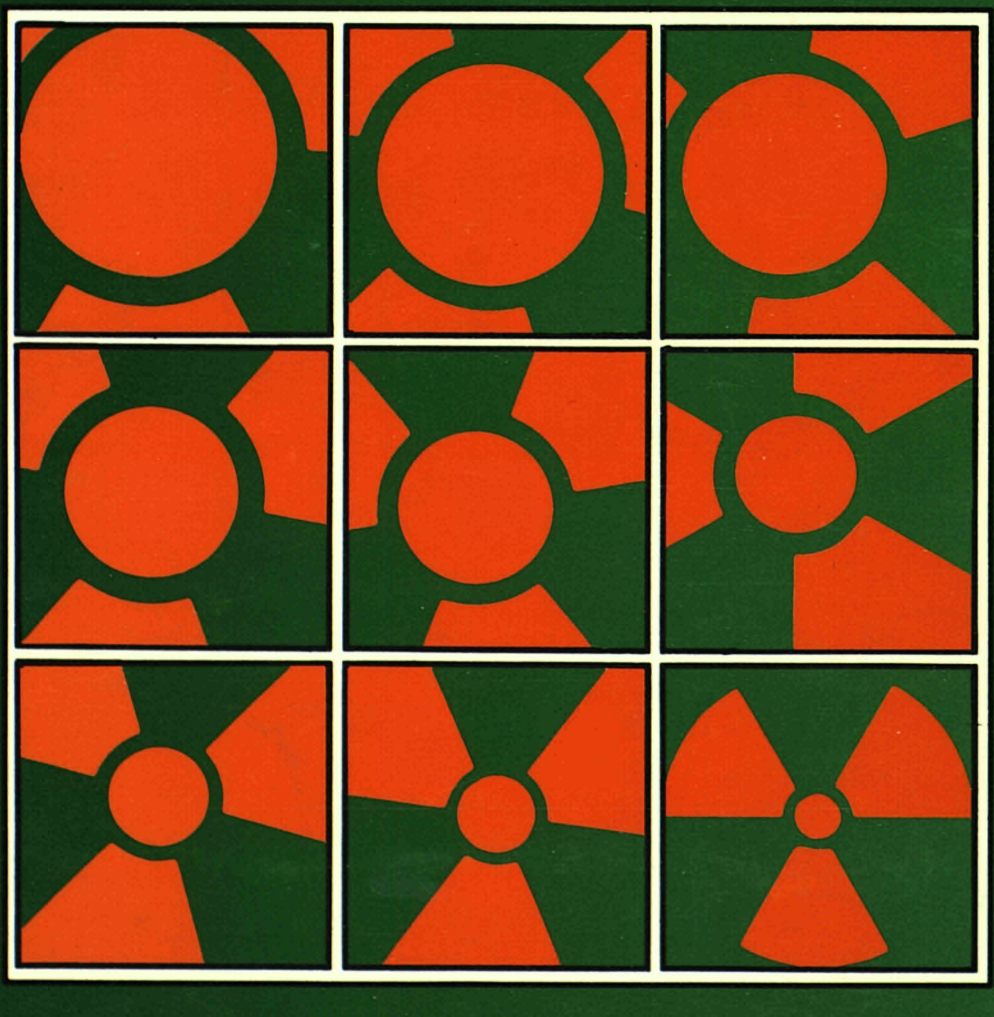
Commission of the European Communities

nuclear science and technology

The Community's research and development programme on radioactive waste management and storage

Shared cost action

Annual progress report 1988
Volume 2



Report

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TASK No 4

Research in support of the development of disposal facilities; shallow burial and geological disposal studies

TASK No. 4 : RESEARCH IN SUPPORT OF THE DEVELOPMENT OF DISPOSAL FACILITIES; SHALLOW LAND BURIAL AND GEOLOGICAL DISPOSAL STUDIES

A. Objective

Evaluation and modelling of the long-term behaviour of the geological barrier

Development of disposal facilities.

B. Research topics dealt with under the 1980-1984 programme

a) Work related to sites and their characterization

- General survey of geological formations and development of measuring techniques with a view to develop large scale in-situ characterization of the geological formations by direct or indirect methods

- Geoprospective studies : development of an operational method for the prospective analysis of the characteristics of geological containment

- Rock mechanics studies.

b) Work related to geological repositories and barriers

- Improvement of the designs and technologies required for the setting up of repositories in geological formations (salt, granite, clay)

- Development of long-lived containers for vitrified waste and of methods for the backfilling sealing of openings in geological repositories.

c) Work on radionuclide migration in the geosphere

- The work mainly comprised integral experiments on migration simulation, laboratory studies concerning the properties of materials from specific sites, hydrogeological investigations, research on natural geological migration systems and the role of micro-organisms, and, finally, the development of calculation tools and the intercomparison of codes regarding transport and geochemistry.

d) Shallow land burial

- Studies dealt with migration phenomena, improvement of barriers and radiological assessments.

C. 1985-1989 programme

The work is mainly a continuation of the research started during the 1980-1984 programme; however, special emphasis is being put on calculation tools and their intercomparison, on investigations attached to specific sites as opposed to laboratory work of general nature, on the role of colloids and complexes in radionuclide migration, on studies of natural analogues, and on the development and assessment of various backfilling materials and concepts.
Coordination is ensured by a structure of projects or working groups :

COSA	:	Comparison of Rock Mechanics Codes for Salt
COMPAS	:	Container Mechanical Performance Assessment
B & S	:	Backfilling and Sealing
MIRAGE	:	Migration of Radionuclides in the Geosphere
COCO	:	Colloids and Complexes
CHEMVAL	:	Geochemical Benchmark for Mirage
NAWG	:	Natural Analogue Working Group

D. Programme implementation

The available information on the contracts signed is listed hereafter.

4.1 RESEARCH RELATING TO SITES AND THEIR CHARACTERIZATION

4.1.A. General survey of geological formations and development of measuring techniques

The 600 m borehole project:
"Development of a surveillance method during dry-drilling of a 600 m deep borehole in salt and performance of geotechnical measurements in the 600 m hole"

Contractor : Netherlands Energy Research Foundation (ECN)
Petten, The Netherlands
Contract No. : FI-1W /0084
Working Period: August 1986 - December 1990
Project Leader: J.R. van Seuren

A. OBJECTIVES AND SCOPE

The experiments performed in the Asse II salt mine in the FRG under the contract with the CEC during the previous programme (1980 - 1984) were carried out in a drilled hole of 30 cm in diameter and 300 m in depth; since then a dry drilling technique was developed for larger diameter holes and greater depths. In this project this technique will be tested by drilling a borehole with a diameter of 60 cm, typical for a disposal hole, and a depth of 600 m.
An alternative for the reconnaissance drilling which takes place before the actual drilling, will be developed with GSF.
The free convergence measurements of the salt as a function of depth of the hole, will be carried out. In a subsequent phase of the programme, non-isothermal pressure measurements will be done at three different levels in the hole. At the bottom of the hole convergence measurements with variable back pressure (isothermal lithostatic measurements) will be performed. Because of the complexity of the total construction the measurements will be done in two boreholes (Figure 1). All these results will be used for the validation of analytical techniques and computer codes.

B. WORK PROGRAMME

B.1 Drilling of the borehole.
B.2 Surveillance method.
B.3.1 Isothermal convergence measurements.
B.3.2 Non-isothermal lithostatic pressure measurements.
B.3.3 Isothermal lithostatic pressure measurement.

C. PROGRESS OF WORK AND OBTAINED RESULTS

Drilling of the borehole

Although not in the contract with the CEC, the drilling of the 600 m holes with a diameter of 60 cm is a vital part of the experiment. This drilling is carried out by GSF under contract with BMFT. Several technical difficulties were encountered during the drilling of the first hole. The depth of the first hole is 300 m with a non-acceptable quality for performing experiments. The drilling of this hole will therefore restart early 1989.

Surveillance method

The system for analysing the gases in the flushing air has been designed and built. A principle drawing of this system is shown in Figure 2.

During drilling of the experimental boreholes the gas components in the flushing air were determined. H_2S has not yet been found in a detectable amount. Hydrocarbons have been determined in the range of up to 40 vpm. The CO_2 content of the flushing air coming from the compressor goin g into the borehole is about 350 vpm, it is increased to 500 vpm when it leaves the borehole. The rock salt horizons which were penetrated during drilling were comparatively dry, therefore no additional water content in the flushing air before it enters the borehole and after it leaves the hole, was found.

In addition to determination of the gases in the flushing air, samples of the drilling fines were taken in order to determine their remaining gas content and their mineralogical composition.

Isothermal convergence measurements

The complete experiment is still in its construction phase. The major auxiliary equipment, which serves the complete experiment has been designed and ordered.

This free convergence will be measured at 100, 250, 400, 500 and 600 m level. The construction for measuring at the first four levels will be identical. The construction for measuring the free convergence at 600 m level is especially designed to measure the end effect of the bottom of the hole on the convergence. This construction is shown in Figure 3 and is momentarily in its testing phase.

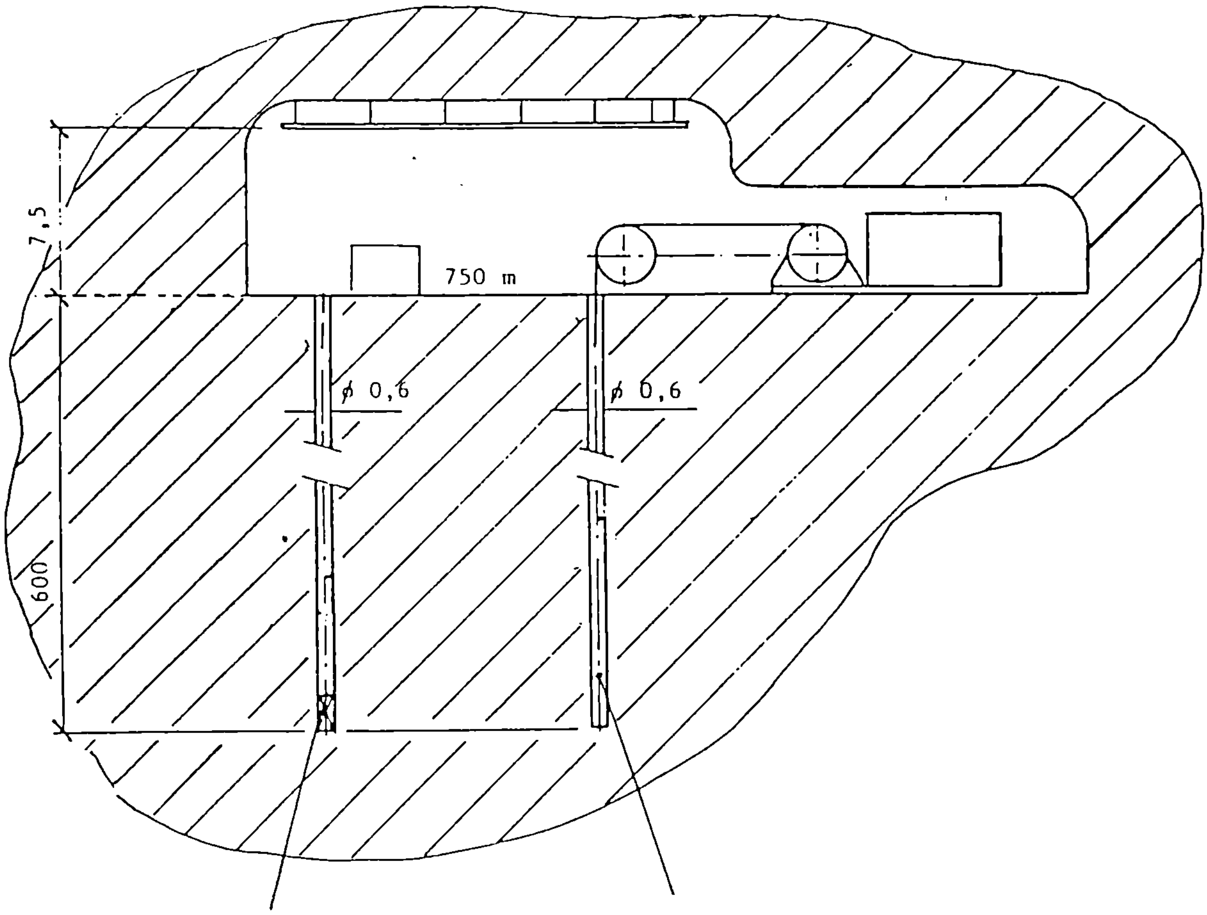
Non-isothermal lithostatic pressure measurements

This experiment will be carried out in the same hole as the free convergence measurement. The deformation on the inserted tube in the hole will be measured.

The tube will be inserted in the hole at different levels and then be heated to close the gap between tube and salt. This experiment is in its design phase.

Isothermal lithostatic pressure measurements

The design of this experiment is based on the working of an inflatable rubber membrane. The construction should be supported by a central column which can withstand the total force of several ton .s, which is exerted on the construction when this membrane is inflated to 200 bar or more. It is the intention to control the process from the 750 m floor in the mine. This construction will operate on the bottom of the first 600 m hole. As a controlling medium a fluid will be used. This experiment is in its design phase. A preliminary design is shown in Figure 4.



DYNAMIC ISOTHERMAL LITHOSTATIC PRESSURE MEASUREMENT

FREE CONVERGENCE MEASUREMENT OVER THE LENGTH OF THE HOLE

NON ISOTHERMAL LITHOSTATIC MEASUREMENTS AT THREE DIFFERENT HEIGHTS

FIG. 1: SKETCH OF THE 600 m BOREHOLE MEASUREMENT SET-UP

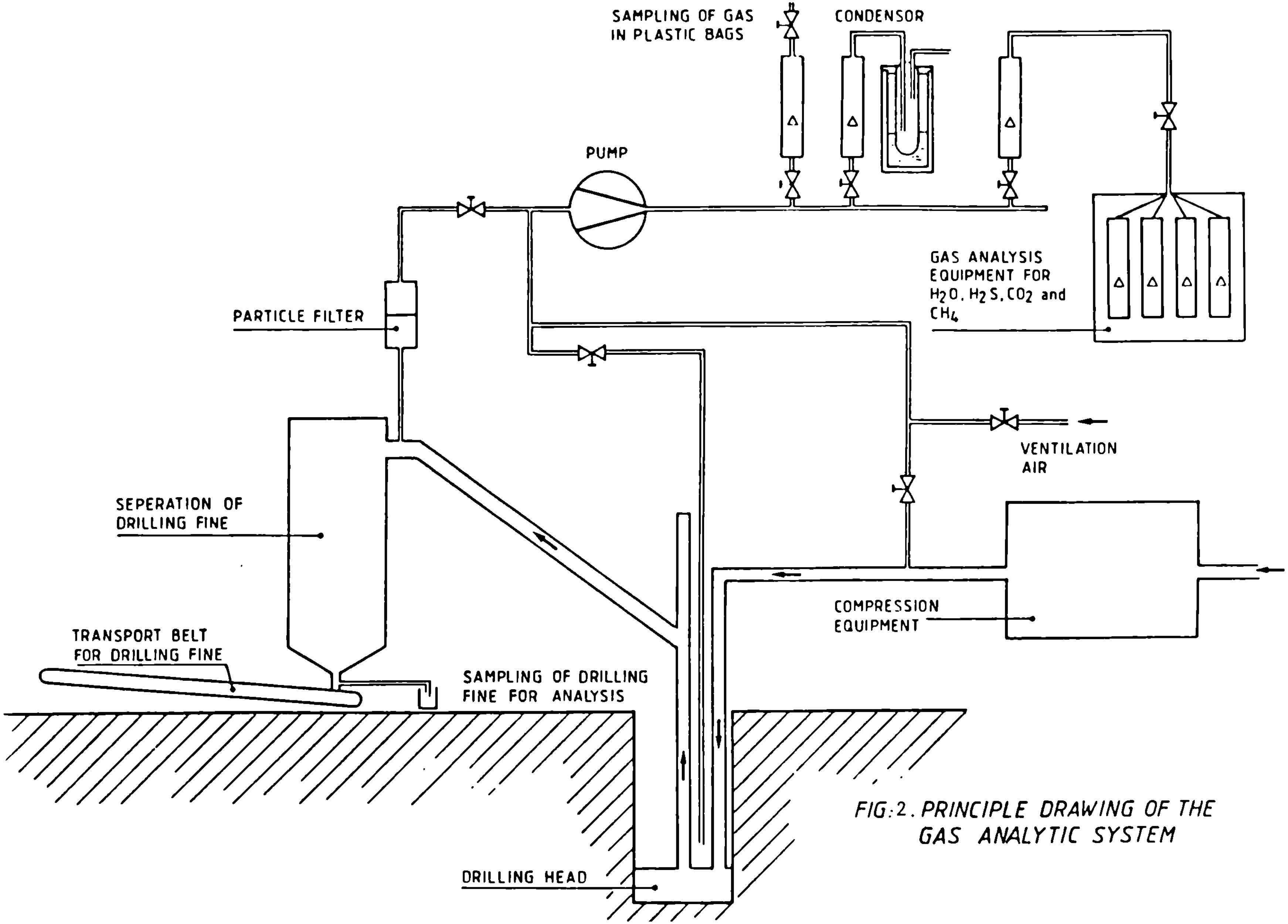


FIG: 2. PRINCIPLE DRAWING OF THE GAS ANALYTIC SYSTEM

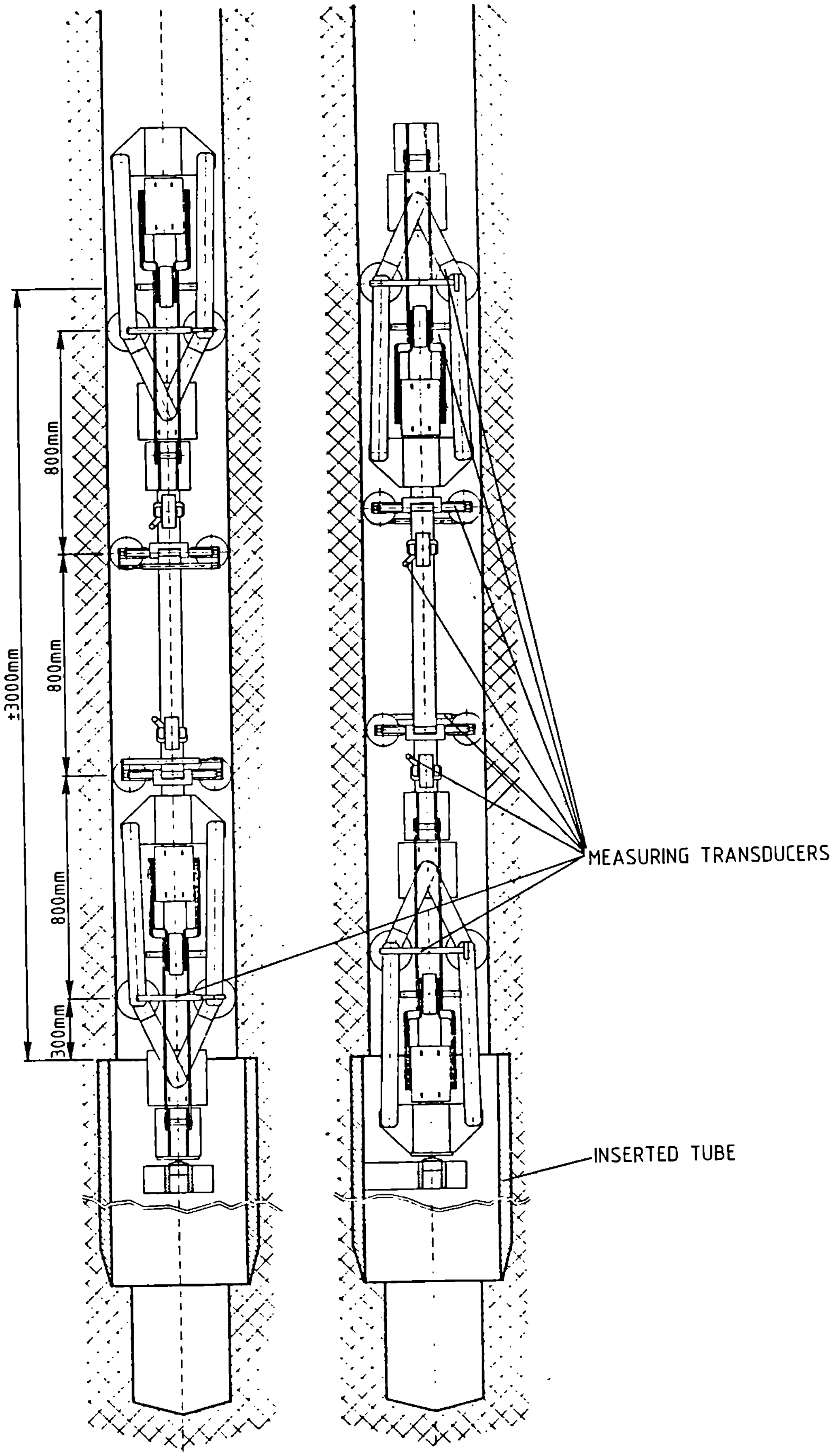


FIGURE 3 MEASUREMENT DEVICE FOR MEASUREMENT OF FREE CONVERSION AT 600m IN THE BOREHOLE

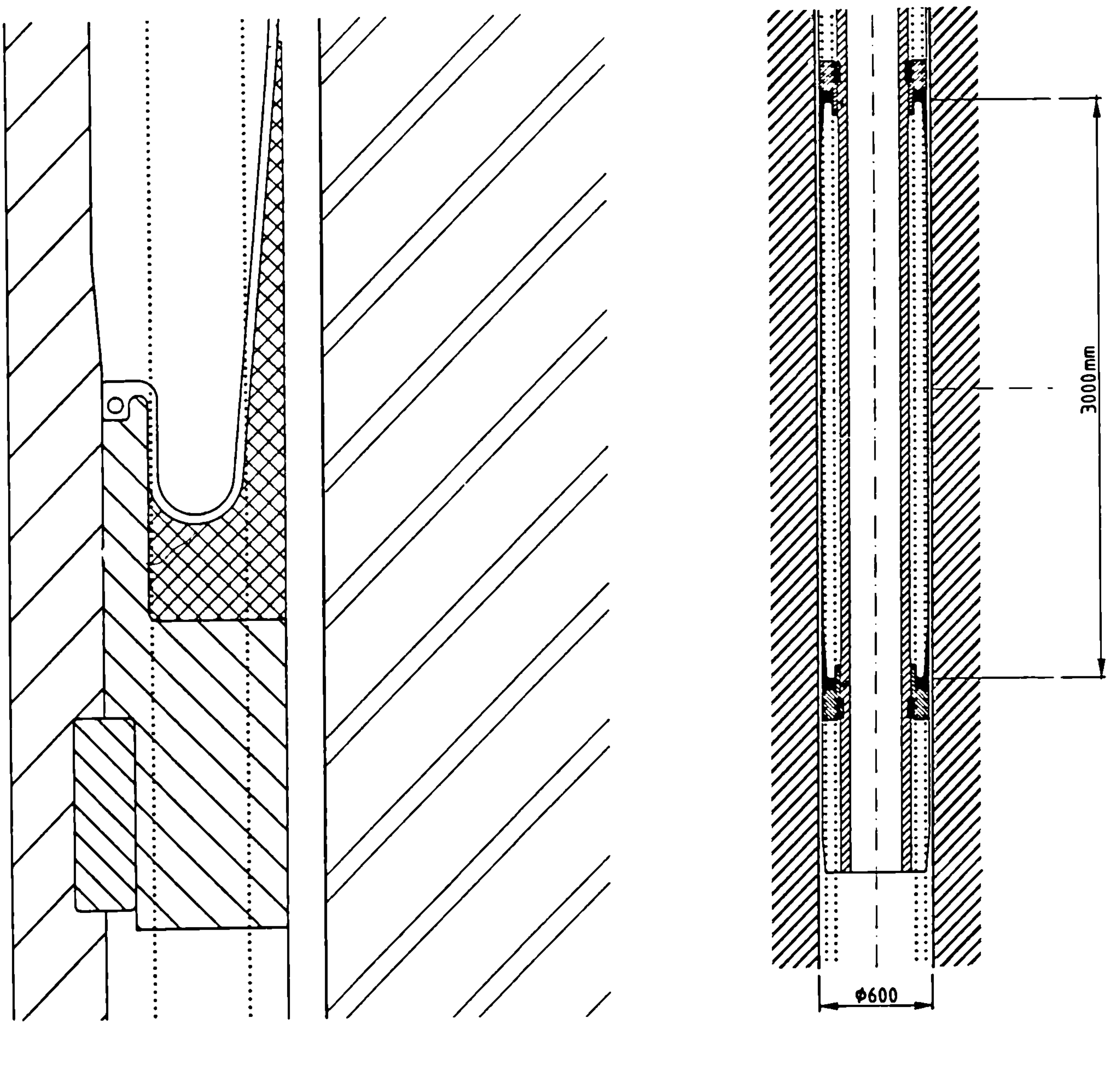


FIG. 4. LITHOGRAFIC PRESSURE MEASUREMENT DEVICE

Faults in clays: their detection and properties

Contractor: BGS, Keyworth, Nottingham, UK

Contract No.: FI1W/0085/UK

Contract Period: October 1986-October 1989

Project Leader: M.A.Brightman

A. Objectives and Scope

Faults occur in most mixed sedimentary environments but their effects on regional groundwater flow patterns are poorly understood. The hydrogeological significance of faulted clay layers is of particular relevance where mudrocks are potential host formations for radioactive waste repositories.

In cooperation with ISMES of Italy two faults through clay layers will be studied (one site in the UK and one in Italy). The project has three objectives :-

1. To develop suitable geophysical techniques to detect water bearing faults in clays. These techniques will aim to differentiate between hydraulically active faults and those which are either sealed or non-water bearing.
2. To measure the hydrogeological properties of faults in sequences of mudrocks and aquifers. This will be achieved by measuring the hydraulic and chemical properties of the fault directly and by measuring the effect of the fault on the underlying aquifers.
3. To define suitable techniques for use in site investigations and methods for assimilating faulted boundaries into flow and transport models in clays and mixed sediments.

B. Work Programme

B.1. Desk study to evaluate a number of potential UK study sites; selection of two preliminary sites.

B.2. Initial geological and geophysical investigations of the preliminary UK sites; selection of the final study site.

B.3. Development of geophysical techniques for fault identification.

B.4. Detailed geophysical survey of the study site.

B.5. Borehole drilling.

B.6. Wireline geophysical logging of the boreholes.

B.7. Hydrogeological testing of the boreholes.

B.8. Synthesis of the results to evaluate the significance of the fault.

C. Progress of work and obtained results

State of advance

A series of exploratory boreholes were drilled to determine the location and geometry of the Down Ampney fault. This was followed by an array of accurately located boreholes drilled perpendicular to the strike of the fault. This array was specifically designed for measuring the geophysical and hydrogeological properties of the fault zone. Detailed geophysical characterisation of the fault is well advanced. Multiple high resolution resistivity measurements have been made in the area straddling the fault. A number of seismic p-wave cross-hole tomograms of the fault zone have been obtained. Hydrogeological testing and groundwater sampling is in progress.

B.5. and B.6. have been completed.

B.3., B.4., and B.7. are in progress.

Progress and results

1. Exploratory borehole drilling (B.5.,B.6.)

A series of exploratory boreholes were drilled at Down Ampney to locate the fault as accurately as possible and to determine its throw and dip. Secondary aims were to obtain resistivity logs to refine the interpretation of resistivity traverses and to obtain core samples for laboratory measurements.

Two fully cored deep boreholes, DA1 and DA2 (Figure 1), were drilled to penetrate the Cornbrash aquifer underlying the clay on either side of the fault. Core samples were taken and wax sealed at regular intervals for a range of laboratory investigations which include pore water squeezing, physical property determinations, geochemical and mineralogical studies. Vertical and horizontal resistivity has been measured on a large number of Oxford Clay specimens and the results show a considerable degree of anisotropy with the vertical resistivity being approximately twice to three times the horizontal resistivity. Detailed geological core logging showed a total displacement of the top of the Cornbrash between the two boreholes of aproximately 47m. A comprehensive wireline logging program was performed in both holes which consisted of natural gamma, neutron, density, focussed resistivity, 16-64" normal resistivity and caliper logs.

Seven shallower cored boreholes, 18-36m deep, numbered DA3 to DA9 in Figure 1, were drilled across the predicted line of the fault. Two boreholes, DA7 and DA8, penetrated the entire fault zone and a full set of cores were recovered. Geological examination of the core from these boreholes showed that the clay for some distance above the fault was disturbed with bedding dipping up to 40°. The fault zone itself consisted of two distinct shear planes approximately 3m apart and detailed lithological examinations estimate the throws across these two planes are approximately 25 and 27 metres. Correlations between the two boreholes indicate the fault plane dips at approximately 70° to the north.

2. The array of measurement boreholes (B.5.,B.6.)

An array of 13 boreholes (Figure 1) was drilled perpendicular to the fault strike specifically for the characterisation of the geophysical and hydrogeological properties of the fault. The main aims of this phase of drilling were to ensure that the boreholes were suitably positioned relative to the fault for the detailed measurements, that the borehole completions were of a very high standard and to obtain core samples for laboratory measurements.

The boreholes were not fully cored, but particular attention was paid to obtaining core from the fault zone and sections to be completed with well screens or piezometers. In total some 230m of core was cut. The boreholes confirmed that the Down Ampney fault is a normal fault striking east-west and downthrowing approximately 50m to the north. The fault plane/zone dips c.70° towards the north. The core from the measurement array boreholes has only been briefly examined in the field, however a few initial observations of the structure of the fault may be made. The upper part of the fault zone at Down Ampney has only been observed in the Oxford Clay where it consists of gently to steeply dipping bedding planes with occassional steeply dipping fracture planes. Different lithological types grade into each other and sedimentary features can be recognised. This disturbed zone appears to be approximately 5-10m thick. The main fault zone is 1-2m thick and is generally very near the bottom of the disturbed zone. In the main fault zone different lithologies are sheared against each other and the boundaries between units are of a variety of dips and directions. In some sections of the main fault zone the mudstone appears to be completely re-worked and totally devoid of sedimentary features, but in others it is heavily fractured, shattered and slickensided shear zones are present. The upper boundary of the fault zone and the edges of the main fault zone are not easy to define absolutely due to the gradational nature of the fault and variations in character between boreholes which are close together. The lower boundary of the fault zone is sharper and generally more easy to define.

Four of the boreholes (DA10, DA11, DA15 and DA18) were completed with 100mm internal diameter casing and well screen and the remaining nine boreholes were completed as 19mm internal diameter piezometers. The boreholes are shown in section in Figure 2.

3. Detailed geophysical characterisation of the fault zone (B.3.,B.4.)

Multiple high resolution resitsivity measurements have been made in an area 50m square which straddles the fault adjacent to the boreholes. Measurements were taken with current flow parallel and perpendicular to the fault. The resultant resistivity values have been contoured and clearly show the fault. More detailed interpretation of these data is progressing.

A series of cross hole seismic p-wave measurements were performed using six pairs of the shallow exploratory boreholes penetrating and adjacent to the fault. This was followed by a more comprehensive cross-hole seismic p-wave survey utilising the array of measurement boreholes. Five p-wave cross-hole tomograms of the fault zone were produced. A new spark source, which has been further developed in-house as part of this project to provide a 1.2 kHz frequency, 1.5m wavelength signal, was used as the p-wave source. Ten hydrophones were used as receivers, and these were supplemented by four surface hydrophones for one section. Each tomogram incorporates at least 900 rays giving excellent ray coverage with typically one metre ray separation. Figure 3 shows the preliminary straight wave tomogram for the section between DA10 and DA18; the fault zone is immediately obvious in this simplest of interpretations. Full processing is currently in hand which will resolve some of the features in the sections more clearly and lead to estimates of the dynamic properties of the fault zone.

4. Hydrogeological testing and groundwater chemistry (B.7.)

The array of measurement boreholes will allow depth profiles of groundwater head, hydraulic conductivity amd groundwater chemistry to be measured in unfaulted rock on both the downthrow and the upthrow sides of the fault and in the fault zone itself. Groundwater head measurements and pulse tests have been performed in all the piezometers and constant rate abstraction tests have been performed in DA11 and DA18. The data from these tests are currently being interpreted and further tests performed. Groundwater samples have been obtained from the Cornbrash aquifer from DA11 and DA18 and from the White Limestone from DA10. Pore water samples of approximately 20cm^3 have been squeezed from 16 core samples taken from the exploratory boreholes DA1 and DA2.

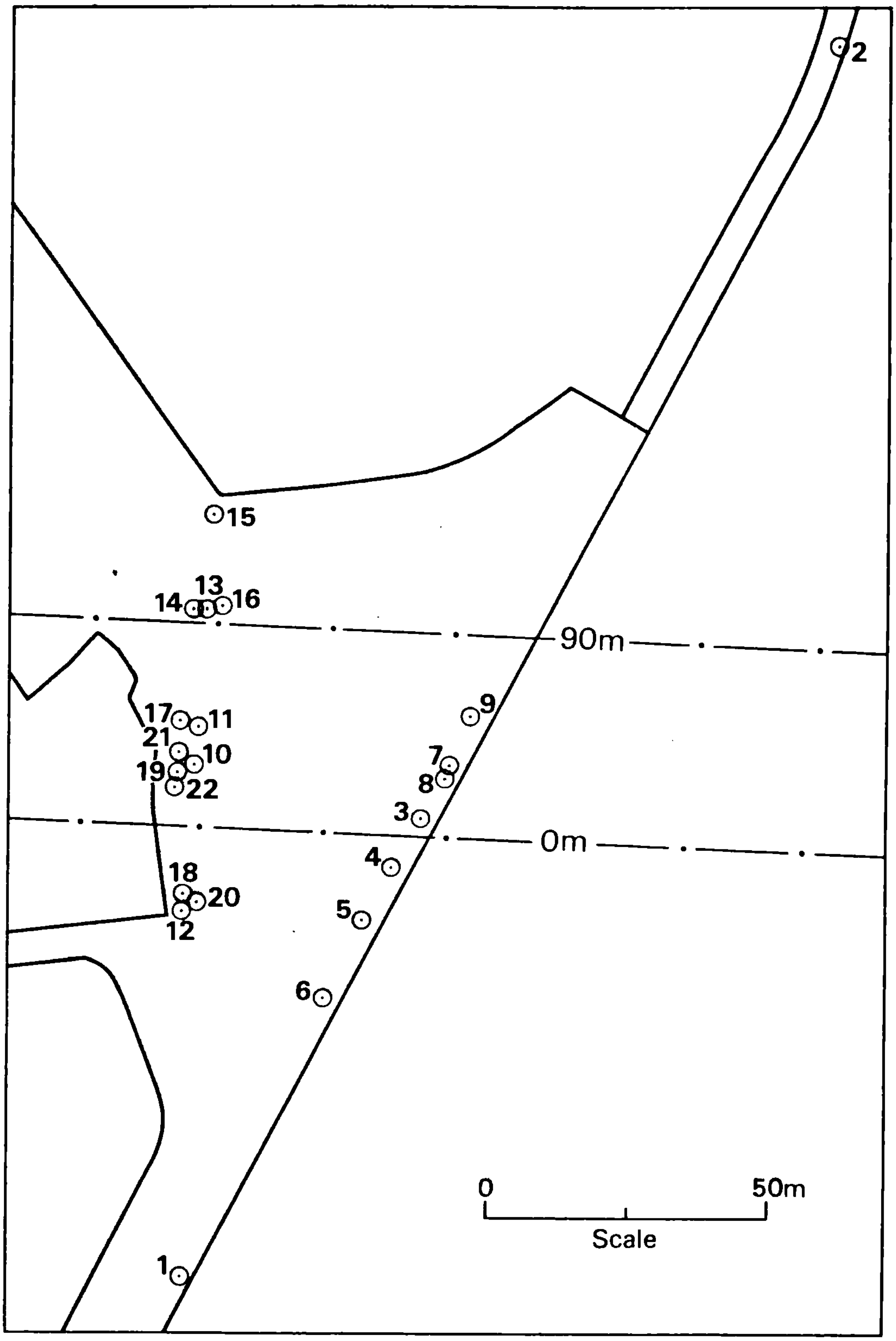


Figure 1. Borehole location plan for the Down Ampney Research Site showing contours on the fault plane at 0 and 90m below ground level.

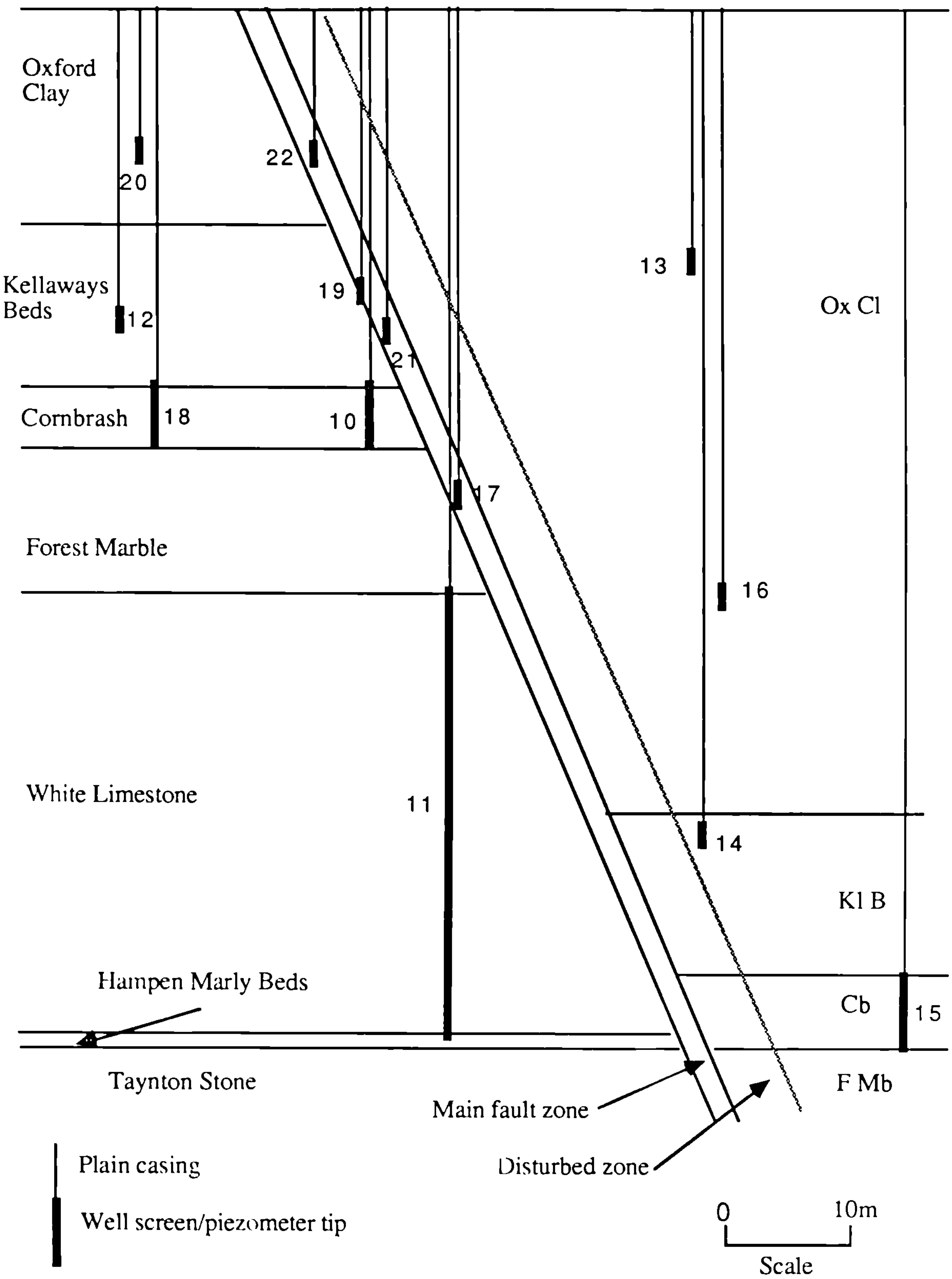


Figure 2 Section through the measurement array of boreholes at Down Ampney

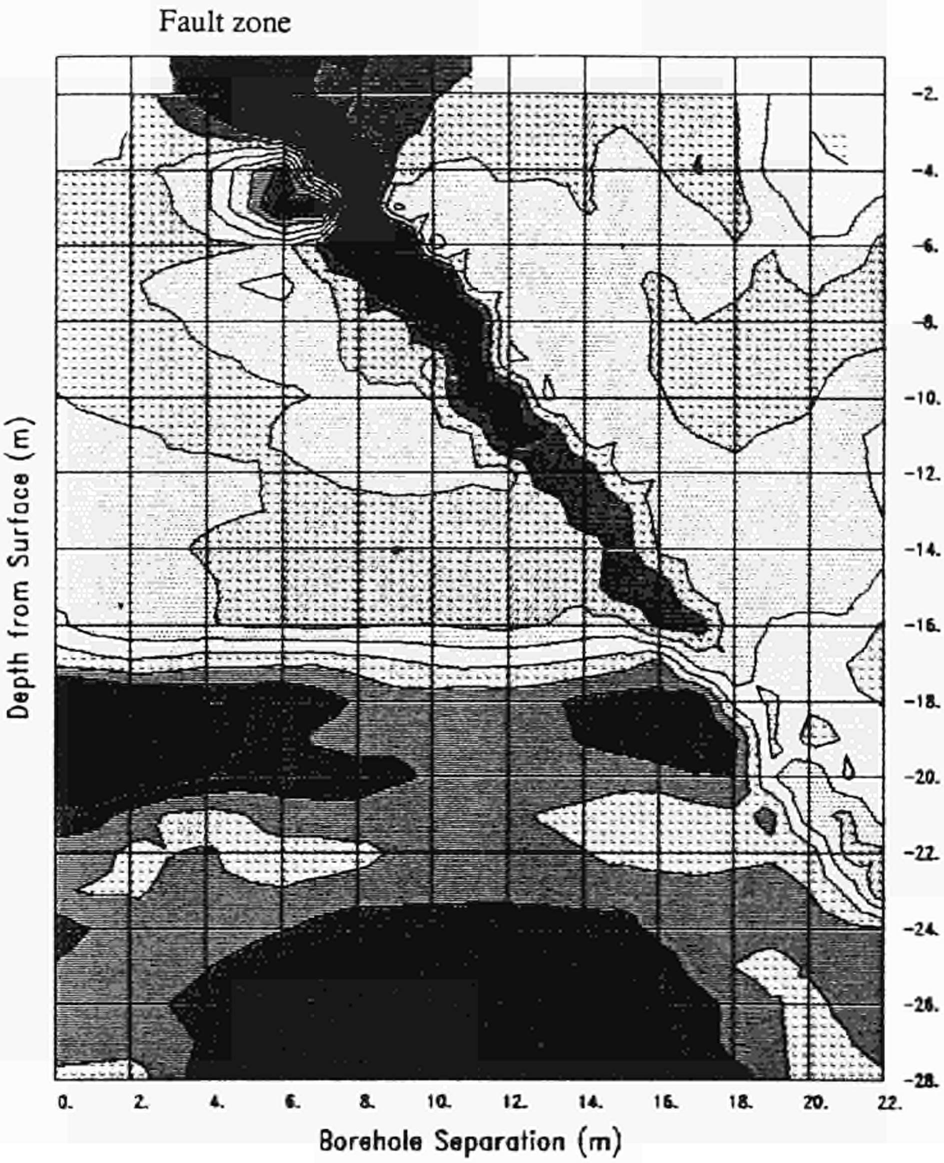


Figure 3. Seismic p-wave cross-hole tomogram between boreholes DA10 and DA18.

METHODOLOGY FOR APPLICATION OF ELECTRIC AND ELECTROMAGNETIC BOREHOLE TECHNIQUES FOR DETAILED EXPLORATION OF FRACTURED ROCKS

Contractor : BRGM, Geophysics department, Orléans, France.
Contract n° : FI1W/0086/F.
Duration of contract : July 1986 - June 1989.
Project leaders : G. POTTECHER, P. VALLA.

A. OBJECTIVES AND SCOPE

The aim of the research work is first to complete the technical developments of three borehole geophysical methods for which prototypes have been built, and second to develop a methodology for these tools applied to detailed investigation of fractures.

These tools are :

- ELIAS, an electrical imaging technique to investigate the borehole wall and determine the depth, strike and dip of conductive or resistive fractures,
- ROMULUS-ERIC, a set of induction transmitter-receiver probes to point out conductive fractures and measure their conductance with a one to a ten meters radius of investigation,
- ARLETT, a three axis induction receiver used together with a surface elecromagnetic transmitter, to help assess the geometry of the more conductive fractures in a ten to one hundred meters range.

The first and third systems are still in the technical development stage while the second is now operational. Numerical modelling of the methods is needed to fully assess their capabilities.

B. WORK PROGRAMME

1. Technical and theoretical development of electric and electromagnetic probes.
1.1. ELIAS
1.1.1. Final step of prototype development
1.1.2. Design of data acquisition and processing software
1.2. ROMULUS-ERIC
1.2.1. Design of modelling software for thin sheet conductors
1.2.2. Set up of a catalog of theoretical response curves
1.2.3. Study of complex geometry of thin conductors
1.3. ARLETT
1.3.1. Study of improvements to be made in the probe design
1.3.2. Development of a new prototype
1.3.3. Numerical modelling

2. Field tests and methodological studies
2.1. Tehnical field tests of ELIAS and ARLETT
2.2. Methodology for detailed exploration of fractured rocks
2.2.1. Data acquisition on available test sites
2.2.2. Data processing and interpretation
2.2.3. Analysis of results and methodology assessment

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Technical developments done this year include the completion of the imaging probe ELIAS and of the analogic modules in the 3-axis electromagnetic probe ARLETT.

Methodological work on the ROMULUS and ERIC systems has been completed with model computations, complementary data acquisition and throughout interpretation work.

Operational capability of ELIAS is proven. Data is now being interpreted.

Progress and results

1.1, 2.1, 2.2. ELIAS probe

The prototype probe including an orientation module and 16 measuring pads has been constructed.

A data acquisition system has been designed for a field microcomputer, enabling image display and magnetic data storage in real time.

Other software developments include a basic processing module (image normalization and orientation) and a program for enhanced display and fracture picking.

The complete system has been field tested in granite and proved to be successful in fracture delineation. A large set of data has been acquired for methodological purposes (Figure 1 : ELIAS images).

1.2, 2.2. ROMULUS and ERIC probes

An extensive model catalogue has been computed and synthetized for efficient use. It contains the following conductive thin structures in various attitudes : intersected disc, missed square plate, missed half plane, infinitesimally small plate.

Data has been acquired at 3 sites in fractured metamorphic schists. The interpretation process based on the model collection was used to exploit them. It could be shown that valuable information can be derived from electromagnetic measurements regarding fracture exploration in this kind of rock.

1.3.2, 1.3.3. ARLETT system

The monoaxial ARLETT probe was modified in order to increase the maximum operating frequency from 900 Hz to 3000 Hz and to make it easier of use. These improvements will be kept for the triaxial probe.

The set of 3 sensors for ARLETT have been designed and constructed. The crosstalk level meets the requirements. Additionnal developments include specific amplifiers and filters and an A/D converter. These last circuits are being tested.

A rich model catalogue has been established, involving conductive square plates and half planes in various attitudes for all field components. An interpretation procedure based on these data could be established.

2.2. Complementary methodological work

A survey has been carried out with an electrical crosshole array in granitic rock. It enabled to determine fractures connecting both boreholes and yield results in good agreement with crosshole hydraulic tests. These features make it a usefull complement to electromagnetic methods.

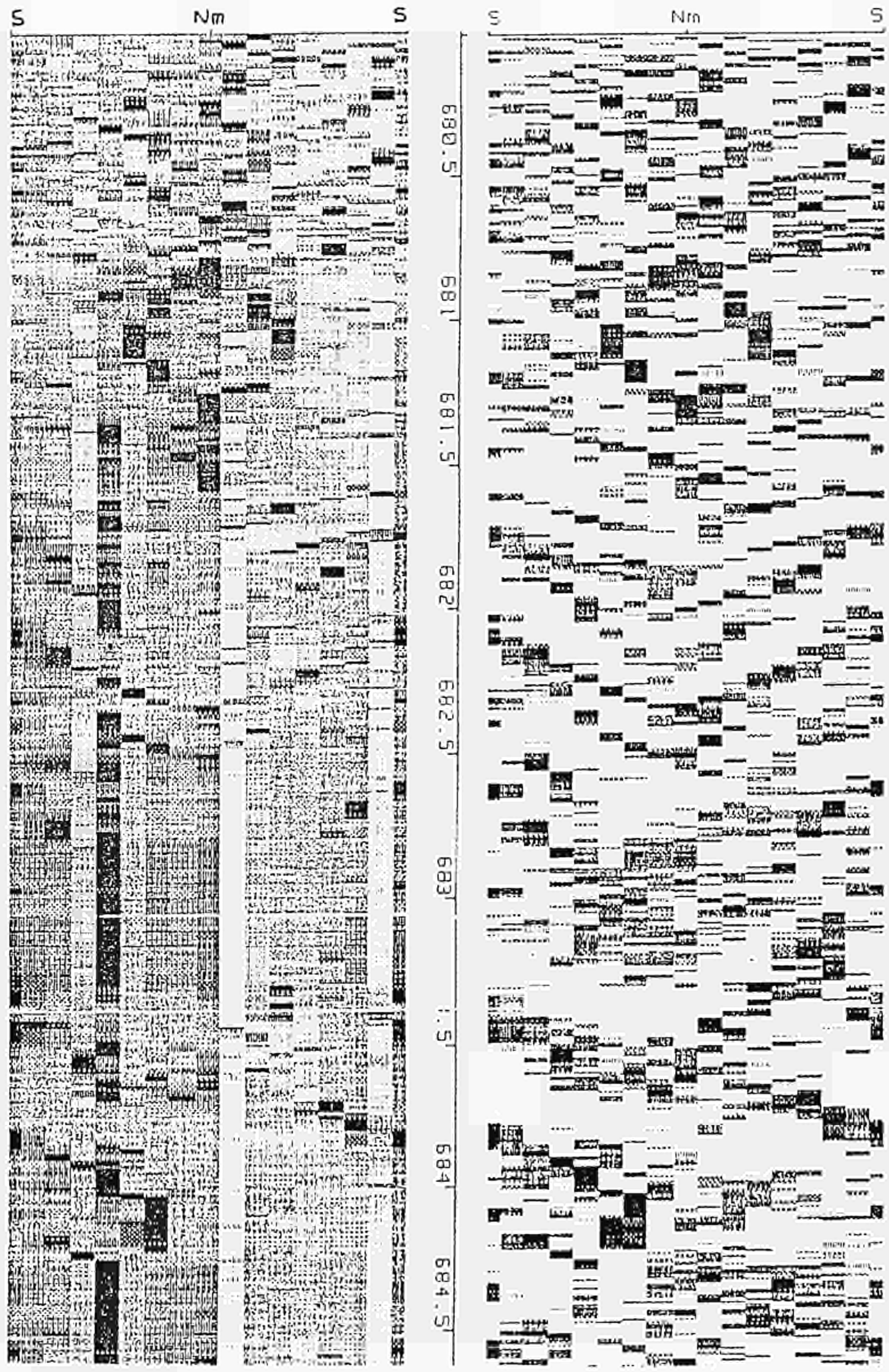


Figure 1 : Image obtained with the borehole imaging probe ELIAS. Fractures appear as sinusoidal curves.

Left : raw data.
Right : processing attempt.

DEVELOPMENT OF A SELF-CONTAINED DRILL-HOLE CHROMATOGRAPHIC PROBE

Contractor	:	CEA - CEN CADARACHE F 13108 - Saint Paul Lez Durance
Contract N°	:	FI1W/0087
Working period	:	1 Nov. 1986 - 1 Nov. 1989
Project Leader	:	J.M. Vinson

A. OBJECTIVES AND SCOPES

The study of the transuranian nuclide migration from radioactive waste storage places is based on the knowledge of the natural environment and, in particular, of the chemical composition of the water : the transfer vector.

In addition to major elements, the water also contains trace elements which play a prominent part not only on the general equilibrium, but also on the radionuclide migration possibility.

This is in particular the case of the Lanthanides, present in the granitic and argillaceous environments, whose role of sorption competitors with respect to the Actinides has been exhibited throughout numerous experiments.

The scope of this contract is to manufacture and operate a chromatographic probe to be used in a deep drill-hole, so designed as to acquire a representative sample under conditions of equilibrium of the natural environment, to preconcentrate it by elimination of the saline content and to store it for ulterior analysis at the surface.

B. WORK PROGRAM

B.1 Process Development

B.1.1 Principle laying-down and delineation. In-laboratory model

B.1.2 Definition of final probe design after study on model. Realization and adjustment on CADARACHE site

B.2 Probe Qualification in the Deep Drill-hole of AURIAT

B.3 Application to 3 reference sites

C. WORK PROGRESS AND RESULTS OBTAINED

Summary

Samples of Auriat water are currently being studied to test the feasibility of chemical separation and analytic sensitivity.

Analytic studies are continuing in all fields which may improve and diversify information which can be obtained from soda.

The main activity in 1988 was the tool manufacturing study, which has almost been completed. The first on-site assembly tests have taken place. Auxiliary equipment necessary for the smooth operation of the tests and the experiment were defined. They are currently being manufactured.

Progress and results

. Laboratory model

The above mentioned study has shown that after separation the lanthanides and some transition elements can be analyzed at concentrations of about 10^{-10} ion.g/liter.

A study on water taken from the AURIAT well is currently under way in order to build up a library of well-defined peaks for conditions applicable to natural water rather than to standard specimens. Moreover, it must determined if the analytic sensitivity is adequate in different environments, depending on the real concentration of elements ; in the respect conventional sampling does not give the best guarantees. Other investigations will be necessary, especially during early site experiments.

Therefore, analytical work is continuing in several directions :

- Quantitative evaluation of lanthanides and transition elements in the AURIAT well.

  Based on samples, elements are column fixed and the saline charge is eliminated on-line in order to improve neutronic activation analysis of fixed elements.

- Evaluation of possibilities provided by the "thermal lens" method, which could improve the performance of the current technique by at least an order of magnitude.

- Spectrofluorometry sensitivity study for some lanthanides, with or without prior separation.

- Study of the possibilities provided for analysis of anions, especially those similar to TcO_4^- technetates.

- Evaluation of other types of cation supports.

In addition, the current development of some techniques such as "ICPMS" makes it possible to consider much elementary analysis better performances. They could be even more advantageous in that removal of the saline charge and/or divalent elements could add a certain amount of qualitative information for very low concentration conditions, since the main advantage of the probe is to sample elements volumetrically on specific supports (cation or anion columns or ultrafilters for colloid analysis columns) with a minimum of disturbance.

2. MANUFACTURE AND DEVELOPMENT OF THE EQUIPMENT

2.1 Development

In parallel with the FORALAB program, some modifications have been made both to function controls and to component testing.

2.1.1 Upper valve (Id. 20)

The valve rating on the tank level has been adjusted such that it is possible to keep all circuits at a pressure slightly above that in the borehole, so that the acid pump cannot discharge freely alone.

2.1.2 Electro-valves

The solenoid power supply system has been modified in order to restrict the electrical power supply required for opening. Thermal release remains at a reasonable level thus avoiding the risk of a power cut during long period operation.

2.1.3 Machining

In order to limit the possibility of solenoid-valves being blocked by stray chips, machining procedures have had to be modified : especially for drilling.

2.1.4 Component tests

During operation of the "FORALAB" probe and during workshop tests of elements of the "chromato" probe, it became apparent that the operation of some components - especially solenoid valves - did not comply with nominal characteristics supplied by the manufacturer.

In addition, the use of a poor thermal conductor under the special geometry conditions over long periods and inside a stainless steel duct is not favorable for the long life of these characteristics.

It is therefore necessary that all components be tested individually, simulating their real conditions of use.

Some of the initially selected components had to be replaced.

2.1.5 Logistics

Some systems such as the "FORALAB" and "Chromato" probes differ from conventional geophysical probes due to their length and weight (greater than 100 kg). Consequently, many modifications were made to the entire drill hole support system. The cable support, pulleys and brake support plates have been changed.

2.2 Appendices

In preparation for on-site laboratory experiments, essential equipment has been defined and is currently being manufactured.

2.2.1 Assembly rig and surface tests

During tests the equipment must be in its complete operating condition, and vertical. Due to its length, a support must be available to position the stages side by side and connected by hydraulic and electric connections. It must be possible for this support to be horizontal for assembly and vertical for operation.

2.2.2 Pressurizing rig

To simulate drilling conditions and to carry out sealing and smooth operation tests on the surface, a rig is necessary to pressurize the probe and the acid tank piston.

This rig contains one 1 liter tank, an external chromato pump make up supply, a nitrogen supply for pressurising, a water level indicator, a purge, a pressure sensor and an external circuit, and a CO_2 supply to purge air before adding water.

Before each experiment, it will be necessary to test that circuits and valves are tight. Filters shall be changed and the discharge checked.

2.2.3 Electrical test rig

Before putting the tool down the bore-hole, all active and passive electrical functions must be tested : solenoid valves, wate pumps and detectors, electrical and electronic continuity in the control and power supply racks, at the cable support and each stage of the equipment.

An exhaustive list of systematic tests to be carried out is under preparation.

<u>List of publications</u>

International symposium

"Hydrogeology and safety of radioactive and Industrial Hazardous waste disposal".

ORLEANS - FRANCE 7-9 June 1988

The Detection and Measurement of Faults in Clay

Contractor: University of Exeter, Exeter, UK.
Contract No: FI1W-0088-UK
Working Period: September 1986 - August 1989
Project Leader: Dr. E.M. Durrance

A. Objectives and Scope

If faults occur in the rock mass surrounding a nuclear waste repository, there is a risk that the return of hazardous radionuclides to the biosphere will take place by migration along these zones of higher permeability. However, the detection and characterisation of faults is difficult, especially in soft rocks such as clay, and little development of techniques has taken place. The objective of this programme is to develop techniques that will be suitable for routine use in both the preliminary and detailed stages of site investigation. The approach used is based on the observation that faults act as zones of preferential migration in the natural degassing of the Earth. Soil gas exploration methods are applied to detect zones of anomalous gas geochemistry. The procedure followed is based upon samples obtained from a depth of about 0.5m along a series of traverse lines. Once a fault has been located, spiking of the high permeability zone from a borehole drilled to intersect the fault plane, will take place with specific gases of different compositions. The ground will then be resurveyed to determine the migration characteristics of the gas within the fault. Test sites in the UK and Italy are to be investigated in co-operation with BGS (Keyworth) and ISMES (Rome). BGS and ISMES are responsible for site selection and the drilling programme, but some trials will be conducted at sites near Exeter.

B. Work Programme

B.1. Equipment development.
B.2. Site selection.
B.3. Soil gas geochemistry.
B.3.1. Reconnaissance soil gas surveys measuring ^{4}He, ^{220}Rn, ^{222}Rn, O_2, CO_2 and some organic gases.
B.3.2. Detailed soil gas surveys of anomalous zones identified in the reconnaissance surveys.
B.3.3. Spiking of vertical boreholes and resurvey of soil gases.
B.3.4. Spiking of inclined boreholes and resurvey of soil gases.
B.4. Modelling and interpretation of results.

C. PROGRESS OF WORK AND OBTAINED RESULTS

Summary

The running of the ^{4}He mass spectrometer in the mobile laboratory powered by portable generators has been improved. A gas chromatograph for additional soil gas analyses has been evaluated. At a selected clay site (Westrow) a major fault has been detected. A two week visit to the ISMES site at Orte (Italy) in October 1988 was necessarily cancelled due to unavoidable access problems and exceptionally heavy rain. A short successful reconnaissance visit was made to the Narni site in December. Equipment and supplies for initial gas injection tests at Down Ampney are complete.

Equipment development (B.1.)

Electrical smoothing apparatus has been purchased for use with the ^{4}He mass spectrometer installed in the mobile laboratory. This reduces interference on the helium response trace when the detector is run from mobile generators. Use of a local mains source is still preferred, however there may be scope for further improvement to ensure that low background He values are not obscured by noise.

At present, for the detection of CH_4 a gas chromatograph with a thermoconductivity detector is being tested using argon as carrier gas. The CH_4 detection limit is currently 30ppm, and CO_2 can be analysed at the same time. This provides a back-up to the portable analyser and should prove valuable where water saturation of the ground allows only a little gas to be withdrawn without water being sucked into the sampling equipment.

Soil gas geochemistry (B.3.2.)

Westrow, the site selected for independent testing of the soil gas technique, is on Oxford Clay in north Dorset (U.K.). Two north-south striking faults, the Goat Hill - Westrow and the Crouch Hill Faults are believed to cut the area. Soil gas signatures from the current work correspond to the projected positions of the faults as indicated by other displaced strata on the B.G.S. (British Geological Survey) Shaftesbury map. The presence of marshy zones and natural ponds provide supporting evidence for the fault locations. Explanation of some anomalies in soil gas distribution is not yet clear, though some may correspond to shear faults associated with strike-slip movement on the Goat Hill - Westrow Fault. Further work is planned.

A site assessment visit to the ISMES (Rome) faulted clay site at Narni (Italy) was made in December 1988. Sampling was at 6.25m intervals from two closely spaced traverses. Both soil gas CO_2 and He increased from near atmospheric levels (0.035% and 5220ppb) to 1.6% and 5446ppb respectively over the anticipated position of the fault. Consistent changes in CO_2 over 3 to 5 sample points suggest the potential for a reconnaissance survey with wider spacing, covering more ground, followed by detailed measurements in selected areas. No modification of results occurred over ploughed ground where the standard 0.5m sampling depth was increased to 0.75m. Future work in 1989 will increase the number of background data values away from the fault to confirm that the high values found do not result from some other feature. The Orte (Italy) site is thought to offer less potential for soil gas work due to complications related to vegetation cover.

Spiking of Boreholes (B.3.3.and B.3.4.)

Information has been obtained from B.G.S. concerning boreholes drilled at the Down Ampney (U.K.) faulted clay site. All are vertical, the north dip of the fault being sufficient to obviate the need for inclined holes. One borehole (No.10), with a screened section in the permeable Cornbrash, 3 to 5m distant from the fault plane, is the closest intersection to the fault and will be used for gas injection experiments. A packer with a 1 metre long rubber sleeve is available to seal the borehole above the test section in the permeable horizon. Concentration of CO_2, passing up through the fault, will be measured on the surface. Later experiments will use helium as well. Drilling of the boreholes through the concrete runways may give problems with measurement of induced soil gases due to the capping effect. Soil probes will be placed in adjacent ground as near as possible to the fault plane. Work will commence in late February 1989, depending on availability of boreholes.

FAULTS IN CLAYS: THEIR DETECTION AND PROPERTIES

Contractor: ISMES S.p.A., Bergamo, Italy
Contract N°: FI1W/0103
Duration of contract: November 1986 - May 1990
Period covered: January 1988 - December 1988
Project leader: Ferruccio Gera

A. OBJECTIVES AND SCOPE

The contract has to be coordinated with a companion contract to BGS to perform similar work in UK clays.

Faults and fractures are known to intersect argillaceous formations that might be considered suitable as host rocks for a radioactive waste repository. In some cases these structural discontinuities have been observed to enhance the hydraulic conductivity of mudrocks. Consequently any argillaceous formation considered for the location of a waste repository should be characterized thoroughly from the points of view of occurrence of fractures and their hydraulic significance.

Available geophysical techniques with the capability of revealing the existence of structural discontinuities in mudrocks, particularly water-bearing ones, could be developed, then site investigation studies would rely more on geophysics and less on drilling with significant advantages from different points of view.

The objectives of the contract are:

- to develop suitable geophysical techniques to detect water-bearing faults in mudrocks. The main intended application is for surface investigations, but possible application in tunnels and boreholes should be considered;
- to carry out hydraulic testing across deep faults to determine their hydraulic conductivity and the hydraulic conditions of any measurable water flow;
- to define suitable techniques for use in site investigation campaigns.

B. WORK PROGRAMME

The project consists of the following activities:

1) a survey within the country to identify sites where faults intersect mudrocks;
2) chosing the most suitable sites for carrying out field work;
3) drilling at least two boreholes in such a way that the fault plane will be intersected at a suitable depth;
4) performing geophysical investigations of the fault zone both from the surface and down hole;
5) hydraulic testing of the fault zone (responsibility of BGS);
6) geophysical logging of the boreholes;
7) geotechnical measurements of samples obtained from the cores;
8) hydraulic modeling of groundwater flow in the fault zone.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

3) An extensive characterization of the clay formation preceeded the choice of 3 potential drillhole locations; it consisted of: a) a geophysical electrical reconnaissance survey; b) a topographic survey. Following the characterization, c) a first borehole was drilled to a depth of about 90 m with core recovery for stratigraphic reconnaissance; d) in a second borehole, 5 m apart (about the same depth), 22 undisturbed samples were collected.
4) The following geophysical surveys have been carried out at the Orte site: a) reflection seismic; b) single-hole (sonic, electrical, natural gamma) logs; c) cross-hole.
5) Hydraulic testing of the fault zone (under responsability of BGS) was not performed because of the constant characteristics of the clay body and because of logistic problems of our English colleagues.
6) See point 4).
7) Geotechnical measurements of samples obtained from the cores started some weeks ago.

Progress and results

3.a GEOPHYSICAL ELECTRICAL RECONNAISSANCE SURVEY

It consisted of resistivity measurements by means of horizontal and vertical logs along a line approximately orthogonal to the suspected fault plane (Figure 1). Obtained results (Figure 2) show limited variations in the resistivity values indicating a sedimentary sequence with constant electrical resistivity (the clay formation) and a thickness of at least 300 meters. From the geoelectrical survey no useful indication about the existence of the suspected fault plane has been obtained.

3.b TOPOGRAPHIC SURVEY

A planoaltimetric survey, covering both the quarry and the immediate surrounding area, has been carried out in February 1988 to locate precisely 3 points for drilling. The objective was to drill through the fault plane at a depth between 55 and 80 meters. The probable position of the fault plane at depth has been estimated by extrapolating the fault plane recognized and measured on the quarry face.

3.c FIRST BOREHOLE

The first borehole (OS1) was drilled with a simple core bit without using either mud or water up to -18.30 m; from that point to the bottom a double corer and water have been used, and the final diameter, at depth 90.50 m, was 146 mm. Continuous coring allowed to investigate the characteristics of the clay formation. The recognizable sedimentary features were: some gravel levels in the first 18 m and then only some very thin sand layers (1-5 cm at the most), very few fossils and some metallic nodules (mainly pyrite). Fractures have been encountered only in very few occasions: at about -32/-33 m, -55.80 and -60/-61; this last discontinuity could correspond to the foreseen depth of the fault plane for OS1, even if somewhat shallower. However, notwithstanding the slickensided texture of several fracture surfaces, no sign of present or past water seepage was noticed and the discontinuities seemed to be watertight.

3.d SECOND BOREHOLE

The second borehole (OS2) was drilled with a rock bit: coring was limited to collecting 22 undisturbed or semi-disturbed samples concentrating the sampling around the depth where the intersection with the fault plane was expected.

4.a REFLECTION SEISMIC SURVEY

Four high-resolution seismic lines have been carried out (Figure 1). None of the investigated lines seems to indicate dislocations, even if acquisition parameters were set up to detect faults with a throw as small as of 1.5 - 2 m. In particular the line ORTE 1 shows shallow reflections with a dominant frequency of about 200 Hz, but no evidence of a fault. In some sections of the prospection lines (particularly ORTE 2 and ORTE 4) there are seismic signals that may be interpreted as tectonic disturbances, but they are probably due to a local low seismic coverage. On the line ORTE 1 some different energy sources have been tested; they were: betsy, hammer, air-gun and sparker. Field seismograms and their correspondent power spectra show that in this area betsy is the most valid source.

4.b SINGLE-HOLE LOGS

Sonic logs have been carried out, before casing, in both OS1 and OS2 boreholes. They have been performed by means of a sonic probe designed and built by ISMES: it has a diameter of 45 mm and the measure base is 1 m long. Measurements have been carried out in two ways. In the first run, P waves velocities at depth intervals of 0.5 m have been measured. In the second run, a continuous log with measures every 2.5 cm has been recorded. Velocity values range between 1800 and 2100 m/s; only at a depth between 45 and 60 m small velocity variations are present, but variable density representations show substantially uniform propagation characteristics (Figure 3).

Electrical logs (spontaneous potential and 16 - 64" normal resistivity) have been also performed in borehole OS1. Both resistivity and SP curves are extremely flat, indicating the absence of relevant lithological variations; resistivity values are generally low due to the presence of saline fluids.

Finally, natural gamma logs have been performed in both boreholes, OS1 and OS2, cased with a plastic tube. Recorded values are not relevant: the only appreciable variation is a lower gamma emission at a depth between 46 and 62 m.

4.c CROSS-HOLE

A cross-hole survey of P and S waves propagation has been carried out between the two boreholes; measures have been taken at depth intervals of 1 m. The P wave velocity curve is similar to those recorded with sonic logs (Figure 3). The S wave velocity diagram shows constant values (about 500 m/s) up to the depth of 60 - 65 m. Below this depth there is a gradual decrease of velocity and the seismic signals received show a low energy level. Because no significant variations were pointed out by single-hole logs performed in the uncased borehole, we are inclined to attribute the decrease of energy and velocity to the poor quality of the casing grouting.

7. GEOTECHNICAL MEASUREMENTS ON SAMPLES

An extensive program of laboratory testing has started on samples collected in borehole OS2. In that borehole 22 undisturbed samples at depths ranging between 31.00 and 66.80 meters have been taken and some of them have been sent to BGS for water squeezing and chemical analyses (Table 1). The geotechnical determinations planned for ISMES's samples are addressed to:

- classification (granulometry, density, Atterberg limits, calcimetry);
- consolidation history of the formation (edo Ko);
- P and S waves velocity (for different frequencies);
- permeability (mainly on samples near the suspected fault plane).

From very preliminary results, in agreement with literature data, the clay seems to be strongly overconsolidated (OCR >5); for that reason some tests (permeability and oedometric) need to be executed in high pressure cells; this will imply a much longer time than previously foreseen.

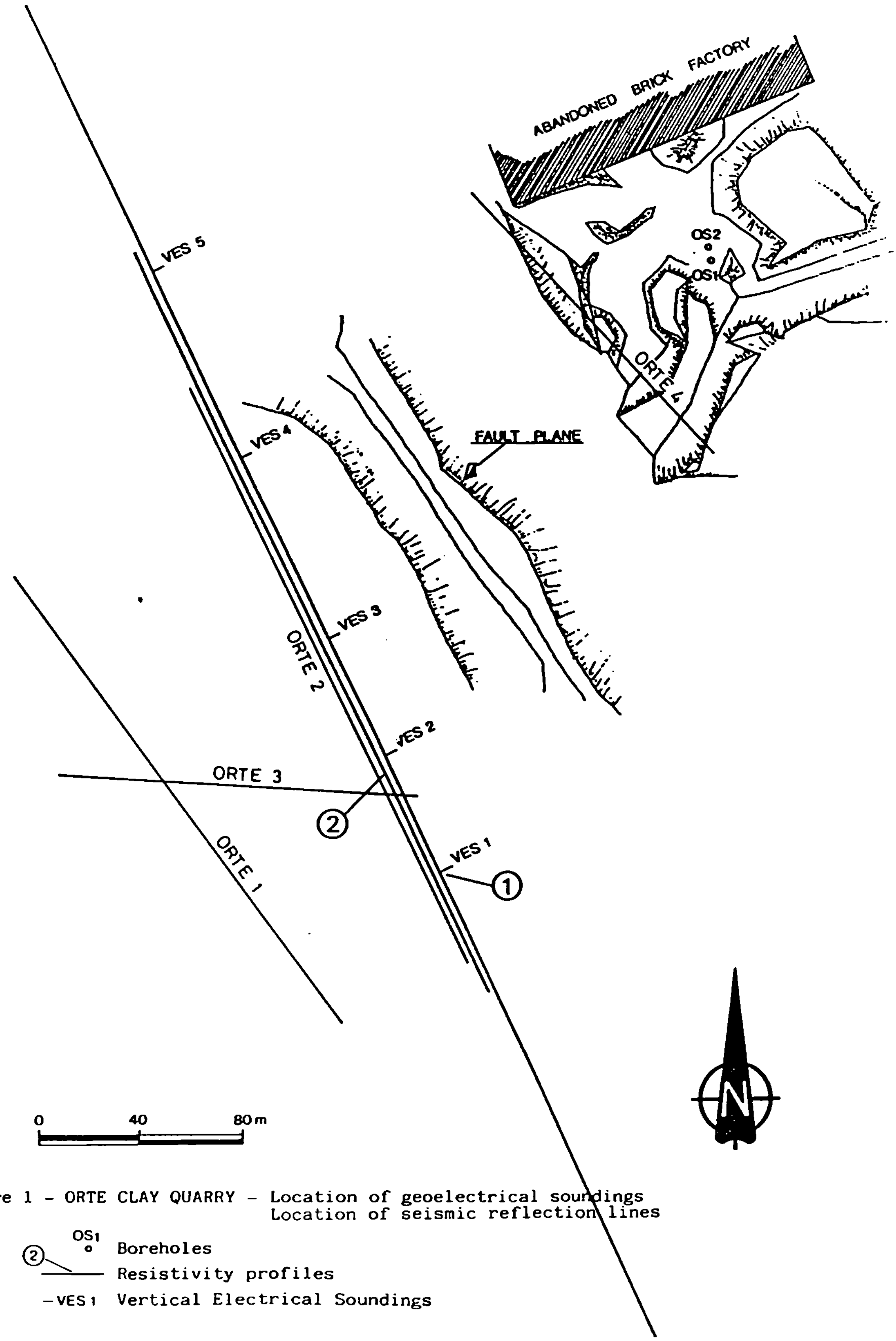


Figure 1 - ORTE CLAY QUARRY - Location of geoelectrical soundings
Location of seismic reflection lines

OS1 Boreholes

2 Resistivity profiles

-VES1 Vertical Electrical Soundings

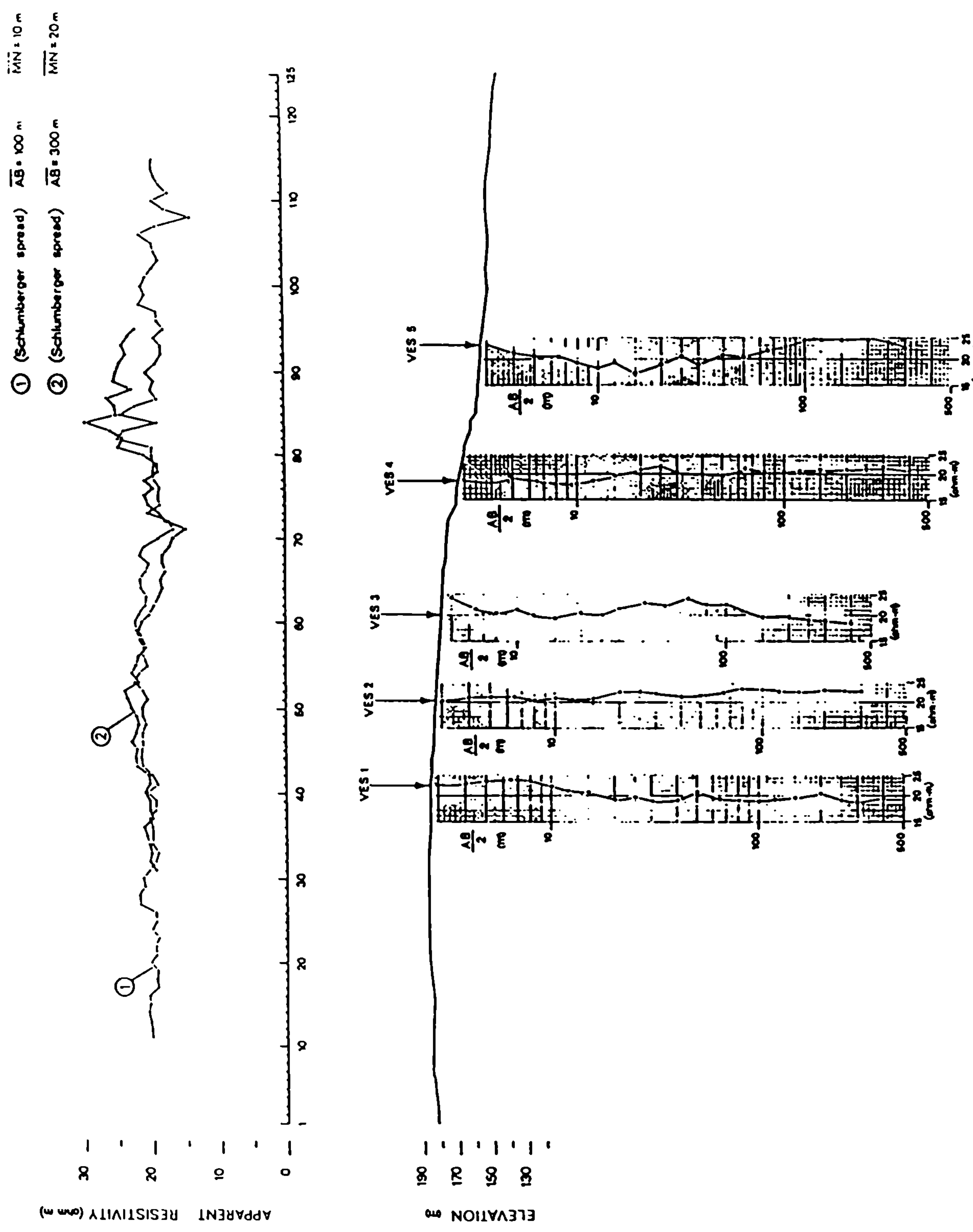


Figure 2 - Results of geoelectrical soundings

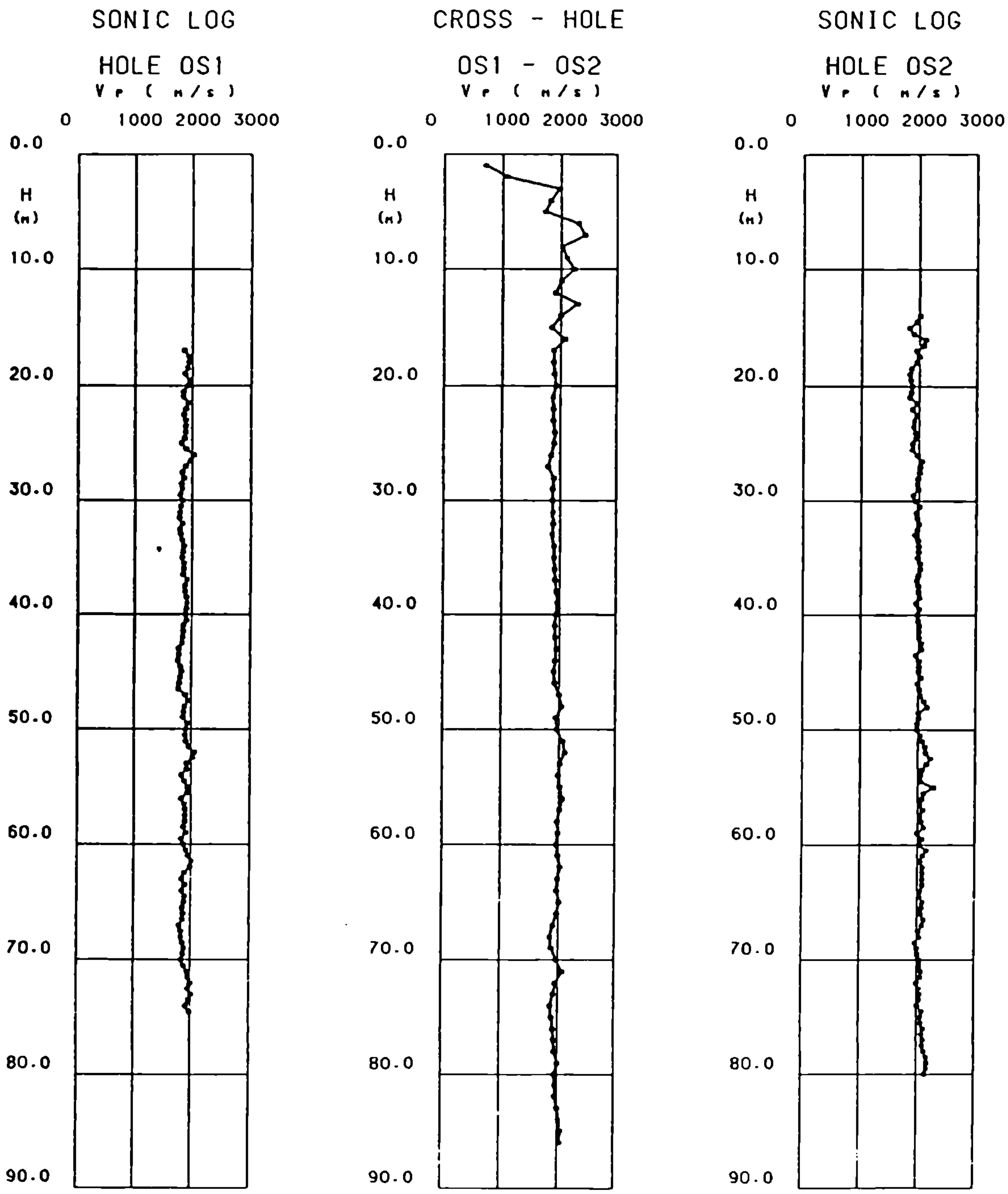


Figure 3 – Single hole and cross hole sonic velocities

CEC – FAULTS IN CLAYS PROJECT
SITE: ORTE CLAY QUARRY

N.o	DEPTH (m)		LENGTH (cm)	CASING	DESTINATION
1	31.00	31.50	50	metal	spare
2	34.00	34.60	60	metal	BGS
3	37.00	37.70	70	metal	spare
4	40.00	40.70	70	metal	BGS
5	43.00	43.40	40	metal	spare
6	46.00	46.60	60	metal	BGS
7	46.60	47.30	70	metal	ISMES
8	47.30	48.00	70	metal	ISMES
9	48.00	49.00	100	PVC	ISMES
10	49.20	49.80	60	metal	ISMES
11	49.80	50.50	70	metal	ISMES
12	50.50	51.20	70	metal	BGS
13	51.20	51.90	70	metal	ISMES
14	51.90	52.60	70	metal	BGS
15	52.60	53.00	40	metal	ISMES
16	53.00	53.50	50	metal	BGS
17	53.50	53.90	40	metal	ISMES
18	53.90	54.50	60	PVC	BGS
19	55.20	56.20	100	PVC	ISMES
20	60.00	61.00	100	PVC	BGS
21	63.00	63.40	100	PVC	ISMES
22	66.00	66.80	80	PVC	BGS

TABLE 1:
CHARACTERISTICS OF UNDISTURBED CLAY SAMPLES COLLECTED IN BOREHOLE OS2 AND THEIR FINAL DESTINATION

EVALUATION AND DEVELOPMENT OF GEOHYDROLOGICAL SURVEYING METHODS IN AREAS WITH SALING GROUNDWATER

Contractor: RIVM, Bilthoven, the Netherlands
Contract No: FI1W/0160
Duration of Contract: from November 1987 to May 1989
Period covered: January 1988 to December 1988
Project Leader: P. Glasbergen

A. OBJECTIVES AND SCOPE

Both deep and shallow sediments have been explored by oil and water companies respectively. A lack of the knowledge of geohydrological parameters and limited research experience exist with respect to Oligocene, Eocene and upper Cretaceons sediments in the subsurface of the Netherlands.

The aim of the project is:

- to investigate, evaluate and to indicate necessary developments of in-situ measurement systems and methods of geohydrological parameters in boreholes;
- to give recommandations for the set-up of geohydrological boreholes near salt structures in phase 2 of the Dutch nuclear waste disposal research programme;
- to test and compare measurement techniques.

B. WORK PROGRAMME

1.1 Investigation and evaluation of well test methods, the possibilities of wireline logging, downhole pH/Eh measurements, isotope dating techniques and sample methods of both undisturbed unconsolidated rock and pore water.

1.2 Recommendations for the construction of boreholes.

1.3 Recommendations for an optimized borehole and survey programme.

2.1 Selection of existing deep boreholes in the Netherlands for well tests.

2.2-2.8 Performing of well tests and sampling of formation water. Evaluation of the results of different methods and comparison with results from analyses on formation samples.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Current systems and methods, which are considered of practical use for the determination of the hydrogeological parameters of sediments around and above salt formations, have been investigated and evaluated. The results of this study have been reported in the first interim report no 728515001, which covers item 1.1 of the work programme described previously /1/.
Until the moment of the preparation of this report no salt structure has been designated for a geohydrological survey programme and it is unlikely this will happen before the termination of the present research project. Therefore item 1.2 and 1.3 of the work programme will be restricted to some general recommendations, which will be included in the final report.
Using existing wells, it is not possible to perform tests, which will lead to parameter values, representative of Oligocene, Eocene and Cretaceous formations around and above salt structures in North East of the Netherlands. Therefore the second part of the work programme will be focussed on the comparison of several existing techniques in low-permeability formations.

PROGRESS AND RESULTS

1.1 Investigation and evaluation of hydrogeological surveying methods of sediments overlying salt formations.

- Different well test methods have been evaluated with respect to the determination of the permeability and the formation pressure including, pumping tests, slug tests, packer tests, drill stem tests and wireline formation tests. The investigation showed that the vertical permeability in anisotropic formations can be difficult to determine with current well test methods. Formation pressure measurements can be subject to errors caused by the conversion of the water head into formation pressure. For long duration measurements this problem cannot be solved by measuring the pressure downhole due to instrumental drift.
- Wireline logs have been considered for the determination of a large number of hydrogeological parameters and characteristics including the clay content, porosity, permeability, temperature and pressure of the geological formation, furthermore the presence of karstic features in the formation and the salinity of the formation water. The determination of the permeability, pressure and salinity using wireline logs can be subject to large errors.
- Current sample methods have been reviewed. Core material turned out to be of large importance in a geohydrological site specific safety assessment. Chemical and mechanical alteration can be minimized using the right systems and methods. Various core systems are available for different formation types. A low recovery may however be expected in unconsolidated sands.
- Current water sample methods have been reviewed. Submersible pumps were considered for a standard chemical isotope analysis. Due to effects of degassing this method is supposed to be inaccurate in the determination of the pH and the Eh of the water of deep wells. Tools which take, in situ pressarised samples were briefly reviewed.
- The french CEA downhole pH/Eh probe was compared with the swedish SKB system. Both were developed for investigation in behalf of the nuclear waste disposal programme.

2.2-2.8 Sampling of formation water

- Several deep saline aquifers have been sampled and analysed for stable isotopes. It seems that sulfur-34 is a valuable tool to find indications for dissolution of evaporites. In shallow aquifers containing brackish water a part of the dissolved salts probably originate from evaporite dissolution (see fig. 1).

  Further research will be focussed on the testing of dating methods of groundwater beyond the limits of Carbon-14 dating. This part of the contract is carried out in cooperation with the Université Paris-Sud.

Reference

/1/. Langemeijer, H.D., Evaluation of hydrogeological surveying methods of sediments overlying salt formations. First interimreport no. 728515001, RIVM, Bilthoven.

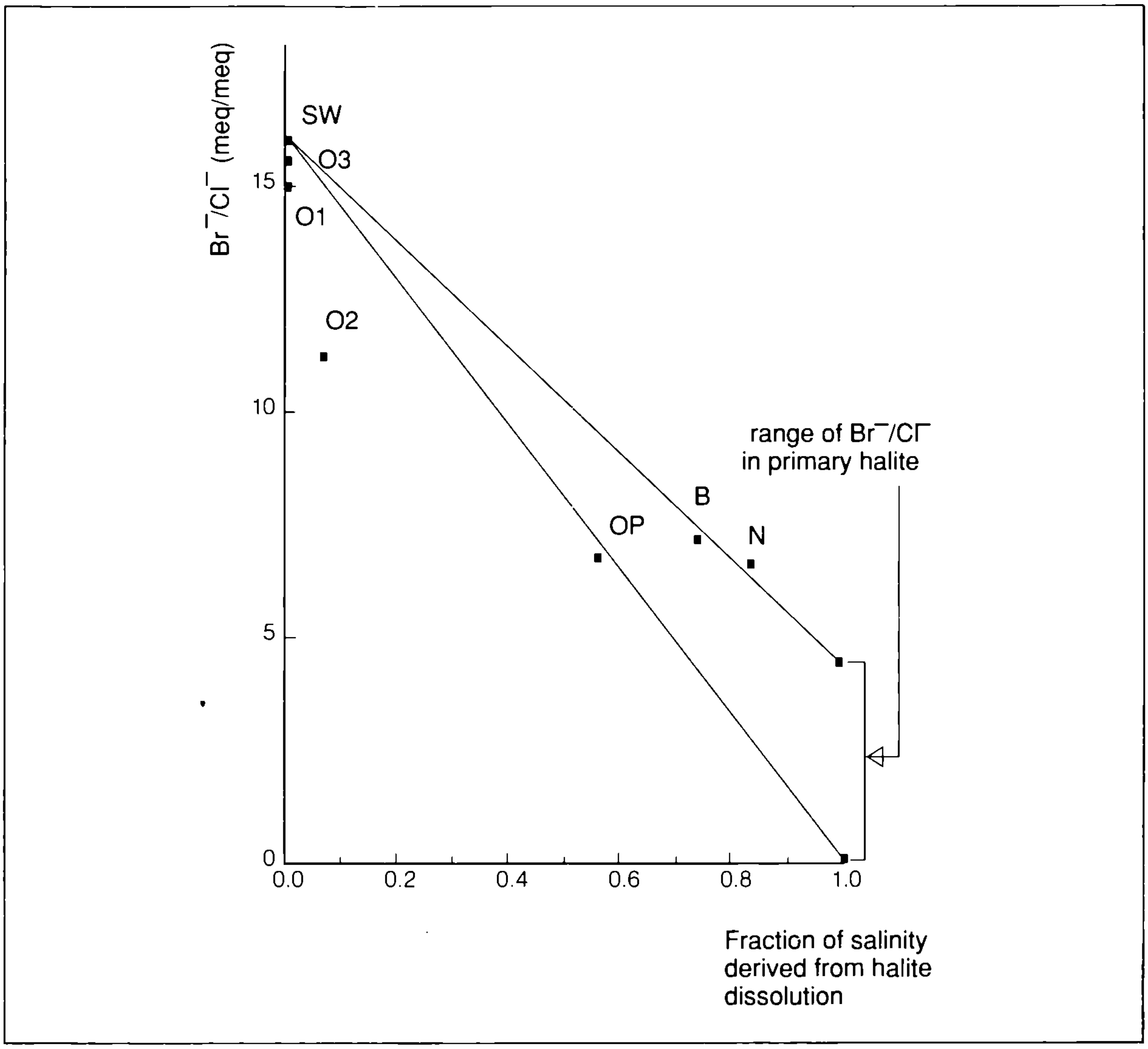


Fig.1. Plot of Br/Cl versus fraction of salinity derived from halite dissolution

Acquisition of physico-chemical properties of clay. Development of methodology for taking samples and making experiments.

Contractor : CEA, CEN Fontenay-aux-Roses, France
Contract n° : FI 1W - 0208
Working period : August 1988 - December 1989
Period covered : October 1988 - December 1988
Project leaders : H. COULON and A. LAJUDIE

A. OBJECTIVES AND SCOPE

The decision of radioactive waste disposal in deep clay formations cannot be taken until a full understanding has been acquired of the in situ water and radioelement transfer phenomena.

The first stage of the investigation consists in taking core samples and studying their properties in the laboratory. The difficulty of this approach resides in the need to take and store the samples without changing their intrinsic properties.

The general purpose of this study is thus the development of methodology which can be used to establish the modes of acquisition of the mechanical and chemical in situ properties required for evaluation of transfer phenomena: listing of the properties to be acquired and the disturbances caused by sample taking, selection of methods of core drilling, core handling and packaging, preparation of samples and analysis procedures.

The approach is essentially based on the experience acquired by CEN/SCK MOL with Boom clay, the observations carried out by ANDRA on the basis of deep clay core sample tests and the methods developed by the CEA for studying clay materials (contracts FI1W 0031 and 0061).

The second stage will consist of validating the methodology by carrying out core drilling at the MOL site using the previously established specifications, then comparing the results of analysis of the core samples with those of in-situ tests carried out in the underground laboratory, under a protocol which remains to be established.

B. WORK PROGRAMME

B.1 Listing of the physico-chemical parameters to be acquired for the purpose of assessing transfer phenomena in clay formations and the disturbances of the clay by the sampling method.

B.2 Selection of methods of taking, packaging and preserving samples. Establishment of experimental procedures. Their implementation at the Mol site in Belgium.

B.3 Comparison of in situ and laboratory readings. Validation of the methodology.

C. PROGRESS OF WORK AND OBTAINED RESULTS

Summary

After briefly describing the laws affecting mass transfer mechanisms in undisturbed clay materials, the parameters corresponding to these laws are reviewed.

An attempt is then made to identify all the types of disturbance associated with the method of taking samples which can affect the representativeness of the previously established parameters.

Finally, consideration is given to establishing a method of taking, packaging and preserving samples of deep clay with which it will be possible to keep or reconstitute the necessary physico-chemical properties before laboratory examination.

B.1 and B.2 are progressing normally.

B.3 is due to begin in September 1988.

Progress and results

B.1

Mass transfer phenomena in porous media are governed by the simultaneous action of two physico-chemical mechanisms : convection and diffusion.

Transfer by convection essentially depends on the properties on the medium and corresponds to Darcy's law :

$$Q = - k A H$$

where Q = the fluid flow rate in $m.s^{-1}$; k = the hydraulic conductivity or permeability ($m.s^{-1}$) ; A = sample area (m^2) ; H = the hydraulic gradient.

The hydraulic conductivity of clay depends on the porosity (n) or more specifically the effective porosity (ε), the tortuosity (τ), the density (ρ), the water content and the degree of saturation (Sr). It also depends on parameters affecting the percolation phase: temperature (which changes the kinematic viscosity of the fluids) and the chemical composition of the fluid (salinity).

Transfer by diffusion also depends on intrinsic properties of the medium, but also varies greatly with other physico-chemical properties of the diffusing substances.

The mechanics of diffusion can be expressed in a simplified manner, in the case of a one-directional model, by the following relationship:

$$J = - D \frac{\partial c}{\partial x}$$

where J = the flux of matter ($moles\ cm^{-2}\ .j^{-1}$) ; D = the diffusion coefficient ($cm^2\ .j^{-1}$) ; C = the concentration of the diffusing substance ($moles.cm^{-3}$).

In practice, a measurement is made of an apparent diffusion coefficient (D_x^*) which allows for the sorption phenomena (distribution coefficient of the substance considered between the solution and the porous medium (Kd)), the porosity (n) and the grain density (ρ_g).

In clay, the phenomena of sorption, precipitation and complex forming vary with Eh-pH conditions, the presence of organic materials and carbonates etc.

The operations of core drilling, taking of samples of clay and their preservation are liable to change the above properties, making laboratory tests pointless.

Indeed, during core drilling, the material which is under high lithostatic pressure is placed in conditions of practically zero external pressure. This results in deconsolidation and cracking of the clay. The interstitial water, which was initially at a positive pressure is placed at a negative pressure. The initially saturated clay becomes desaturated, allowing the migration of drilling fluids and air into the core sample. The initial properties of the fluids, the porosity, the specific density and the water content are thus greatly disturbed.

These phenomena are amplified during removal and handling of the core samples (modification of temperature, oxidation of organic matter, expansion of gaseous phases and modification of the Eh-pH equilibrium). Those in charge of operation of sampling and preparation of the plugs can make the changes in the stress fields, dehydration and oxidation of the clay worse.

B.2

By applying certain precautions and using suitable equipment, it is possible to avoid or limit the above disturbances.

Core drilling:

Of the core drilling rigs currently available, the "Mazier universal geotechnical core drilling rig" would appear to be the most suitable (figure 1). This is a triple-wall machine with a moving cutting sleeve which precedes the diamond-studded crown. This cutting sleeve penetrates the clay and protects it from the drilling fluids. During removal, injection of compressed air completely isolates the core sample, thus preventing any contamination. This core drilling rig could nevertheless be improved by using a less rigid internal casing and could possibly be lubricated to further reduce decompression of the core samples and internal friction. Furthermore, the use of an inert gas instead of compressed air would make it possible to avoid substantial oxidation. Optimization of the size of the core sample would make it possible to improve the geometrical coefficients of the drilling rig (coefficient of entry, wall and advance) and hence the quality of the samples.

Sample taking:

The use of an X-ray unit would make it possible to select the most representative sample, and those least disturbed while limiting the duration of work on the core samples (cutting of casings and exposure of the samples to air).

After cutting, the samples must be immediately packed in cells with which it is possible to re-apply pressure and preserve them under anaerobic conditions. It must be possible to store the samples under stable conditions (hygrometry and temperature of formation).

Preparation of test samples:

All the operations necessary for preparation of the laboratory tests must be carried out under a controlled atmosphere (glove box and anaerobic environment) in accordance with the protocols in force within CEN/SCK, MOL. We have therefore designed a thin-walled sample-taking tube which can be used in a glove box and enables direct transfer of the samples to the cell of a permeameter (CCE contract FI 1W/0031) with minimal disturbance. This equipment is to be tested with MOL clay.

All the protocols mentioned in this progress report are to be supplemented and improved before phase B.3 is started.

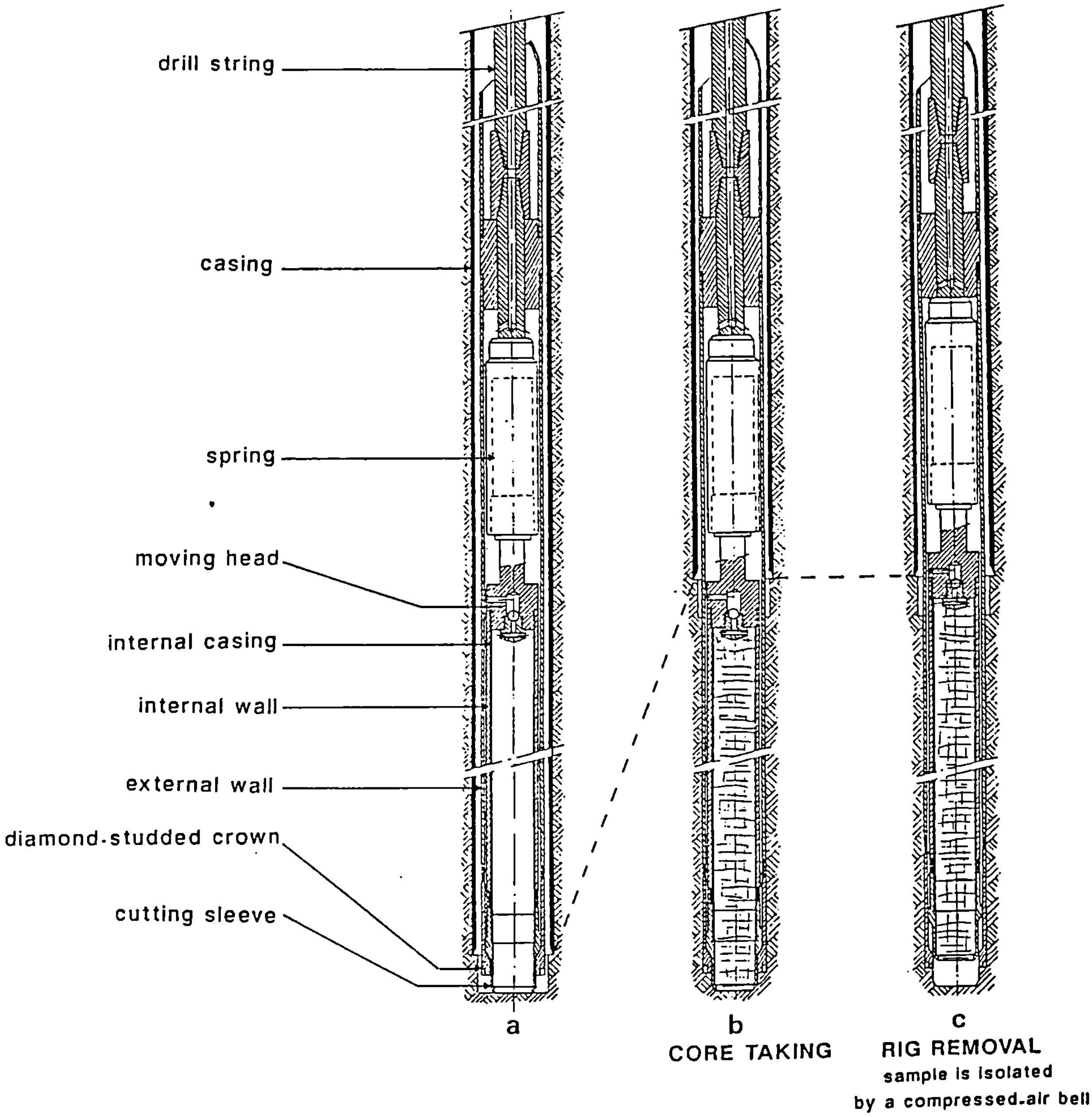


Figure 1 : Mazier universal geotechnical core drilling rig : Principle draft.

DEVELOPMENT AND EVALUATION OF AN X-RAY-ANALYTICAL TECHNIQUE FOR CORES FROM EXPLORATION HOLES DRILLED ON A WASTE-DISPOSAL SITE

Contractor : BRGM - Avenue de Concyr
B.P. 6009 - 45060 ORLÉANS CEDEX

Contract n°: FI 1W/0209

Duration of contract : from january 89 to july 90

Project leader : G. AUBERTIN.

A - OBJECTIVES AND SCOPE

The exploration of sites for the disposal of radioactive waste in various types of deep geological formations, and of possible locations for underground laboratories, is mainly based on the use of cored borehole. It is desirable to study the resulting cores in a non-destructive manner.

X-ray photography is widely used in mechanical and metallurgical industry.

In geology, only a few stationary X-ray machines are used for limited applications.

The aim of the research is thus, for different environments:

- to adapt radioscopic techniques to the study of geological materials that are available as drill core;
- to create a mobile unit that can be transported to the sites where cores are available and must be studied, such as a drill site, an underground laboratory or a vessel engaged in sediment studies;
- to allow non-destructive testing and study of the cores that are taken within a sleeve and require immediate confinement after being pulled from the hole, such as isthe case for clayey and salt-bearing materials that are destined for mechanical and geochemical tests;
- to have access, before any destructive operations, to the internal structure of cores, as a further aid to mechanical tests, and to geological and geochemical study;
- to adapt and develop image-processing software for the geological study of radioscopic images.

The equipment that is the subject of research differes from existing X-ray apparatus in the following points:

- an image-analyser tube will be fitted in order to be able to dispose of video images; image quality could be controlled and adapted to the geolocical material being studied, this by varying the feed voltage of the emitting tube;

- a diaphragm will allow adaptation to the size and shape of the samples to be analysed;

- the video image will allow real-time observation, as opposed to classical radiography where static images are obtained after an exposure of long duration. It will thus be possible to give a movement to the core and to obtain an analog-type log that can be correlated with logs obtained by other techniques;

- the video images can be digitally processed, for graphic restitution and the recognition of geological subjects.

B - WORK PROGRAMME

The contract will comprise the following stages;

a - Drawing up a detailed pilot study of the apparatus (consulting of suppliers of components, accurate definition of the apparatus and its operating environment).

b - Construction of the apparatus with the assistance of specialized suppliers.

c - Testing the apparatus on various cores of geological materials that can be envisaged for the disposal of radioactive waste, such a clay (in a sleeve), salt, granite and schist/shale.

d - Adapting and developing image-processing software with the following objectives:

- geological exploration and identification (lithology, structure, texture) without damage to the structural integrity of the sample;

- testing the suitability of this technique for identification of an homogeneous zone and for localizing mechanical and geochemical tests, in particular in the case of samples covered by a sleeve;

- improving the legibility and use of the images / use of image-processing software developed for remote-sensing purposes);

- correlating the X-ray logs with other borehole or geological logs.

e - Preparing documents that present the results obtained for various types of geological materials, and for different structures and textures being studied.

C - PROGRESS OF WORK AND OBTAINED RESULTS

1 - State of advancement

The programme is in its first month of activity. During this period, contacts were made with sub-contractors for the construction of the X-Core apparatus.

2 - Progress and results

The first orders, for the supply of parts and materials for the X-Core apparatus, have been sent out.

4.1.B. Geo-forecasting studies

GEOPROSPECTIVE MODELLING

Contractor: Bureau de Recherches Géologiques et Minières
BP 6009, 45060 Orléans Cedex - France

Contract n° FI 1W/0048

Duration of contract: from 1986 August 1st to 1989 March 31st

Period covered: 1988 January 1st to December 31st

Project leaders: P. Peaudecerf - J. Fourniguet

A. OBJECTIVES AND SCOPE

Since 1981, the BRGM has been working on the development of a method for systematically studying all the factors which might influence the evolution of a waste storage site and their interactions. One of the work phases consisted in a relative quantifying of the links between the factors and in modelling them so as to complete realistic scenarios. These operations are carried out with a simulator called CASTOR ("Construction automatique de scénarios d'évolution d'un site de stockage de radionucléides" /Automatic design of scenarios evolution of a radionuclide storage site).

The first simulations showed that a few main mechanisms governing site evolution should be represented more realistically.

They are essentially those concerning climate variations, weathering and erosion processes and relationships between stress and hydraulic parameters. The simulation programme will have to be modified accordingly to take these mechanisms and their interrelationships into account simultaneously.

This work should make the CASTOR code operational when applying the methodology to specific sites.

B. WORK PROGRAMME

Work programme will revolve around two main aspects:

1. Increasing knowledge and modelling of mechanisms which appeared essential in previous phases.
   a. Climatology: as well mathematical expression of fluctuations in the near past and future according astronomical data, as search for present climatic equivalents to past climates.
   b. Weathering erosion: better quantifying of rates according to lithology, slopes, vegetal cover, rainfall, temperature, and a tentative modelling of erosion processes.
   c. Relationships between stress and hydraulic parameters mainly in the case of the occurrence of an ice-cap covering the site area.
2. Improving the modelling of the phenomena and the representation of the results obtained.

C. PROGRESS OF WORK AND OBTAINED RESULTS

1. State of advancement

Item 1.a. has been treated and mainly completed in 1987 (see annual report). The work on weathering and erosion (item 1.b.) began in 1987 by a bibliographical analysis which produced a review of all processes (nature, rates, ...). A new and specific model of erosion was designed in 1988 since no available model was able to fit precisely with our requirements. Item 1.c. was also treated during the past year; the resulting model has been completed.

Item 2 has been largely begun in 1988 meanwhile and after the development of the new models.

2. Progress and results

2.1. Erosion and weathering processes

Most of the existing models concern the short term modelling (human scale of some tens of years). They require the supplying of abundant quantitative data on numerous parameters, obtained by field instrumentation during several years. These results are difficult if not impossible to extrapolate at the geologic scale (several tens of thousands years).

Some models deal with more global approaches of erosion but do not provide the quantitative data we need for a geoprospective modelling.

The newly created model allows the simulation of various erosional processes in one or several catchment basins, each of them defined by their topographic values in a rectangular grid.

It also takes into account the weathering of one or several geologic layers whatever its dip. Erosion and weathering are two coupled phenomena in the model.

The driving parameters of the model are as follows:

- erodability of soils (submitted to spatial variations),
- susceptibility of rocks to weathering (with spatial variations),
- climatic action, susceptible of variation as a function of time.

The progression of erosion/weathering front can be followed on screen by the evolution of a longitudinal profile in one or several basins (Figure 1).

2.2. Stress and hydraulic parameters under an ice-cap

The finite element method was used for the evaluation of the hydromechanic behaviour of the geologic formations under an ice-cap. The phenomenon is supposed to be isothermic since the arrival of the next glaciation is foreseen after several tens of thousands years, when the thermic activity of the storage is largely if not totally weakened.

A parametric survey was conducted mainly on the limit hydraulic conditions and on the progression rate of the ice-cap.

A quick progression of the ice stops the removal of interstitial water and is considered as the undrained condition. A speed of 100 m/year was adopted as a minimum. On the downstream part of the fictive site, an imposed pressure seems valuable since it corresponds to the initial stage, after the creation of a permafrost.

On the upstream part, two imposed conditions were tested: pressure and flow.

The permeability of the faults on the fictive site was considered as far greater than the one of geologic layers. This is supposed to be the worst possible case.

The permafrost is considered as impermeable thus stopping any water movement toward the surface.

The progression of the ice-cap induces a sub-horizontal movement of underground waters downstream. But the rapidity of the ice movement stops this flow rather quickly.

3. LIST OF PUBLICATIONS

- International Association of Hydrogeologists - International Symposium hold in Orleans (France) in June 1988: Prévision quantitative de

l'évolution à long terme des caractéristiques hydrogéologiques des sites de stockage profond, by Ch. Filippi & al.
ISPRA courses: Advanced Seminar on Risk Analysis in Nuclear Waste Management, May-June 1988:
Predictive geology in the analysis of the repository evolution; description of the relevant natural phenomena, by J. Fourniguet.
Modelling combined effects and making scenarios, by G. Aubertin.

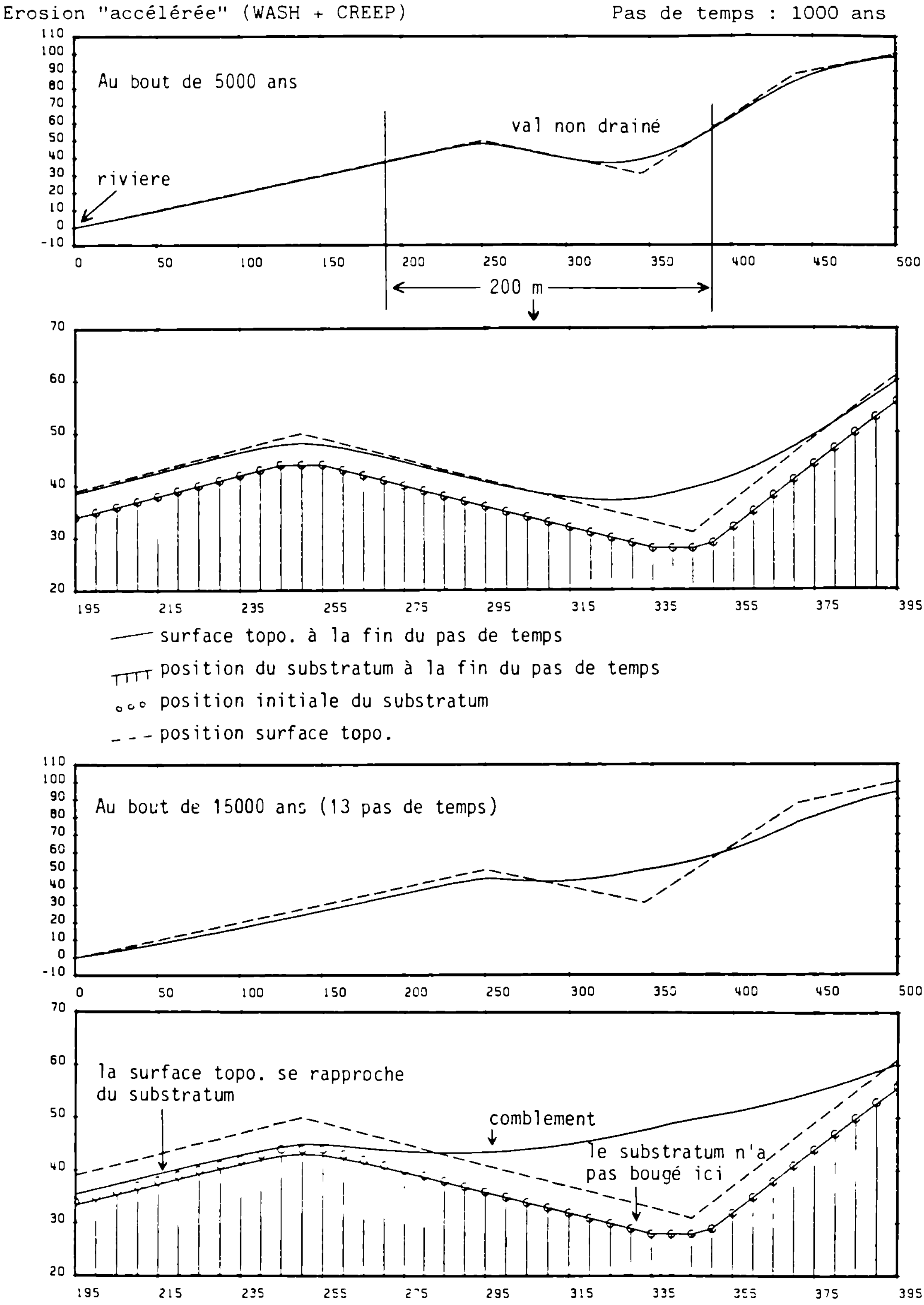


Fig. 1 - Example of evolution of a profil as regard weathering and erosion processes

4.1.C. Rock mechanics

IN-SITU CHARACTERISATION OF THE BEHAVIOUR OF DEEP CLAY LAYERS

Contractor	: ANDRA, PARIS, FRANCE
Contract n°	: F I1W/0049
Duration of contract	: August 86 - December 89
Period covered	: January 88 - December 88
Project leader	: R. ANDRE JEHAN

A - OBJECTIVES AND SCOPE

The objectives of this project are :

- To complete geomechanical investigations in the Boom deep clay formation, under natural conditions and after heating,

- To develop laboratory and in-situ methods to study deep clay layers, and to compare the results obtained by these two approachs.

This work forms a basis for dimensionning the storage facilities in deep clays. The interesting point is to study the time-lag behaviour of this material, at ambient and high temperature.

The sub-contractors of ANDRA for this project are :

- LMS (Laboratoire de Mécanique des Solides de l'Ecole Polytechnique) for the laboratory tests, for the conception of borehole probes in cooperation with SEDITECH-MAZIER and with the BRGM, and for the in-situ test interpretation,

- BRGM (Bureau de Recherches Géologiques et Minières) for the set up, and monitoring of in-situ test,

- SEDITECH-MAZIER for the design and fabribation of borehole probes.

Experiments are performed in the U.R.L. of CEN/SCK at MOL (Belgium).

B - WORK PROGRAMME

1. Experiments at ambient temperature
1.1. Long term borehole dilatometric tests
Three tests are considered, two in horizontal holes and one in a vertical hole. Surveys of experiments are still carried out.
1.1.1. Measurements
1.1.2. Interpretation
1.2. Laboratory tests on thick tube samples of clay
1.2.1. Experiments
1.2.2. Interpretation

2. Experiments at high temperature
Design of apparatus led to construction of the first probe in 1988.
2.1. Apparatus fabrication and test in laboratory
Preliminary tests on components of the probe are still undertaken.

C - PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

In-situ studies : the two first dilatometers are used for creep tests since December 1986 for the horizontal borehole and June 1987 for the vertical one. The third device is used for a relaxation test since December 1987 in horizontal borehole. Their loading states have been increased by stages. As for the heating dilatometer, the realization of the deformable part of the probe has brought up some difficulties relating to the adherence between the rubber body and its reinforcement. New apparatus pieces are fabricated.

Laboratory test : a set of different experiments (creep, relaxation and internal unloading tests) has been performed on Boom clay samples at varying moisture contents.

Progress and results

1. Experiments at ambient temperature
1.1. Borehole dilatometer long term test
1.1.1. Measurements

The detail of the boreholes loading is reported in tables I and II. The creep test principle is to apply a fixed pressure on the hole wall, smaller than the lithostatic stress, and to record the borehole closure. The relaxation test proceeds from the opposite principle : the borehole closure is imposed and the pressure variation is observed. The rough measurement of pressure must be corrected in order to consider the apparatus inertia pressure (about 0.4 MPa). Respectively, the rough convergence measurement must be added to the initial closure value, which occurs during the time-lag between drilling and setting of the probe, to give real convergence value. The initial closure is estimated from previous test in unlined boreholes in Boom clay.

After two previous stages the same pressure has been maintained by the first dilatometer for more than 1.5 year. The closure is yet increasing (figure 1).

For the second test, the decreasing stages of pressure were chosen smaller to multiply the points on the long term closure-confinement diagramm of the material and to better border its limit pressure of instability. However the closure is rather moderate in this vertical borehole (figure 2).

The last stage of the relaxation dilatometer shows that the increase of pressure stabilizes slowly and reaches a final value very near of the previous one (figure 3).

1.1.2. Interpretation

A rheological model of Boom clay has been established from numerous laboratory tests performed by another way. It reproduces in a satisfactory way the results of dilatometer tests, which confirms that scale effects are moderate for Boom clay /4/.

1.2. Thick tube test
1.2.1. Experiments

All the tests begin with an isotropic loading to the lithostatic pressure value of the sampling level. Then three types of experiments are carried out :

- regular internal unloading, with different rates (figure 4),
- creep (figure 5).
- relaxation : contrary to in-situ test, the initial pressure is not zero.

1.2.2. Interpretation

These tests give the short and long term closure-confinement curves of the material, either directly and continuously for the short term one through rapid internal unloading tests, or point by point from the stabilized couples of parameters (Pi, Ui) obtained in the long term tests. The results are coherent with each other and show the difference between short and long term behaviour, that is the effect of ultimate capacities of Boom clay.

2. Experiments at high temperature
2.1. Apparatus fabrication and test in laboratory

The probe body has been made. Two specimens of the deformable tube which surrounds the central probe has been realised but they failed during a preliminary inflation test, due to a bad adherence between the rubber material and its reinforcement. New specimens are being made and in-situ tests should start by mid. 1989.

REFERENCES

/1/ - ANDRE JEHAN, R. and al, OCDE/AEN workshop on excavation responses in deep radioactive waste repositories, Winnipeg, Canada, 26-28 April 1988.

/2/ - ANDRE JEHAN, R. and al, International Symposium on Rock Mechanics "Rock mechanics and Power Plants", Madrid, Spain, 13-16 September 1988.

/3/ - ROUSSET, G. and al, CEC contractor's meeting on "Geomechanics of clays for radioactive waste disposal", Brussels, Belgium, 1-2 December 1988.

/4/ - ROUSSET, G. "Comportement mécanique des argiles profondes - Application au stockage des déchets radioactifs", Thesis E.N.P.C., July 1988.

TABLE I : DETAIL OF LOADING OF BOREHOLES CREEP DILATOMETRIC TESTS

TEST	Stage	Rough Pressure (MPa)	Corrected Pressure (MPa)	Duration (days)
Test 1	1	3.65	3.25	35
(horizontal)	2	3.10	2.70	130
	3	2.40	2.00	> 560
Test 2	1	3.60	3.20	120
(vertical)	2	3.10	2.70	130
	3	2.70	2.30	90
	4	2.35	1.95	120
	5	2.10	1.70	> 90

TABLE II : DETAIL OF LOADING OF BOREHOLE RELAXATION DILATOMETRIC TEST

TEST	Stage	Corrected closure (%)	Duration (days)
Test 3	1	1	100
(horizontal)	2	2.5	> 260

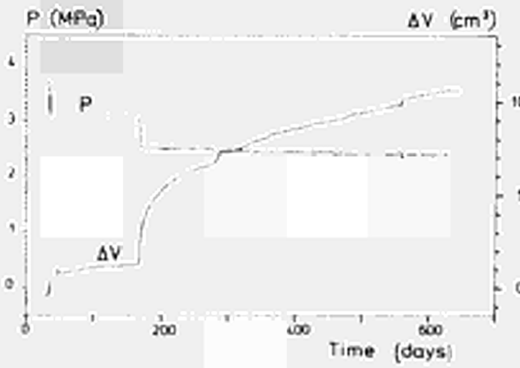


Figure 1 :

Rough measurements of dilatometer n° 1 (horizontal)

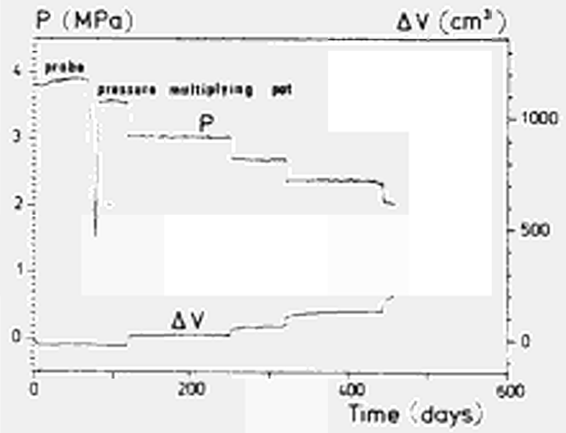


Figure 2 :

Rough measurements of dilatometer n° 2 (vertical)

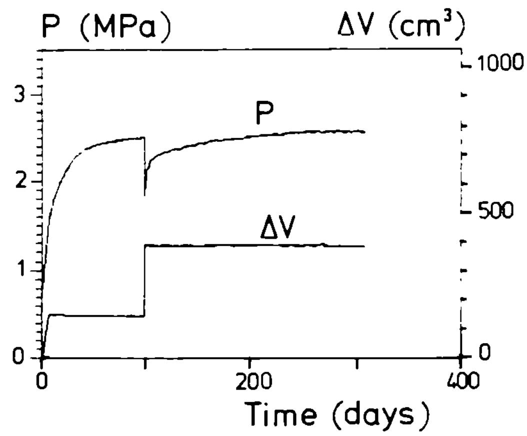


Figure 3 :
Rough measurements of
dilatometer .n° 3 (horizontal)

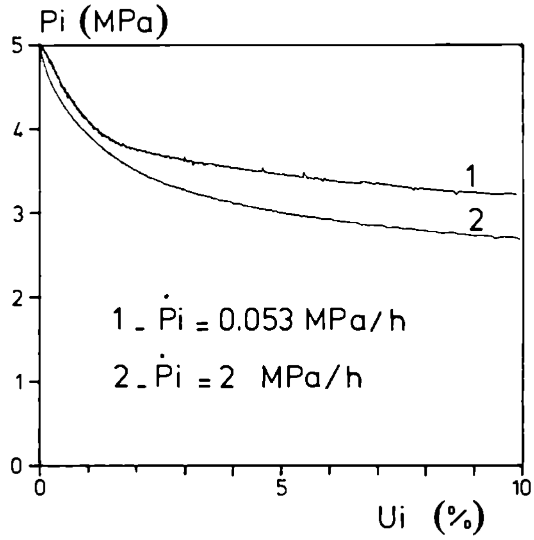
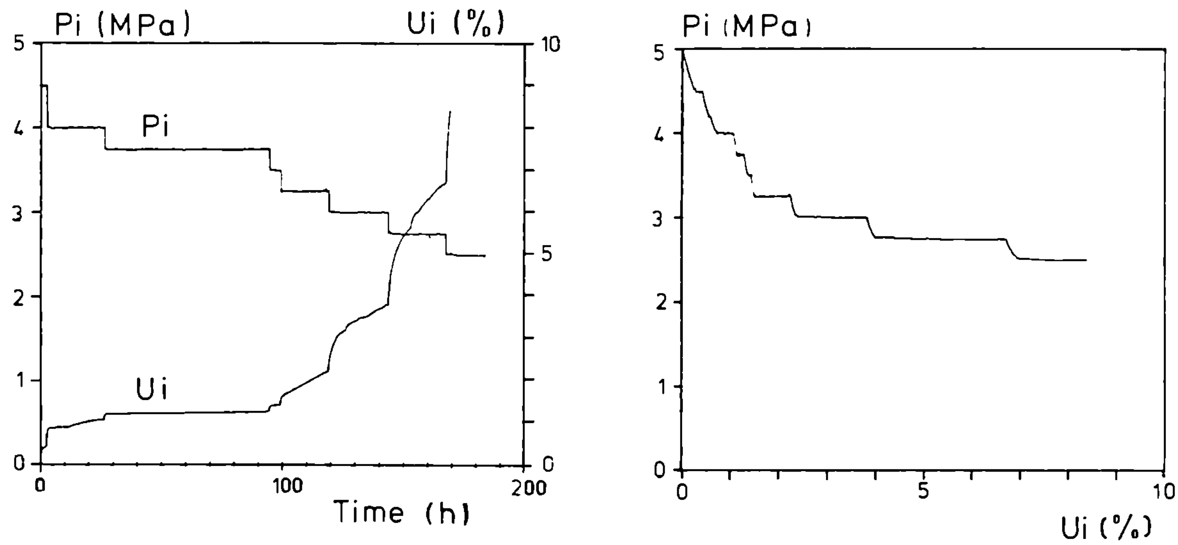


Figure 4 :
Internal unloading tests on
thickwalled tube sample closure
confinement curve



Figures 5 : creep test on thickwalled tube sample

5a : Pressure and closure
versus time

5b : Closure - confinement curve

CENTRIFUGE MODELING OF SALT DOMES

Contractor	: ANDRA, PARIS, FRANCE
Contract n°	: F I1W/050
Duration of contract	: October 1986 - March 1989
Period covered	: January 1988 - December 1988
Project leader	: R. ANDRE JEHAN

A - OBJECTIVES AND SCOPE

The aim of the research is to study experimentally the different stages in the history of salt domes (or diapirs). It completes a previous CEC contract which ended by autumn 85. The major part of the work is the inclusion of three-dimensional models for comparison with two-dimentional ones of the first period.

Tests are performed on equivalent materials on the large centrifuge of CESTA (in Bordeaux). Results can be applied to dome evolution through similitude transposition. The purpose of these experiments is to record the main onset and development mechanisms of salt diapirs.

B - WORK PROGRAMME

1. Two-dimensional tests :
   Different possible starting factors of movement are tested.

2. Three-dimensional tests :
   Comparable tests with two-dimensional ones are performed.

C - PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The experimental equipment, particularly the system to visualize the three-dimensional model inside the cylindrical cell and the cutting device to obtain thin sections, are now completed and operational.

The real geological stratigraphy is schematized by two layers, the salt and its overburden. Equivalent materials used in the models were determined previously, essentially clay mixtures with respect to the difference between average density of the overburden and salt density.

Tests in two- and three-demensional cells were undertaken with different geometries of "overburden" and "salt", in order to record the onset and evolution mechanisms of salt diapirs.

During 1988 the CESTA centrifuge broke down several times, with a very tight planning, so an additional week of tests has to be reported to February 1989, and the contract was given a 6 months prolongation.

Progress and results

1. Two-dimensional tests

Tests in two-dimensional cells were performed to study the starting factors of diapirism : initial notch in the interface "overburden" - "salt" (standing for a preliminary dome), slope of the interface and overloading of the ground surface. Salt is replaced by white clay, with a density of 1.54 g/cm^3 and overburden by a grey clay loaded with corundum to a density of 1.86 g/cm^3.

Effect of the first factor was investigated in 1987.

Tests with different slopes of interface show that a vertical interface and one with an initial slope of 45° cause a very progressive layers reversing beginning for an acceleration of 10 or 20 g. An initial slope of 14° produces an abrupt diapir evolution at about 50 g.

The overloading of the surface represents a topographical irregularity. It is rapidly flattened by centrifugation, but causes however abrupt diapir development, starting at about 40 g.

A last parameter was tested, affecting diapir evolution rather than phenomenon onset : the ratio of width upon thickness of the model. An experiment was made with thin overburden and salt layers, to approach in situ conditions, and with an initial noth in the interface. The acceleration value which started diapirism was the same as for thick layers model, but the evolutions slightly differ.

2. Three-dimensional tests

Four tests were performed for the duration of the contract, with same materials as for the previous experiments : white light clay for salt and grey loaded clay for overburden.

Two of them were realised in 1987, with initial cylindrical notch at the interface of the models.

For the third test, the two materials were separated by an interface with a slope of 45°. Movement started at 25 g, with quiet evolution.

The fourth model supported a loading of the free surface by grey heavy clay. Diapirism began at 30 g.

Study on fracturing and microfissuration of granite

Contractor : Commissariat à l'Energie Atomique, Fontenay-aux-Roses, France
Contract n° : FI/1W/0053 F
Duration of contract : from 01.12.1986 to 01.06.1989
Period covered :
Project Leader : S. DERLICH

A. OBJECTIVES AND SCOPE

Digging of drifts or tunnels with explosives or tunnelling machines and boring holes induce in massive rocks (granite) two kinds of stresses : the first one are stress waves and the second one are variations of the natural stresses induced by the underground openings.

Explosives loads induce cracks of tens of centimetres, or microcracks with thicknesses of some microns. Depending on the value of natural stresses and also on the shape of openings, slabing of walls and boreholes or spalling of cores may occur. Such fractures modify the mechanical characteristics of the medium and create new flow paths for the underground waters.

Few studies have been made in order to evaluate these possible effects.

Our purpose is to determine the extension of fractures around mine openings and to try to quantify fracturing or microfracturing of cores versus lithostatic pressures.

B. WORK PROGRAMME

1. Sampling

Fractures and microfractures will be studied on samples of granite.

1.1. Samples from 100 m depth

Study of explosive induced fractures in drift walls and of stress release in zones far from explosive mining ; release of stress will be studied by overcoring.

1.2. Samples from surface to 1000 meter depth (Auriat borehole).

2. Preparation of rock samples

Successive impregnations will be made with different dyes in order to identify the fractures induced by each operation.

3. Study of fractures

Different sizes will be considered :

. thicknesses from 10^{-3} to 10^{-6} m.

. " or pores less than 10^{-6} m

Several methods will be tested and the best ones used for the study.

3.1. Volume study

. Optical examens on thin sections cut following three directions.
. Electron microscope
. Castaing micro probe
. Porosimetry tests

3.2. Surface study

Fissures will be opened and the two surfaces limiting the voids will be studied (rugosity).

4. Interpretation

It will be a tentative to find a relation between the variation of stresses applied to the sample and the observed mechanical effects of fracturing

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Sampling

1. In mine (100 m depth)

. Coring of explosive induced fractures in drift walls will be achieved in february

. Large diameter cores (0,75 m to 1 m diameter) have been drilled in the groundfloor of the experimental room in order to minimise stresses relieve and to have quite a large volume of rock to observe and study fractures to 2 m depth in the rock mass.

2. In boreholes

Samples of cores were selected every 100 m between 998 m and 200 m below ground surface.

Studies

Two main methods were studied : geometrical characteristics of small fractures in thin sections, porosity of voids by mercury porosimetry.

They give the void distribution in the rock but orientation studies will be made to determine the influence of stresses orientation and petrographic components of the granite.

PROGRESS AND RESULTS

Preliminary results

Porosity

Tests have been realised on fresh samples and on weathered samples of granite from Auriat boreholes.

Porosimeter sensitivity tests

Previous studies have been made on Auriat granite samples. They have shown a 10 % variation between the porosities of good granite and of heated samples to 200°C.

Porosity comes from the fissures which can be opened or shut under mechanical loads and from pores included in crystalline or intercrystalline structures. These pores have also volume variations under loads. These strains obey the law of elasticity and are proportionnal to the stresses. Compressibility tests (1) give a fracture porosity of 0.05 % of the total volume and a total porosity of 2 % (Hg porosimeter). So the fracture porosity is about 2.5 % of the total porosity and to determine one tenth of its variation, a sensitivity of 5.10^{-3} for the total porosity measurement is needed.

Geometrical characteristics of fractures identified in granite samples

The objective of the study is to quantify geometrical parameters such as :

- . number of fractures NF
- . cumulated perimeter PC mm
- . cumulated surface SC mm^2
- . average perimeter PM mm
- . average surface SM mm^2
- . average orientation of fracture OM (o)
- . distribution of fractures limited by different crystals : biotite (BIO), alkali feldspar (FEL), plagioclase (PLA), quartz (QUA)
- . mean length of fractures LM mm
- . mean width of fractures lM mm

Measurements have been made on compact granite and weathered samples in order to estimate the validity of the method. Results are presented in figures 1 and 2 and in Table I.

Comparison of histograms of characteristics such as length and width of fractures or orientations, show few difference between the two groups of samples.

Bibliography

(1) Contrat CEA - Ecole Polytechnique - Laboratoire de Mécanique des Solides
Influence de la fissuration sur les caractéristiques thermiques d'un granite (étude en cours)

Figure 1

Histogramme des longueurs

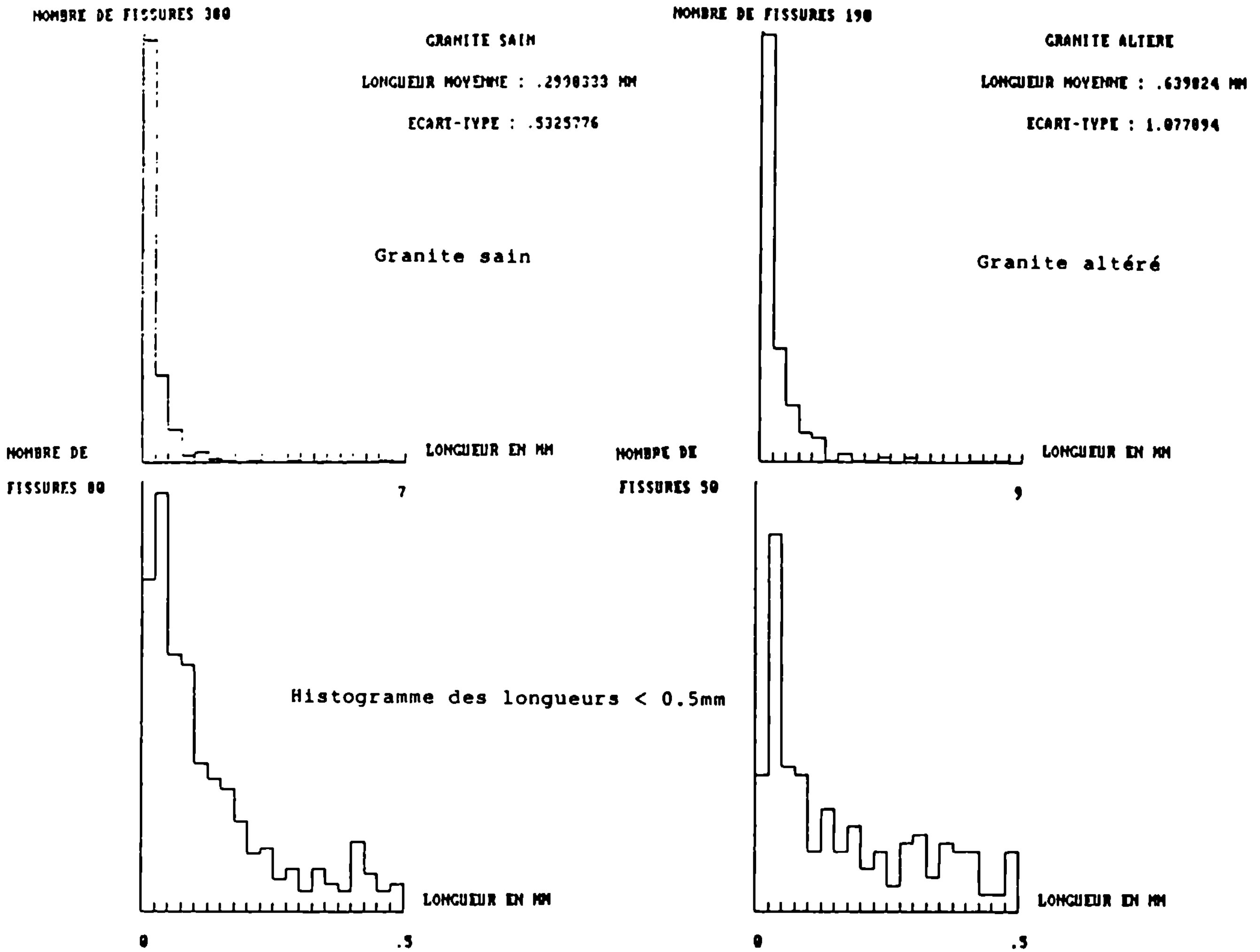


Figure 2

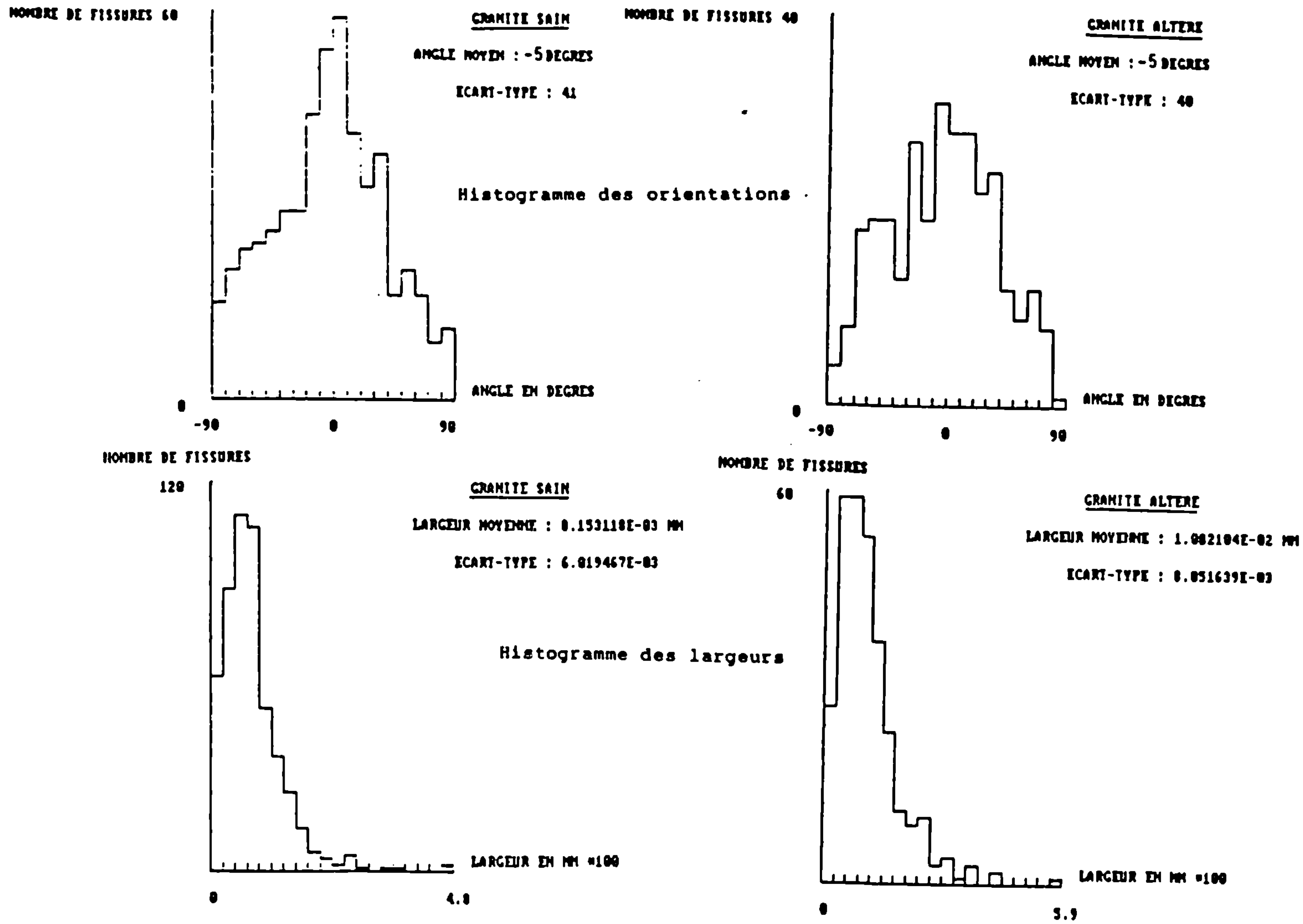

NOMBRE DE FISSURES 60
GRANITE SAIN
ANGLE MOYEN : -5 DEGRES
ECART-TYPE : 41
ANGLE EN DEGRES
0
-90
0
90
NOMBRE DE FISSURES 40
GRANITE ALTERE
ANGLE MOYEN : -5 DEGRES
ECART-TYPE : 40
Histogramme des orientations
ANGLE EN DEGRES
0
-90
0
90
NOMBRE DE FISSURES
120
GRANITE SAIN
LARGEUR MOYENNE : 0.153118E-03 MM
ECART-TYPE : 6.019467E-03
Histogramme des largeurs
LARGEUR EN MM *100
0
4.0
NOMBRE DE FISSURES
60
GRANITE ALTERE
LARGEUR MOYENNE : 1.082104E-02 MM
ECART-TYPE : 8.051639E-03
LARGEUR EN MM *100
0
5.9

Table 1

échantillon	NF	P C (mm)	S C (mm²)	PM (mm)	SM (mm²)	OM (°)	BIO (%)	FEL (%)	PLA (%)	QUA (%)	INTRA (%)	INTER (%)	INTRA-INTER (%)	lM (mm)	LM (mm)
B 11	81	75	0.14	0.93	0.0017	-8	0	62	19	25	91	8	1	0.0047	0.46
B 12	196	170	0.66	0.87	0.0034	-4	1	19	34	53	82	13	5	0.0090	0.43
B 13	77	30	0.12	0.39	0.0016	-12	1	9	48	48	84	10	6	0.0085	0.19
B 14	153	37	0.13	0.24	0.0008	0	1	24	22	65	79	16	5	0.0087	0.11
B 1	507	312	1.05	0.62	0.0021	-5	1	26	30	51	83	13	4	0.0081	0.30

Tableau 1 - Résultats d'analyse des fissures d'un échantillon de granite sain.

échantillon	NF	P C (mm)	S C (mm²)	PM (mm)	SM (mm²)	OM (°)	BIO (%)	FEL (%)	PLA (%)	QUA (%)	INTRA (%)	INTER (%)	INTRA INTER (%)	lM (mm)	LM (mm)
B21	93	102	0.74	1.10	0.0080	1	6	22	33	44	86	10	4	0.0120	0.54
B22	41	15	0.04	0.37	0.0010	-2	0	15	7	78	88	12	0	0.0061	0.18
B23	58	23	0.15	0.40	0.0026	-2	0	17	41	47	81	17	2	0.0110	0.19
B24	63	17	0.05	0.27	0.0008	-2	2	33	29	43	70	27	3	0.0069	0.12
B2	255	157	0.98	0.62	0.0038	-1	3	22	30	50	81	16	3	0.0096	0.30

Résultats d'analyse des fissures d'un échantillon de granite peu altéré.

FURTHER BENCHMARK EXERCISES TO COMPARE GEOMECHANICAL COMPUTER CODES FOR SALT (COSA II)

Contractor : WS Atkins Engineering Sciences, Epsom UK
Contract No : FI1W/0054
Duration of Contract : November 1986 - January 1989
Project Leader : N C Knowles

A. OBJECTIVES AND SCOPE

Research into geomechanical aspects of RAW repositories in salt formations has been active in the European Community for nearly two decades, with particular interest being placed on problems of heat producing waste. Central to this work is the prediction of stresses and deformations in the host strata, for which a number of computer codes have been used /1/. A preliminary exercise ("COSA 1") to compare the ability of the different codes /2/ provided a limited "snapshot" of the European capability to predict the behaviour of rock salt under well defined conditions. The purpose of the present contract is to extend the comparison to more complex but realistic situations.

Comparison problems in COSA I were relatively simple, and a number of difficulties to do with modelling the in-situ behaviour of rock salt were deliberately avoided. The present exercise is directed at comparisons of realistic, albeit relatively short-term, in-situ behaviour. Emphasis is placed on the requirement to predict (rather than replicate) real-life behaviour and individual participants are allowed considerable freedom to characterise the physical situation and material behaviour according to the dictates of their experience.

There are 10 participants in the exercise, each acting as a sub-contractor to the coordinator. In addition two independent experts provide advice as necessary on aspects of salt rheology (Table I).

B. WORK PROGRAMME

B.1 Participants jointly, at plenary meetings, agree programme of suitable in-situ benchmark problems.
B.2 Coordinator prepares discussion documents as necessary and circulates to participants.
B.3 Coordinator prepares and circulates detailed specifications of agreed problems.
B.4 Participants solve benchmark problems as specified by coordinator, to the best of their ability using appropriate codes.
B.5 Coordinator collects and collates results and other data from participants.
B.6 Coordinator prepares draft reports for discussion at plenary meetings to be held approximately every 6 months.
B.7 Coordinator prepares and issues final reports taking due account of participants comments.

C. PROGRESS OF WORK AND RESULTS OBTAINED

State of Advancement

Three benchmark problems were identified at the start of the exercise. They are based on the series of experiments performed by ECN in the 300m dry-drilled borehole at the Asse research facility.

The first problem is the prediction of the isothermal free convergence (IFC) at the bottom of the borehole over a period of some 800 days. This was largely completed in 1987 but a number of additional results were submitted in the present reporting period.

The second problem was to predict the behaviour (including pressure build-up) as the borehole wall converged onto a heated probe (HPP1). Preliminary results for this benchmark were collated and subsequently discussed at a plenary meeting of participants at Ismes Bergamo February 2-3.

The third problem was to predict the free convergence of the borehole during a heater test (HFC1). The specification of this benchmark was discussed at the plenary meeting in Bergamo, it was then finalised and calculations were performed, prior to comparison and discussion of the results at a plenary meeting in Madrid September 20-21. The predictions again agree qualitatively with the experimentally measured behaviour, but quantitatively there are considerable differences, which can be attributed to the various constitutive models used.

A final report for the project as a whole has been drafted and issued to participants for comment, prior to submission to the CEC in January 1989.

Progress and Results

1. IFC Benchmark

This benchmark was reported briefly in last years report and will not be duplicated here. The additional results submitted this year are in broad agreement.

2. HPPI Benchmark

The experimental test on which this benchmark was based, involved monitoring the variation with time of pressure and temperature on a heated probe fixed in the borehole at a depth of 262m over a period of 60 days. Participants were asked to predict this behaviour. As with the IFC benchmark, the specification gave no details of the measured behaviour and defined only the physical arrangements for the test. It was left for each participant to decide how best to model the test, especially with regard to the material constitutive behaviour. There was also some discussion about the importance of the inevitable small gaps between the probe and the borehole wall, but nevertheless perfect contact was specified in order to confine the model variations to reasonable bounds. Subsequently the effect of gaps was investigated by a number of participants and was shown to be relatively minor compared to other influences.

In general there was good agreement among the predictions of the temperature field but the predictions of the temperature at the borehole were all higher than measured. The reasons are attributed to a variety of sources, including uncertainty about thermal properties of the salt, heat loss in the cables and experimental error.

Predictions of the geo-mechanical behaviour agree qualitatively but differ substantially in absolute terms (eg predicted peak pressures differ by a factor of approximately 2.5). The behaviour is dominated by the early thermo-elastic response to a steadily increasing temperature and accordingly is sensitive to spatial temperature gradient and to the values used for Elastic modulus and coefficient of expansion.

3. HFCI Benchmark

The heater test on which this benchmark was based, followed chronologically the HPP test and was conducted at a depth of 237m down the borehole. The benchmark required the prediction of the temporal and spatial temperature and stress variations is the salt mass adjacent to the borehole over a period of 20 days due to a heat source of approximate 5KW in the borehole.

As before, participants were, quite deliberately, allowed complete freedom in modelling the test. In practice the key issues again centred on the material constitutive model.

The temperature predictions for the salt generally agree well with each other. Moreover, given the difficulties encountered in obtaining reliable temperature measurements during the test, they appear to be consistent with reality.

The stress and convergence predictions exhibit wider variation (maximum displacements vary by a factor of approximately 4) but are qualitatively, similar. In contrast to the HPP benchmark the behaviour at the borehole wall exhibits a complex interaction between the visco-elastic response to a time varying thermal straining and the thermally accelerated creep observed in the IFC. The predictions bound the measured behaviour and the variations can be attributed to the different models used to account for the material behaviour.

The 3 benchmarks highlight the difficulty in establishing reliable material models for "blind" predictions. It appears that most models can be "tuned" to replicate measured behaviour satisfactorily, but the reliability of such models to predict unknown behaviour is unproved.

List of Publications

1. Lowe M.J.S., Knowles N.C. 'Project COSA - A Benchmark of Computer Codes for the Geomechanical Behaviour of Rocksalt'. Proc. Int. Conf. Reliability of Methods for Engineering Analysis, Swansea, 1986.
2. Knowles N.C., Lowe M.J.S. 'A Benchmark Exercise on a Thermal Elasto-Creep Problem'. Structural Analysis Systems World Conference, Paris, 1986.
3. Knowles N.C. 'Project COSA - A Benchmark of Computer Codes for the Thermal-Mechanical Behaviour of Rock Salt'. Workshop on Mathematical Modelling for Radioactive Waste Repositories, Madrid, 1986.
4. Knowles N.C., Lowe M.J.S. 'Some Experiences of Finite Element Calculations in a European Benchmark Exercise'. NAFEMS Int. Conf. on Quality Assurance and Standards in Finite Element Analysis, Brighton, 1987.
5. Knowles N.C., Lowe M.J.S., Piper D. 'An Update on Project COSA' Trans. 9th Int. Conf. on Structural Mechanics in Reactor Technology, Lousanne, 1987.
6. Come B. 'Benchmarking Rock Mechanics Computer Codes : The Community Project COSA' Proc. 6th ISRM Congress on Rock Mechanics, Montreal, 1987.

7. Come B. 'Le Projet Communautaire COSA : Un Exemple d'Intercomparaison de Codes de Calcul Geomecaniques pour le Sel' Revue Francaise de Geotechnique, 1987.
8. Piper D., Knowles N.C. 'Some Computational Experiences of a Geomechanical Benchmark in Rock Salt'. Proc. 6th Int. Conf. on Numerical methods in Geomechanics, Innsbruck, 1988.
9. Knowles N.C., Come B. 'A Progress Report on Project COSA'. Workshop on Excavation Response in Deep Radioactive Waste Repositories, Winnipeg, 1988.
10. Lowe, M.J.S., Knowles N.C. 'Further Benchmark Exercises to Compare Computer Codes for Salt' Draft Final Report to CEC. WS Atkins Engineering Sciences December 1988.

References

/1/ BROYD, T.W., et al. CEC EUR Report 8669 (1985)

/2/ LOWE, M.J.S.L., KNOWLES, N.C., CEC EUR Report 10760 EN

Table I : List of Organisations involved in COSA II

WS ATKINS ENGINEERING SCIENCES - Epsom (UK)	Co-ordinator
FORAKY - Brussels (B)	Calculation Team
LGC - Louvain-la-Neuve (B)	Calculation Team
KfK - Karlsruhe (D)	Calculation Team
RWTH - Aachen (D)	Calculation Team
CEA-DEMT - Saclay (F)	Calculation Team
EMP - Ecole des Mines - Fontainebleau (F)	Calculation Team
LMS - Ecole Polytechnique - Palaiseau (F)	Calculation Team
ISMES - Bergamo (I)	Calculation Team
ECN - Petten (NL)	Calculation Team
ENRESA/ETSIM - Madrid	Calculation Team
GSF - Braunschweig (D)	Salt Specialist
Technical University Delft (NL)	Salt Specialist

Geomechanical behaviour of clay at ambient and elevated temperature conditions

Contractor : SCK/CEN, Mol, Belgium
Contract No : FI1W/0055/B
Duration of contract : October 1986 to December 1989
Period covered : January 1988 - December 1988
Project leader : A.A. Bonne, B. Neerdael

A. OBJECTIVES AND SCOPE

In 1974 SCK/CEN launched a R&D-programme concerning the possibilities for disposal of high-level solidified and alpha-bearing radioactive wastes in a continental stratiform clay formation (Boom clay) situated below its own site. Several specific investigations still need to be further undertaken in order to characterise more accurately the argillaceous formation in view of assessing its appropriateness for hosting radioactive waste as well from engineering point of view as for long term safety and performance evaluations. In support of these also further modelling efforts are required in order to improve and confirm our prediction capability.

This research is focussed upon in situ investigations related to the (thermo-)mechanical behaviour of clay.

Several field tests are developed and performed jointly with ANDRA.

B. WORK PROGRAMME

B.1.1. Stress measurements in non-frozen clay
B.1.2. Fracturing of clay
B.1.3. Long-term dilatometric tests.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Most of the tests intended to study the behaviour of Boom clay under elevated temperature conditions have not been started up yet. The temperature increase around the heated corrosion experiment is not yet sufficient to allow any reliable calculations and the specific experimental devices to be developed (e.g. heated dilatometers) are still under construction.

In order to get information about the stress field in zones only slightly affected by the excavation three hydraulic systems have been installed in the clay mass by way of 15 m long drillholes ; the influence of the grouting material confirmed to be very important in the assessments.

A geophysical survey is running for the detection of natural and/or induced fractures in clay ; after a reference borehole logging campaign, a long term seismic survey has been launched in the vicinity of one of the corrosion test tubes at temperature.

Three long term dilatometer tests are running satisfactorily but the installation of two complementary devices, equipped with heating elements, is postponed to next year.

In the frame of the study to be performed by ISMES, on the thermo-mechanical properties of Boom clay, samples were shipped to Bergamo in March. Laboratory experiments and determinations on Boom clay, undertaken by ISMES are in progress and results are evaluated in close collaboration.

Progress and results

1. Stress measurements in non-frozen clay (B.1.1)

The first three Glötzl cell cylinders (pre-orientated cells grouted in a cylindrical form) were emplaced in boreholes to get information on the stress field far from the underground laboratory. The results obtained and recorded in similar experimental conditions can be compared satisfactorily (rather isotropic stress field). The pressure level however remains, even after more than 2 years, relatively low which illustrates the time dependant behaviour of the clay (influence of a disturbance previously caused by the drilling to emplace the cells).

As an example the recorded values of the measuring system SMA (ring 9, grout modulus of 250 MPa) consisting of 4 total pressure cells and 1 pore water pressure cell are shown on figure 1 : the pressure decrease observed on the diagramme during the last weeks gives an idea of the sensitivity of the measuring device to monitor pressures changes ; these changes were caused by the drilling of a hole (cased immediately) in the vicinity of the measuring system.

Improvement of the physical characteristics of the grout (higher deformation modulus) has more beneficial influence on the pressure build-up than on the pressure level. In view of reproducing by numerical simulation the pressures recorded and to perform sensitivity analysis in order to assess the influence of several experimental parameters, a finite element approach has been launched by way of the 2D-linear elastic computer code EFINISAR, installed on the CEN/SCK mainframe. Other elasto-plastic simulations performed by our geotechnical consultant TRACTEBEL confirm our first deductions.

2. Fracturing of clay (B.1.2)

Observation of clay fracturation by optical means rapidly appeared to be inadequate due to the quick convergence of the wall of unlined holes and due to the limitations in dimension and the remote illumination.

The detection of micro-fracturation, natural or induced by temperature, has been launched by a geophysical survey from observation boreholes (single hole and cross-hole technique). A clay volume including the three vertical corrosion test tubes operated in the Hades-URL was compared with another volume of clay in an intact clay portion. The survey is performed by the "Laboratoire de géologie de l'ingénieur et de prospection géophysique" (ULg) in collaboration with the "Laboratoire du génie civil" (UCL). The classical borehole logging (caliper, gamma-ray, gamma-gamma, neutron-neutron, sonic) was completed by a seismic and resistivity survey.

The location of these experiments is shown on figure 2. The short term measurements developed for the "heated" section were between rings 3 and 8 and the "intact" zone is situated between rings 8 and 15. In both of zones, the previously frozen zone (3 to 4 m around the gallery) is clearly enhanced. When considering a more detailed examination of the results, a disturbance at the level of the heating elements (ring 3) can be detected in spite of the low temperature gradients now involved (the corrosion test tubes had to be disconnected temporarily). Confirmation has to be brought by long term investigations launched now in combination and along the restart of the heating of vertical corrosion tubes. The sensors (geophones for seismic and electrodes for resistivity measurements) were definitively locked in the PVC casing of two earlier exploration holes in the heated section.

The rock quality factor (Q) is one of the parameters mentioned on figure 2. The reciprocal of Q is a measure of the energy fraction absorbed by a non elastic medium when a wave is propagating through it ; it is of course an indication of the mechanical characteristics of the ground.

In the framework of the ME2I/ANDRA research programme concerning the study of clay behaviour around a heat source by way of spectral analysis, four holes have been drilled and cased for lodging the instrumentation around the location of the next future corrosion tube (ring 7). Zero measurements are scheduled for beginning 1989.

3. Long term dilatometric experiments (B.1.3)

The three long term dilatometer experiments, considered in the characterization programme of ANDRA (application of the convergence/confinement theory), are running satisfactorily. For the two experiments performed at a constant pressure, the stress levels now in operation are ranging from 2.2 to 2.5 MPa.

For the experiment performed at constant volumetric deformation, the third volume increment has been set and November 1988. The results of these experiments are detailed by ANDRA in their corresponding contribution.

The emplacement of the "heated" dilatometers, intended to perform similar experiments at higher temperature, in view of simulating at small scale the mechanical behaviour of clay in a thermal field around heat emitting wastes is scheduled to start after March 1989. Other experimental conditions (location, dimension, distance from the gallery) will remain comparable to the experiments already running at clay body temperature.

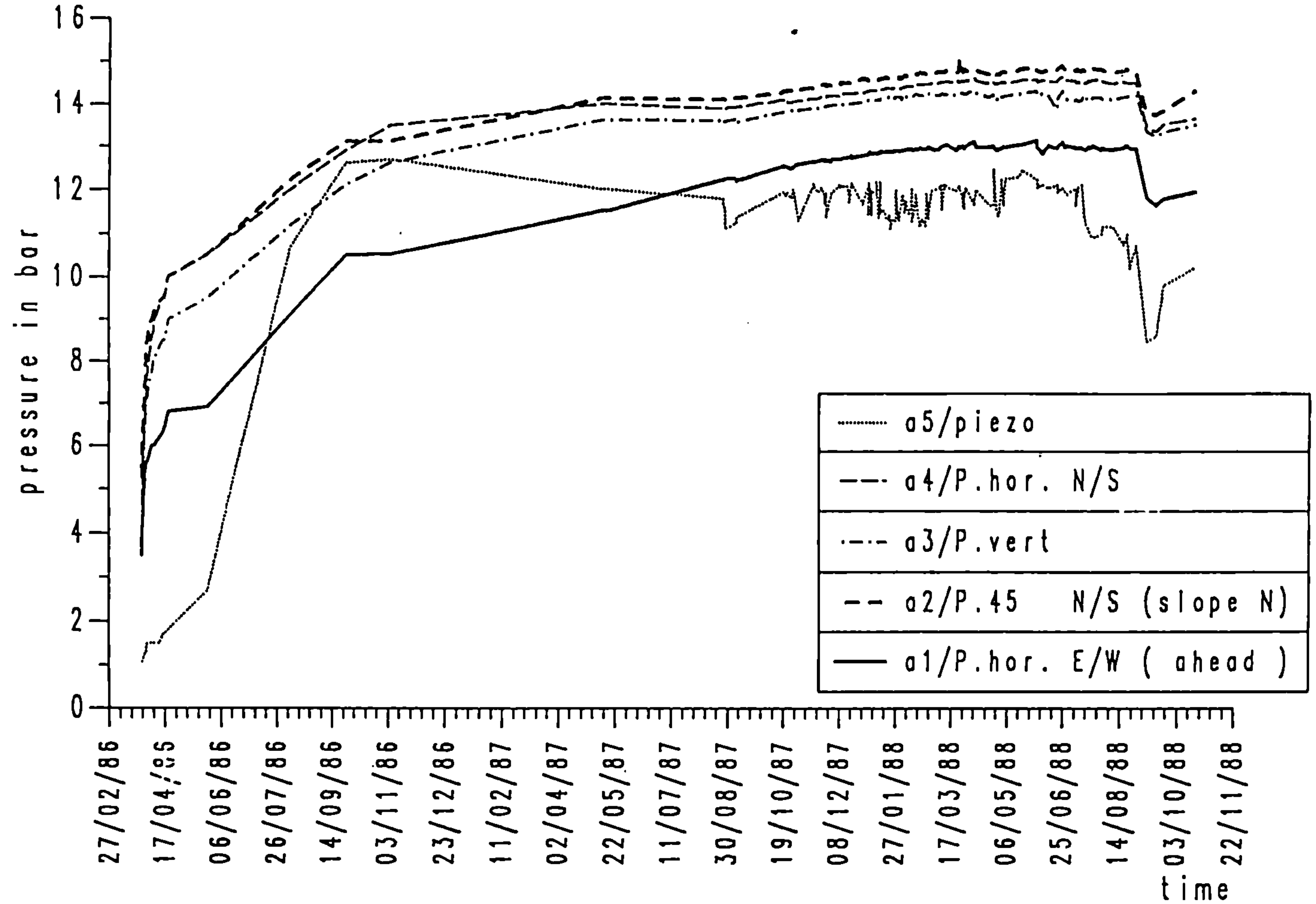
PUBLICATIONS

NEERDAEL B., DE BRUYN D.
Geomechanical research in the underground laboratory at Mol
CEC Contractor's Meeting on Geomechanics of clays for radioactive waste disposal, Brussels, 1-2 December 1988.

NEERDAEL B., BONNE A.
Ongoing research on geological disposal of radioactive waste in the Boom clay layer - last in situ developments
Third Meeting of ISAG, Washington DC, 2-4 May, 1988

FIGURE 1.

SMA - ring 9 - March 1986
corrected pressure values
27/10/88



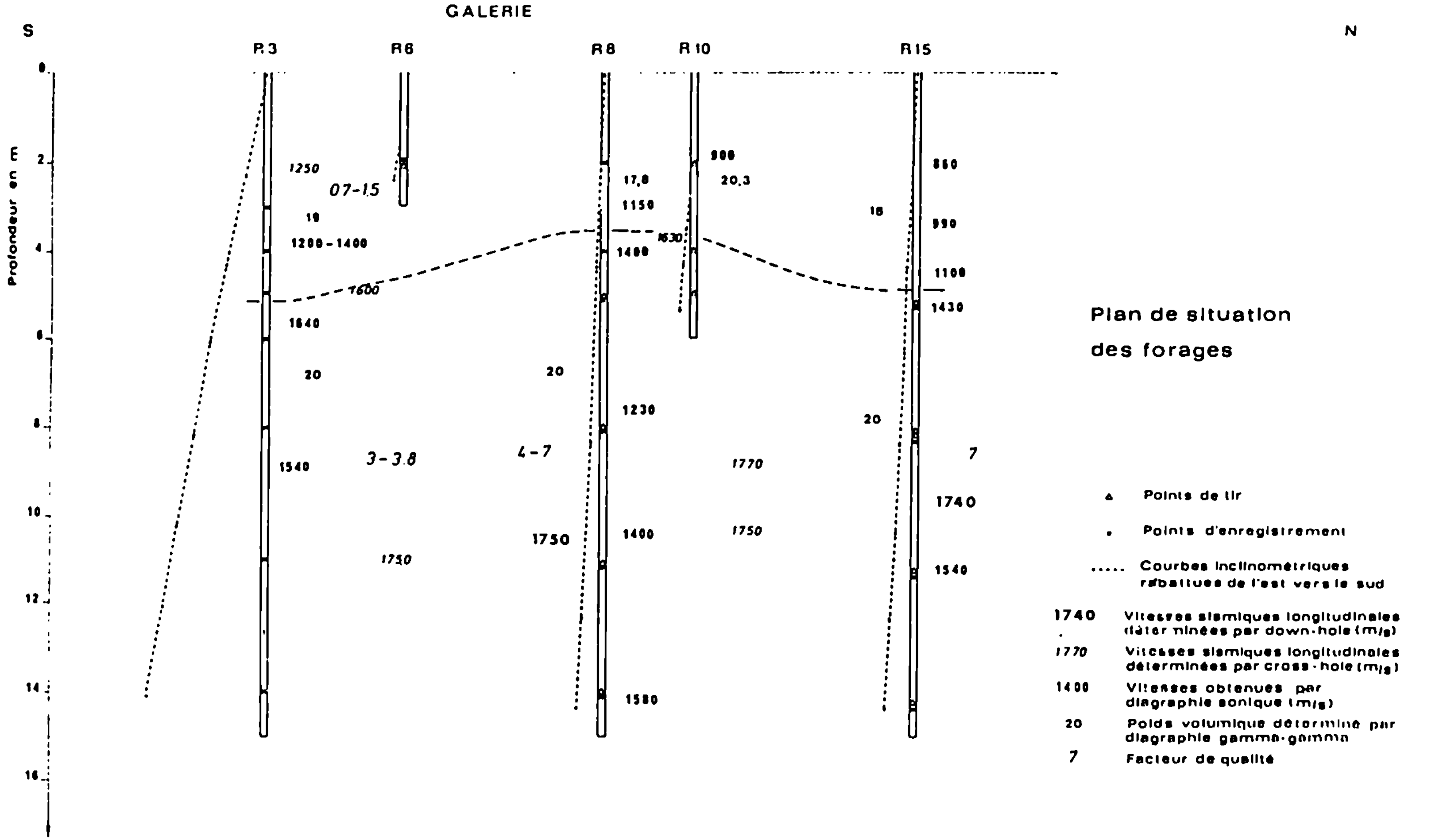


Figure 2.

0 5m

CEN 883

CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

Détection de la microfracturation
de l'argile de Boom

Doc. 1

A THERMO-MECHANICAL BEHAVIOUR OF BOOM CLAY.

Contractor: Ismes S.p.A.- Viale Giulio Cesare, 29 -24100 BERGAMO ITALY
Contract n.: FI1W/0150
Duration of contract: 29 months, from 1.11.1988 to 1.2.1989
Period covered: 1.11.1988 to 1.1.1989
Project Leaders: Dr. A. Peano, Ing. G.Baldi.

A. OBJECTIVES AND SCOPE

The understanding of thermal effects due to decay heat like pore pressure rise, change in mechanical properties and in the hydraulic field under high lithostatic pressures is fundamental in the evaluation of safety and in the design of nuclear waste repositories in clay.

For this purpose a thermo hydro-mechanical model has been developed in ISMES. Its suitability in reproducing the thermo mechanical response of the samples tested under different loading conditions has been positively verified with experimental tests performed on the HITEP triaxial cell operating in ISMES laboratories on various types of stiff clays, either reconstituted and undisturbed.

Furthemore, the interpretation procedures of thermal tests showed that for some of the clays tested the Campanella an Mitchell procedure can lead to inconsistent results.

In order to study possible reasons for this inconsistency, first thermal expansion coefficient values will be studied referring to the conditions of water in stiff clays, which are suspected to affect significantly its thermal-behaviour. This study will be conducted with AECL of Canada.

The results of this research could affect also the predicted undrained response of clay in terms of pore pressure rise.

In the meantime a better understanding of thermo-mechanical Boom clay behaviour is foreseen in order to improve the model.

Thermo-mechanical effects on clay due to realistic disposal geometries will be then studied with this model with the cooperation of SCK/CEN.

Similar studies are planned also for sand/betonite-mixtures, widely adopted for backfilling.

B.WORK PROGRAMME

1. Microstructural studies
2. Sampling of the Boom clay in the Mol site.
3. Experimental tests in the HITEP apparatus on natural and artificial clay soils.
4. Identification of material parameters on the basis of experiments on Boom clay.
5. Introduction of the options expected from activiy 1 in the mathematical model.
6. Check on the results of the laboratory tests using the above model.
7. Definiton of the boundary value problems to be treated on the basis of prospective in-situ experiments and/or typical disposal technology at Mol site.
8. Simulation of clay mass behaviour with reference to the problems envisaged in activity 7.
9. Evaluation of simulation results.

C. PROGRESS OF WORK AND OBTAINED RESULTS.

State of advancement.
Theoretical studies on microstructure of hard clays have been undertaken in order to assess the role of adsorbed pore water layers in clay response to nuclear waste heat. A constitutive model for thermal expansion coefficient of pore water was proposed together with a mathematical framework to account for adsorbed water presence in the theory of mixtures of porous deformable continua.

Laboratory tests at ambient and elevated tempoerature are in progress for specification of the thermo-mechanical model of clay skeleton response developed at ISMES to Boom clay. Activities include improvement of testing devices.

Experiments aimed at supporting the modeling of thermal water expansion coefficient of pore water for Boom clay have been included in a parallel research performed by AECL on the behaviour of hard clays.

PROGRESS AND RESULTS

1. Microstructural studies
A phenomenological relationship[1] for thermal expansion coefficient of interstitial water based on the results obtained in isotropic heating experiments on the HITEP apparatus[3] on reconstituted Pontida silty clay at high stresses and temperature has been developed at ISMES.

It considers not only temperature dependence, as usual for pure water, but also bonding pressure dependence. Predicted values of thermal expansion coefficient are considerably lesser when the high pressures occurring in the adsorbed water layer act.

Strains calculated following this relationship agreed well with experimental strains obtained from the axial strains.

A lower water expansion coefficient in hard clays, even not very active, is likely to be present due to the state of interstitial water.

To get experimental evidence with other devices, Boom clay and Pontida Silty clay were included as test materials in a research performed by WNRE of Atomic Energy Board of Canada Ltd. on microstructural properties of clays.

Estimates on quantity of free, adsorbed and bound water present in a clay soil will be made with the Pressure Plate/Pressure Membrane apparatus, in wich moisture content-capillary suction equilibria are measured. This will assist in the analysis of measurements of thermal expansion and associated porefluid expulsions from samples in tests on the prototype 1- Dimensional High Temperature High Pressure installed at AECL. Tests on Illitic and on Pontida clay samples are in progress.

The 1-D cell was ready for tests by late October 1988. The first heating test was made employing pure water as testing material: good prediction of water expansion values compared with the theoretical ones was observed. Heating tests on Silica Sand, Sodium Bentonite mixed with Silica Sand are in progress. Tests on Crushed Illitic Shale, Pontida Silty and Boom clay will follow. The choice of these materials is justified by the need to properly assess the influences of surface forces/clay mineral type and soil structure on thermal expansion properties of clay.

2. Sampling of the Boom Clay in the Mol Site.
25 Boom clay samples have been provided by SCK/CEN in early spring 1988,

obtained in 1987 during the construction of a test drift gallery. 15 samples are packed in tin boxes and 10 in wooden boxes.

3. Test on Boom Clay

Tests on Boom clay are in progress at ISMES using conventional geotechnical devices and the HITEP apparatus[3].

This last has been provided with drainage at the top and the bottom and with high entry value porous stones. For thermal tests, water expansion calibration in the supply circuit has been improved and tests were performed to verify membrane permeability at high temperature in presence of different types of cell fluids.

Epichloreidrine and Bromide Buthile membrane performance at 100°C and under 2MPa of pressure gradient was studied in HITEP employing an inox steel sample and water, castor oil, silicon oil as cell fluids. It resulted a considerably higher permeability of the first blend compared to the other. Fluid flow through the membranes was opposite to that of pressure gradient in the case of silicon an castor oil: this was thought as osmotic effect. Further similar tests on a new Bromide-Buthile blend membranes are planned in February 1989.

Classification and mechanical tests have been performed on Boom clay sample R116-W112.

Measured clay content and plasticity index are greater than those measured for Boom clay at 240 m depth tested at ISMES during CEC research contract 380.85.7.1. (S). Anyway they lay within the experimental range of variability for this clay.

X-ray diffractometry on random powder and on the $\leq 2\ \mu m$ fraction has been performed to characterize the mineralogy of clay fraction, togheter with DTA,TG and DTG analyses. The types of clay and interlayer minerals (chlorite, smectite) found in Boom clay lead to consider this material as potentially expansive.

From stress and strain controlled oedometer tests higher compressibility was found with respect to the clay at 240m depth. Preliminary evaluation of the results seems to confirm that maximum consolidation vertical stress is around 5MPa and K_0 value around 0.7.

Two triaxial drained test in the range of high OCR have been performed (0.8 MPa and 1.7 MPa initial isotropic stress). These tests are being used to mechanically finalize the constitutive model of Boom clay. The stress paths were characterized by loading-unloading cycles. Results showed a marked volumetric strain recovery which was supected to be due to swelling.

A triaxial undrained test at high OCR has been performed for comparison with one analogous test performed on Boom clay at 240 m depth: the new material revealed more compressible and with lesser residual resistance.

The first test on HITEP consisted in isotropic loading-unloading cycles in drained conditions, from 2 to 8 MPa of effective pressure under 2MPa of back pressure. Remarkable strain recovery has been measured again during unloading.

4. Mathematical modeling

Two main activities were forseen in this research to obtain a rigorous mathematical model for use in a finite element general code:

1- constitutive modeling for Boom clay skeleton thermo-mechanical behavior (a) and for interstitial water thermal expansion (b);
2- accounting for consistency of constitutive mathematical expressions

from thermo-dynamics principles and study for possible ways of accounting for the absorbed interstitial water in the framework of the porous deformable continua theory.

As for point 1 (a), clay response at low and intermediate OCR was seen worth to be further studied. FE calculations showed how this range is most probably that ruling the yielding response of the heated clay around a waste container.

From a preliminary study on the set of results obtained from laboratory tests on Boom clay at 240 m depth it resulted the need for a specification of the yield limit shape and of flow rule.

The results of the mechanical test compaign in progress will outline the main features of this specification.

As for point 1(b), a possibility is that of using the constitutive law mentioned in Sec.1 if validated by AECL experiments, or to introduce alternative options coming from the parallel research with AECL.

As for point 2 the ruling equations of the theory of mixtures were studied[2] and have been specified for the case of saturated plastic clays under thermo-mechanical loadings, accounting for the presence of adsorbed water in clay pores.

Two principle ways of approach have been selected: mass transfer (MT) and average water (AW) approach. Here clay is considered as a two-phase material in which adsorbed water is considered as a part of the solid fraction (MT approach) and of the water fraction (AW). Consistent definition of parameters and of mass exchange during thermal loading has been studied for these two cases. Implication on stress repartition has been also specified.

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2. HUECKEL, T. (1988). Vincoli da rispettare nella modellazione matematica del comportamento termomeccanico del sistema acqua-argilla , (in english) ISMES Report DMM-4734.
3. TASSONI, E.; HUECKEL, T. and BORSETTO, M. et al. (1985). Comportamento termico dei materiali argillosi (in italian). EUR Report 10190IT,37-127.

STUDY OF THE CLAY BEHAVIOUR AROUND A HEAT SOURCE

Contractor: ME2i, 8 Rue Eugène Oudiné, 75013 PARIS
Contract No: FI1W.0152.F
Duration of contract: 1 December 1987 - 31 December 1989
Period covered: 1 January 1988 - 31 December 1988
Project leader: P. De Sloovere

Progress report not yet available.

FRACTURE MECHANICS FOR HARD ROCK

Contractor: J. Gramberg, The Netherlands
Contract No: FI1W/0153/NL
Duration of contract: July 1987 - December 1988
Period covered: January 1988 - December 1988
Project leaders: J. Gramberg

Progress report not yet available.

Experimental study of the mechanical behaviour of argillaceous rock

Contractor: Commissariat à l'Energie Atomique, Fontenay-aux-Roses, France
Contract N°: FI1W/0163.F
Duration of Contract: October 1987 - December 1989
Period covered: October 1987 to December 1988
Project Leader: J.Y. BOISSON

A. OBJECTIVES AND SCOPE

Changes in the mechanical behaviour of clays according to the temperature are not yet well known. If, for the short term behaviour studies, one can admit that there is no drainage effect and, that, a temperature rise leads, at the short term, to a clay volume increase, this cannot be stated concerning the long term, where the drainage effect must be taken into account.

Recent studies have shown that, thanks to their texture rearrangement, saturated remolded porous soils, replaced in a normal consolidated state, exhibit, in the long term, a volume decrease, with, as consequences, settling effects and crackings.

It is possible that the long term response of the clays at such thermal prompting is a fonction of their over consolidation degree : normal consolidated clays will decrease, in fact, in volume under thermal sollicitation but this could be different for the over consolidated clays. The aim of this study is to bring clear and quantitative experimental answers to these questions both with a theoretical interpretation of these phenomena.

This research will be performed with the scientific support of the Centre de Géologie de l'Ingénieur (ARMINES-Ecole des Mines de Paris).

B. WORK PROGRAMME

2.1. Choice of the sites, and sampling of clay

The selection will be made first considering that the sites answer to some textural granulometric criteria (clays silts) and mechanical criteria (normally or over consolidated clays).

After that, we will take these materials under predetermined conditions to avoid remolding and overconsolidation due to surface dessication.

We will determine then, in a precise way, their initial overconsolidation degree and their texture, considering that these two points are essential for this study of the behaviour of clays towards thermal sollicitations.

2.2. Responses study of the selected material towards the thermal mechanical sollicitations

These responses are mainly textural rearrangements which will be studied in the laboratory by precise texture identifications and by comparisons between initial and final state.

2.2.1. Basic petrographic, mineralogic and textural identification

Different procedures will be used : X ray diffractometry analyses, adsorption tests with methylene blue, Atterberg limits, porosimetry, observations with scanning electron microscope, and permeabilities.

2.2.2. Determination of the overconsolidation degree.

2.2.3. Control thanks to blank tests of the thermal behaviour of the experimental apparatus used.

The aim will be to well separe, in the experimental results, the information concerning the clay geomechanical behaviour, from artefacts due to the thermal behaviour of the tests apparatus itself.

2.2.4. Creep tests in oedometer cells and permeability measurements.

An axial backpressure will be applied so as to be able to scan the temperature range between 20°C and 120°C without pore water loss. The stress will be equivalent to the one in situ, and will not be less than 0.01 MPa.

In case of swelling during the sample saturation, the applied stress will be equivalent to the swelling pressure. The thermal prompting applied to each selected clay sample will correspond to the 4 following temperatures : 20, 50, 80 and 120°C. Different temperature rises will be applied, favouring the exploration of parameters linked to the thermal prompting and to the creep :

a) direct temperature rise (creep study at mid term during 3 months, tests at the 4 mentionned temperatures).
b) direct temperature rise, (creep study at long term during 9 months, tests at 20° and 80°C).
c) progressive temperature rise at 20° and 120°C (for each temperature level, creep study at mid term for 3 months).
d) progressive temperature rise and then progressive decrease, creep study during 3 months (for each temperature level) : one test from 20° to 50°C and then from 50°C to 20°C ; one test from 20°C to 80°C and then from 80°C to 20°C ; one test from 20°C to 120°C and then from 120°C to 20°C.

A certain number of these tests will be repeated (two or three times) so as to ascertain the reproductibility of the results.

2.2.5 Textural study

This study will be done by applying the mercury porosimetry tests, scanning electron microscope, permeability tests on each sample (initial and final state). The compared analysis of the whole results will certainly give a contribution to the thermal mechanical behaviour knowledge related to the texture and overconsolidation degree.

The basic equipement for the textural study and for the mineralogical, petrological identifications and classical geotechnical tests is available. Nevertheless, for the thermomechanical tests it will be necessary to adapt existing devices.

C. <u>PROGRESS OF WORK AND OBTAINED RESULTS</u>

2.1. <u>Choice of the sites, and sampling of clay</u>

A bibliography study and numerous contacts with clay quarries people have led us to a pre-selection of several sites where sampling has been done. Necessary precautions have been taken to obtain representative samples, avoiding any kind of remolding due to the sampling or any dessication phenomena.

Seven samples have been taken in quarries :

- mixed red an white clays (Barremian) from <u>Saint-Germain de Fly</u> - Pays de Bray ;

- white clays (Sparnacian) from quarries of Fontaines and Fontbouillant - Charentes ;
- clays (Sparnacian) from Limay-Vexin at two levels :
  . plastic mixed clay : Limay 1
  . black clay (5 meters above the first one) : Limay 2.
- clays (Sparnacian) from Provins-Brie, coming from two sites : Montbron and Saint Genest
- green clays (Sannoisian), from South-East Lagny - Brie taken from a core.

The complete identification of each clay is presented in tables I to IV. The selection of clay samples which will be used for the study, has been done on the following bases : mineralogical criteria (type of clay minerals), granulometry (silty fraction percentage) and geomechanical criteria (over consolidation ratio ; plasticity ; natural void ratio which may determine the creep extent ; swelling potential, and natural sucking which may generate a remolding due to the swelling).

2.2. Responses study of the selected material towards the thermal mechanical sollicitations

2.2.1 - 2.2.2. Choice and basic identification - Overconsolidation degree

Four samples have been selected :
- green clay from Lagny : important illitic fraction, relatively high plasticity, and high void ratio ;
- clay from Saint-Genest : same characteristics but with an important kaolinitic fraction ;
- plastic clay from Limay 1 : high plasticity ;
- black clay from Limay 2 : highest silty fraction, and presence of organic matters which may be sensitive to the temperature.

These four clays are all overconsolidated but at different level : Lagny (low OCR), and in the order of importance : Limay 1, Limay 2 and Saint-Genest.

The clay from Saint-Genest will also be studied in remolded state : the remolding is obtained by free swelling during saturation.

2.2.3. Control tests of the experimental apparatus

A specific prototype for the creep tests has been designed (Fig.1). It is an oedometric cell with back pressure, but with specific layings out due to the temperatures range, the thermostatisation with oil immersion, and long test periods :
- the lateral friction of the piston is minimised thanks to a waterproof "BELLOFRAM" membrane between piston and cell ;
- the thermal dilatation is minimised using INVAR for the parts of the apparatus involved in vertical displacements measurements ;
- the vertical stress is applied with a pneumatic jack (max : 2 MPa on the sample) ;
- the cell is submerged in a silicon oil, with low viscosity and chemically inert ;
- a back pressure of 0,3 MPa is imposed to avoid water vaporisation of the pore water sample ;
- saturation sample at the beginning of a test, and permeability measurements are made via a pressure-volume automatic controller ;

- the axial deformation is measured through an electronic sensor of 1 μm sensibility.

Schematic view of the set up is given (Fig.2). There are two benches of eight cells each.

Qualification tests have been run so as to be sure of the reliability of the system and to determine the necessary corrections to be used, taking into account temperature variations on the whole device. Calibration tests have been made for this last point and it is shown that thermal equilibrium of the device is reached after several hours.

2.2.4 - 2.2.5. Example of results for a preliminary test

A preliminary experimental study has been done at room temperature (20°C), on the consolidation and creep behaviour in a standard oedometric cell. The case of the Saint-Genest clay is considered.

Samples are saturated for one week, preventing or not the swelling, and submitted during one month to a vertical stress equivalent at least to the pre-consolidation pressure. Swelling characteristics are analysed. Textural analysis are made before and after tests.

The swelling of the Saint-Genest clay (under 5 kPa, after 5 days) during the saturation phase gives a swelling curve (Fig.3) which can be represented by an hyperbolic law :

$$\varepsilon = \frac{dh}{ho} = G \frac{t}{B+t}$$

with a final swelling rate G = 4,94 % (after theoritical infinite time, and a time of half swelling B = 300 min.).

Our sample is then submitted to a vertical stress of 405 kPa during one month. The results of the test give the curve (Fig.4), which can be compared to the theoritical creep curve calculated from FELIX (1980) using the equation :

$$C(t) = h_f \left(1 - e^{-\alpha t^{\beta}}\right)$$

with final settlement h_f for an arbitrary duration of 100 years.

M.E.B examination of the intact St Genest-clay sample (Fig.6 - photos 1 and 2) reveals a relatively compact texture of the turbulent type (after GRABOWSKA-OLSZEWSKA et al. 1984) with a certain anisotropy. The results of the mercury porosimetry tests (Fig.5a) indicate a very fine porosity with the most representative threshold radius of 7.5 10^{-3} μm and an ultra porosity (< 0,1 μm) equal to 77 % of the total porosity.

Swelling leads to a significant increase of the total porosity, mainly due to the increase of the microporosity (0,1 à 10 μm) and the appearance of a new pore family with mean access pore radius of 0,4 μm (Fig.5b). This fact can be related to the microfissure openings observed with M.E.B (See Photo 3). The trapped porosity increases too.

The settlement reached at the end of the oedometric test seems to induce the disappearance of the micro fissures which were observed with the M.E.B (See Phote 4), and a decrease of the microporosity with a mean access pore radius of 0,4 μm (Fig.5c). The ultra porosity does not seem to be affected. Nevertheless the final texture appears to be slightly different from the intact one, with higher percentages of microporosity (0,1 - 10 μm) and macroporosity (> 10 μm).

Références

1. FELIX B., 1980. Le fluage et la consolidation unidimensionnelle des sols argileux. Rapport de recherche des Laboratoires des Ponts et Chaussées n° 94 (1980), 176 p.
2. GRABOWSKA-OLSZWKA B., OSIPOV V., SOKOLOV V., 1984. Atlas of the microstructure of clay soils. Panstowe Wydawnictwo Naukowe, Warszawa.

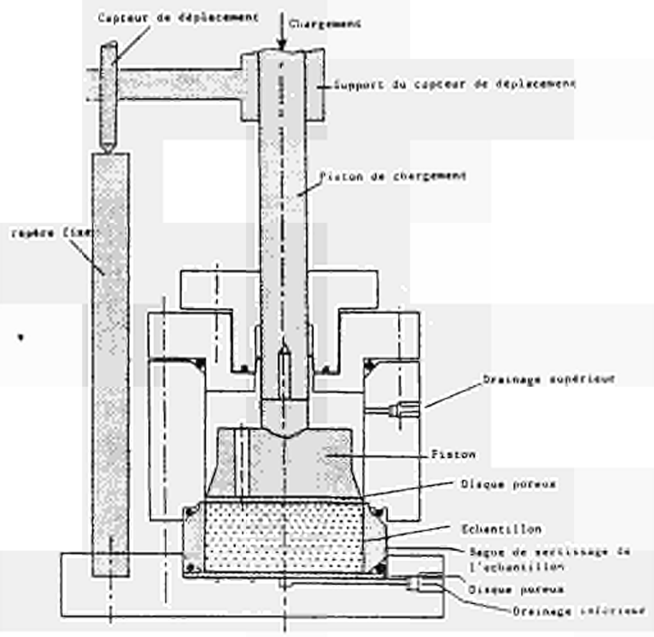


Fig.1 : Schéma de principe de la cellule oedomètrique

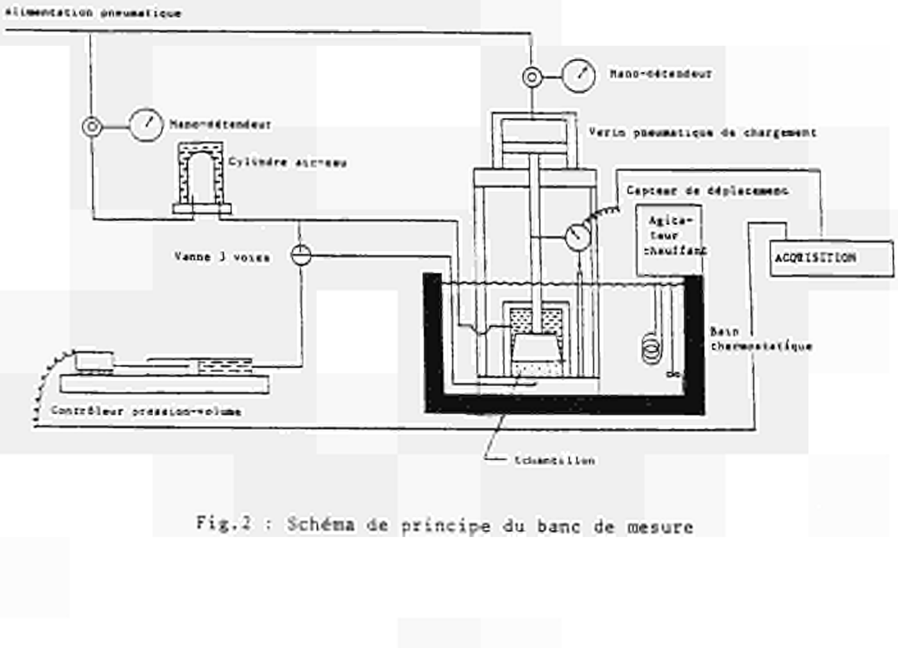


Fig.2 : Schéma de principe du banc de mesure

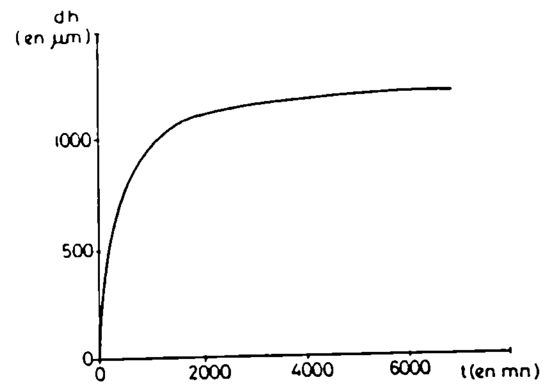


Fig.3 : Courbe de gonflement de l'argile de St Genest

$$\varepsilon = \frac{dh}{24000} = 0.0494 \frac{t}{300+t}$$

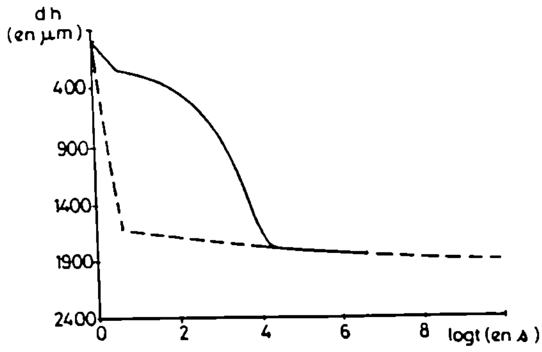


Fig.4 : Courbe de tassement de l'argile de St Genest

En pointillé, la courbe de fluage : $dh = 1951\ (1-\exp(-1.70\ t^{0.039}))$

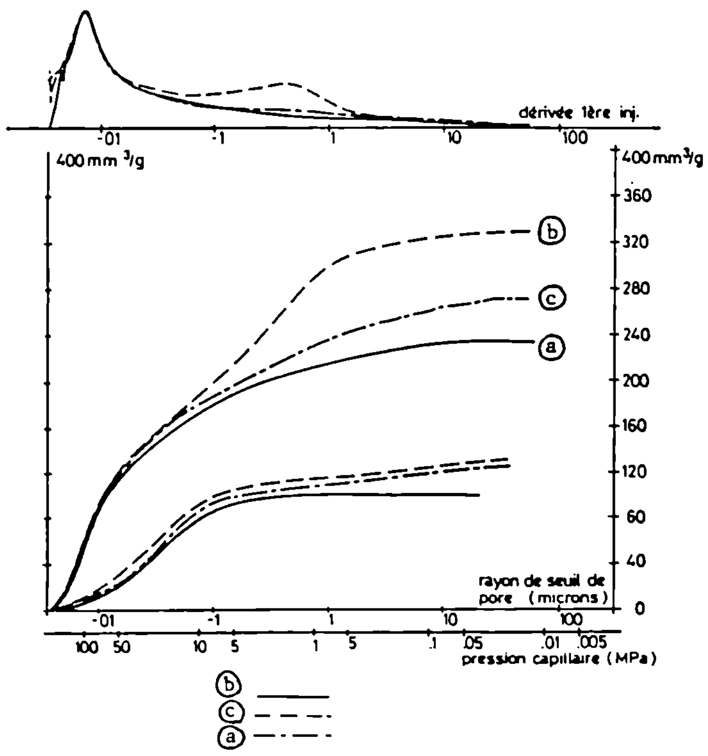


Fig.5 : Courbes porosimètriques de l'argile de St Genest intacte (a), gonflée (b), gonflée puis tassée (c)

10 µm

Photo 1 - argile intacte
(Parallèlement à la stratification)

10 µm

Photo 2 - argile intacte
(perpendiculairement à la stratification)

10 µm

Photo 3 - argile après gonflement

10 µm

Photo 4 - argile après gonflement
et tassement

Fig 6. : Observations au microscope électronique à balayage de l'argile de Saint Genest

SITE	Kaolinite	Illite	Smectite	Interstratifié Illite Smectite	Goetite	Quartz
St Germain de Fly	xxx	-	-	x	-	x
Fontaines	xxx	x	-	-	-	xx
Fontbouillaud	xxx	xx	-	x	-	-
Limay 1 (plastique bariolée)	xxx	-	-	xx	x	x
Limay 2 (noire)	xx	xx	xxx	-	-	x
Montbron	xxx	x	-	-	-	-
Saint Genest	xxx	-	-	-	x	x
Lagny	-	xxx	xxx	-	-	-

Tableau I : Composition minéralogique

xxx : forte proportion
xx : faible proportion
x : traces
- : absence

SITE	ρh g/cm³	ρd g/cm³	Wo %	Sr %	eo	Wl %	Ip %	Bleu g/100g	Ssp m²/g
St Germain de Fly	2.14	1.81	18	0.99	0.49	51	24	2.9	85
Fontaines	2.09	1.77	18	0.96	0.48	46	20	1.3	32
Fontbouillaud	1.80	1.31	38	0.99	1.01	60	22	1.7	37
Limay 1	1.89	1.43	32	0.98	0.87	130	84	14.1	316
Limay 2	1.99	1.66	20	0.87	0.63	74	40	11.3	376
Montbron	2.10	1.79	18	1	0.48	54	30	5.5	153
Saint Genest	1.88	1.44	31	0.98	0.82	91	49	5.4	118
Lagny	1.91	1.45	32	1	0.86	78	35	10.0	255

(Bleu : valeur de bleu ; Ssp : Surface spécifique totale)

Tableau II : Identification géotechnique

SITE	Sable fin (%)	Silt (%)	Argile (%)
St Germain de Fly	0	30	70
Fontaines	4	20	76
Fontbouillaud	1.5	10.5	88
Limay 1	0	7	93
Limay 2	2	33	65
Montbron	3	26	71
Saint Genest	0	4	96
Lagny	1	21	78

Tableau III : Granulométrie

Sable fin = > 64µm
Silt = 64µ à 2µm
Argile = < 2µm

SITE	$\sigma'g$ (kPa)	$\sigma'c$ (kPa)	Cc
St Germain de Fly	70	130	6.10^{-2}
Fontaines	40	100	$0,9.10^{-2}$
Fontbouillaud	40	200	1.10^{-2}
Limay 1	230	340	3.10^{-2}
Limay 2	30	85	12.10^{-2}
Montbron	100	210	3.10^{-2}
Saint Genest	200	350	8.10^{-2}
Lagny	20	70	13.10^{-2}

Tableau IV : Caractéristiques mecaniques

$\sigma'g$ = pression de gonflement
$\sigma'c$ = pression de préconsolidation
Cc = indice de compression

SITE	k (m/s) permeabilité	pF succion
St Germain de Fly	non déterminé	3,4
Fontaines	1.10^{-10}	4,3
Fontbouillaud	1.10^{-10}	4,1
Limay 1	5.10^{-10}	3,9
Limay 2	$3.7.10^{-11}$	-
Montbron	1.10^{-10}	3,9
Saint Genest	5.10^{-12}	4,0
Lagny	1.10^{-10}	3,7

Tableau V : Caractéristiques hydraulique et hydrique

DEFORMABILITY AND FAILURE BEHAVIOUR OF A SALIFEROUS FORMATION NEAR TO OPENINGS

Contractor: BRGM, BP. 6009, 45060 Orleans (F)
Contract No: FI1W/0206
Duration of contract: 1 August 1988 - 1 August 1990
Period covered: 1 August 1988 - 31 December 1988
Project leader: P. Peaudecerf

Progress report not yet available.

4.2. REPOSITORIES AND ENGINEERED BARRIERS

4.2.A. Repository design and disposal techniques

N.B. : No particular research contracts have been concluded on this subject

4.2.B. Engineered barriers

4.2.B.1 HLW container development (COMPAS)

ASSESSMENT OF STRUCTURAL PERFORMANCE OF HLW CONTAINERS (COMPAS)

Contractor :	Ove Arup and Partners, London, England
Contract No. :	FI1W/0111
Duration of Contract :	April '87 - Dec '89
Period Covered :	Jan '88 - Dec '88
Project Leaders :	J Miles; S Hendry

A. OBJECTIVES AND SCOPE

The COMPAS project has been designed to look at the mechanical performance of those containers which will be used for overpacking and disposal of high level radioactive waste. The following partners are also participating in the project: CEA (F), Equipos Nucleares (E), NAGRA (CH), PSI (CH), SCK/CEN (B) and STEAG (D). By agreement of the Partners it will be restricted to the examination of containers for vitrified waste rather than containers for the direct disposal of spent fuel. The project is not concerned with the production of a specific design for licensing purpose; it is only intended to investigate the characteristics of representative designs.

The objectives of the COMPAS project are to look at the mechanical performance of these containers and to develop an understanding of how they will behave when subject to the most extreme conditions which can be foreseen in realistic disposal scenarios.

In order to predict the ultimate mechanical performance of the disposal containers it will be necessary to use computer aided modelling techniques. The early part of the COMPAS project therefore includes a considerable amount of computational work which is aimed at developing confidence in the use of these techniques.

B. WORK PROGRAMME

During the year the contract was extended to include a more comprehensive programme of testwork and associated computing. The revised project plan now comprises the following activities:-

(i) Directory of Computer Codes
(ii) Containment Concepts
(iii) First Benchmark Exercise
(iv) Preliminary Ring Tests
(v) Intermediate Testwork
(vi) Advanced Testwork
(vii) Prediction of Ultimate Performance.

C. PROGRESS OF WORK AND RESULTS OBTAINED

State of Advancement

The second year of the project has seen an increasing emphasis on experimental work. The Directory of Computer Codes and the report on the First Benchmark Exercise, which were prepared during 1987, have been submitted to the CEC for publication.

The revised project plan now includes three distinct phases of experimental testwork and associated computational calculations. The Preliminary Testwork has been completed, the Intermediate Testwork is underway, and the Advanced Testwork programme is being prepared.

The project is on schedule and should be completed by the end of December 1989.

PROGRESS OF WORK

(i) Directory of Computer Codes

During 1987, various finite element (and finite difference) codes used by the project partners, along with a number of commercially available codes, were investigated to establish a directory of codes suitable for the stress analysis of containers for high level nuclear waste.

The directory included information on each code's supplier, its distribution and availability, technical background and pre- and post-processing capabilities. This directory has been submitted to the CEC for publication.

(ii) Containment Concepts

This work, performed during 1987, confirmed the state of development of repository plans for the various project partners.

(iii) First Benchmark Exercise

The purpose of this study was to set up a series of problems against which the partners could test their favoured codes. Two different container concepts were investigated during this exercise - a thick-walled 'corrosion tolerant' container and a thin-walled 'corrosion resistant' container. For both containers, the calculations performed investigated the effect of an external pressure on an annular slice through the container. Both geometrical and material non-linearities were considered. A total of 18 problems was specified.

All of the partners took part in the exercise, although not all of them attempted every problem. During 1988 the report on this work was submitted to the CEC for publication under the title "Stress Analysis of HLW containers". Figure 1 shows a typical set of results - for the elastic-plastic analysis of a non-uniformly corroded thick-walled container.

(iv) Preliminary Ring Tests

The first computer benchmarking exercise confirmed that several of the codes used were capable of calculations involving large deflections, in problems typical of those experienced by HLW containers. However no experimental information was available to assess the accuracy of the codes' predictions. To address this shortcoming the Preliminary Ring Test programme was set up. In these tests ring specimens (160mm diameter, 16mm wall thickness) were loaded transversely across a diameter using parallel plattens.

In addition to tests on standard ring specimens, a number of tests were carried out on rings with representative defects. These include rings with internal or external notches and rings with an external layer of weld deposit. Four different ring types were manufactured, and three tests on each type were performed. The project partners were invited to model these tests, and most of the partners attempted all the problems. Results were received during October and November 1988, and a report on this part of the project is being prepared.

(v) Intermediate Testwork

The Intermediate and Advanced Testwork programmes involve isostatic pressure tests on scale models of HLW containers. The intermediate tests look at simplified one-third scale models of thick-walled containers. By including several variations to the standard container, these tests give an opportunity to examine the effect of these variations on the performance of the container. The test programme investigates the effects of reduced wall thickness, non-uniform corrosion and different welding methods.

Eight containers were manufactured by Equipos Nucleares, Spain, during November and December 1988. The testwork will be performed at the Schlumberger test facility in Paris during January and February 1989. The containers will be subjected to a uniform external pressure of up to 100 MPa, and will be extensively instrumented to allow post-test comparisons with computational predictions by the project partners.

(vi) Advanced Testwork

Following on from the intermediate tests there will be a small number of tests on more realistic scale models of actual HLW containers. These containers will be as near as possible to an existing design, but there are restrictions on the cost, size and strength of the containers. In these tests features such as lifting pintles and end closures will be modelled in greater detail.

These Advanced Tests will take place in July, August and September 1989. They will be extensively instrumented and should give a great deal of information concerning the likely behaviour of real containers under disposal conditions.

(vii) Prediction of Ultimate Performance

This study is intended to predict the ultimate performance of the containers under extreme disposal conditions. It will draw on the conclusions from the experimental programme, especially the Advanced Testwork. By its nature it can only be attempted once the majority of the work in the later part of the project has been completed. However some preliminary hand calculations were performed at the beginning of the project in order to give a general appreciation of the problem. This preliminary work formed part of the planning studies for the experimental work mentioned above.

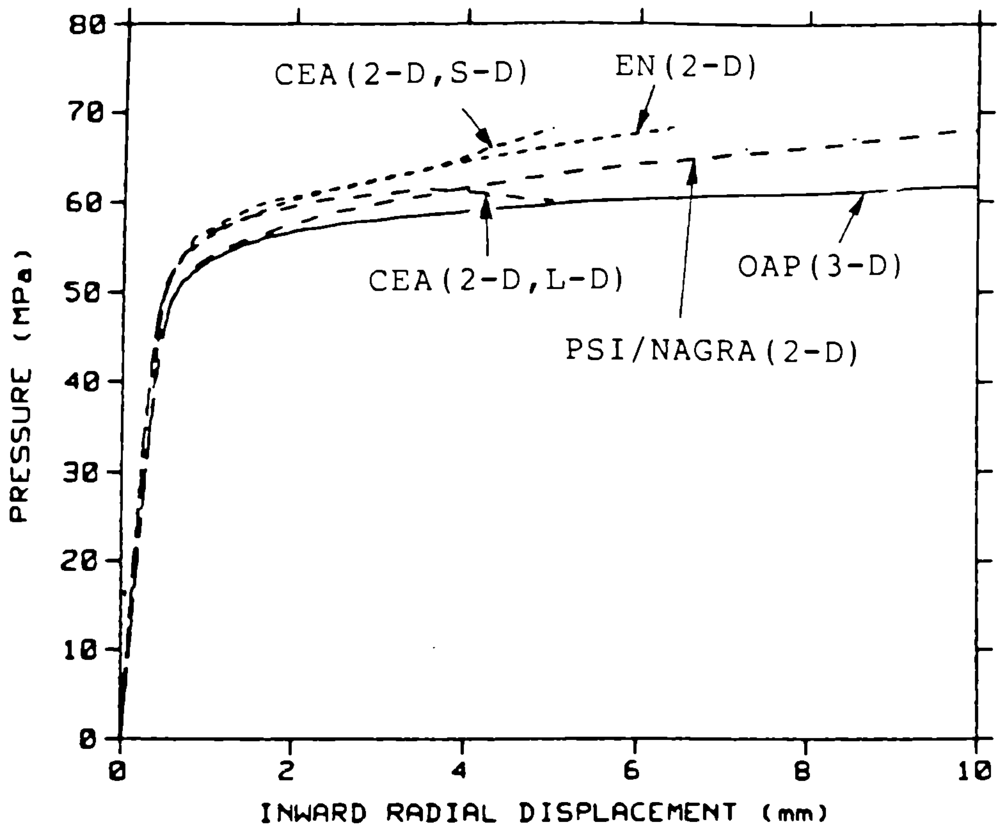


Figure 1 : Typical results from the First Benchmark Exercise

4.2.B.2. Backfilling and sealing of radioactive waste repositories

In-situ reduced scale backfill and heater experiment

Contractor : SCK/CEN, Mol, Belgium
Contract No : FI1W/0055/B
Duration of contract : October 1986 to December 1989
Period covered : January 1988 - December 1988
Project leader : A.A. Bonne, B. Neerdael

A. OBJECTIVES AND SCOPE

In 1974 SCK/CEN launched a R&D-programme concerning the possibilities for disposal of high-level solidified and alpha-bearing radioactive wastes in a continental stratiform clay formation (Boom clay) situated below its own site. Several specific investigations still need to be further undertaken in order to characterise more accurately the argillaceous formation in view of assessing its appropriateness for hosting radioactive waste as well from engineering point of view as for long term safety and performance evaluations. In support of these also further modelling efforts are required in order to improve and confirm our prediction capability.

This particular item aims at investigating in situ the performances of an argillaceous seal and its immediate surrounding clay in a thermal field.

The experiment is developed jointly by the CEA/DRDD and the CEN/SCK where the field test is taking place in the HADES-URL (Underground Research Laboratory).

B. WORK PROGRAMME

B.2.1. Investigations about materials and instrumentation
B.2.2. Integral reduced scale in situ experiment (Bacchus)
B.2.3. Validation of heat transfer and thermo-mechanical computer models

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The Bacchus experiment ("BACkfilling Control for High level wastes Underground Storage") has been designed in 1987 ; the experimental conditions prevailing at Mol have in particular led to the definition by the two parties involved (CEA/DRDD and CEN/SCK) of the composition of the materials to be used.

Emphasis is laid on the thermal and hydraulical transfers through a highly compacted material placed at the periphery of the heater and in the surrounding clay mass.

Much time was devoted this year to the choice and/or development of measuring devices including calibration and testing. Among the instrumentation now installed in both materials (temperature pressure, humidity), the measurement of water content has required the development of specific moisture sensors : thermal probes in compacted material and TDR sondes (Time Domain Reflectometry) in the surrounding clay.

The test cell was installed in a vertical access hole in clay 14 m underneath the gallery lining end November and sealed off in the clay mass by a plug of recompacted Boom clay.

After improvement of the contact of the test cell with the clay body and the restoration of the local conditions, the experiment is scheduled to start in the course of February 1989.

Progress and results

1. Investigations about materials and instrumentation (B.2.1)

Characterization of materials

The natural clay selected by CEA/DRDD (Ca-smectite) is mixed in a proportion of 50 % with sand (45 %) and graphite (5 %) in order to adapt respectively the swelling pressure (5 MPa) and the thermal conductivity (1.7 W/m° C) to the experimental conditions at Mol.

The characterization of the backfill material and the reused Boom clay has been made by CEA. An isostatic compaction level of 20 MPa has been chosen for final characterization and fabrication ; it corresponds to volume weights convenient for in situ conditions and does not compromise the mechanical behaviour of large cylindrical cores (D = 300 mm).

Instrumentation

The clay host instrumentation, in charge of CEN/SCK, has been installed by way of access drillholes prior to the installation of the test cell and located as illustrated by figure 1.

The measuring devices are located between 50 and 100 cm of the test cell, the minimal distance being related to the precision of drilling 15 m long holes and the maximal one is required for experimental purposes (minimal temperature evolution).

Have been placed in the surrounding clay (figure 2) :

- 18 temperature sensors (Pt100) by way of 2 boreholes ;
- 10 screen piezometers (2x5) by way of 2 boreholes ;
- 10 other Pt100 for each of the screen piezometers ;
- 8 hydraul earth pressure cells by way of 2 boreholes ;
- 2 pore water pressure cells coupled with the previous ones ;
- 8 humidity sensors (TDR probe) by way of 2 boreholes ;
- 8 other Pt100 for each of the humidity sensors ;
- 1 access tube for use of a gamma/neutron sonde (one hole).

The TDR method, based on the properties of electromagnetic waves, consists in measuring the transit time of waves along a probe in a soil. If the technique has been proved, the specificity of this application needed to design special bifilary probes where the two rods have to be pushed horizontally in the clay from a vertical access tube, 170 mm in diameter. Calibrations have proved that the reduced length of the rods (8.5 cm in this case with regard to 30 cm usually used) does not seriously affect the accuracy.

The instrumentation placed in the highly compacted backfill material (temperature, pressure, humidity) is described in the corresponding CEA progress report.

Complementary probes (pressure, temperature) were installed at the top of the seal plug (compacted Boom clay blocks) during the placement of the whole system in the hole.

2. Installation of the experiment (B.2.2)

Figure 2 gives the location and dimensions of the different components of the experimental set-up. The set-up includes the experimental cell itself (1.68 m high) including the buffer and the heater and the seal plug (1.47 m high) consisting in Boom clay blocks.

The experimental cell is composed of 5 circular blocks of compacted backfill material, 0.3 m high. The upper plug consists in 4 circular blocks of Boom clay, 0.36 m high. The cables connecting the command and measuring units in the URL with the instrumented and heated lower part of the cell are introduced in the central hoisting pipe.

From a 760 mm diameter access hole in the cast-iron lining, a 11 m long vertical hole was drilled in 450 mm diameter and lined by a steel casing (406/396 mm). From this level, the hole was deepened in smaller diameter (390 mm) over a distance of 3 m and the experimental cell (800 kg), 330 mm in diameter, lowered down the hole.

The drilling phase was particularly time consuming due to the necessity, to retrieve a caliper device abandoned in 1984 at the same location as the Bacchus test.

The heating elements (D = 60 mm, H = 1500 mm) are designed to perform the test at constant heat power between 300 and 500 W corresponding to a temperature increment of respectively 70 to 100 °C at the interface between the heater and the buffer after one year.

3. Results and interpretation (B.2.3)

The data and results obtained will allow the comparison and validation of thermomechanical and heat transfer computer codes. The temperature increment expected at the interface between the heater and the backfill has been calculated with the MPGST-heat transfer code recently validated by the reproduction of the temperature field recorded around the corrosion test tubes.

PUBLICATIONS

NEERDAEL B., VOET M.
In situ reduced scale backfilling and heater test
CEC Meeting on Backfilling and sealing, Paris, 18-20 January (1988)

NEERDAEL B., BONNE A.
Ongoing research on geological disposal of radioactive waste in the Boom clay layer - last in situ developments
Third Meeting of ISAG, Washington DC, 2-4 May (1988)

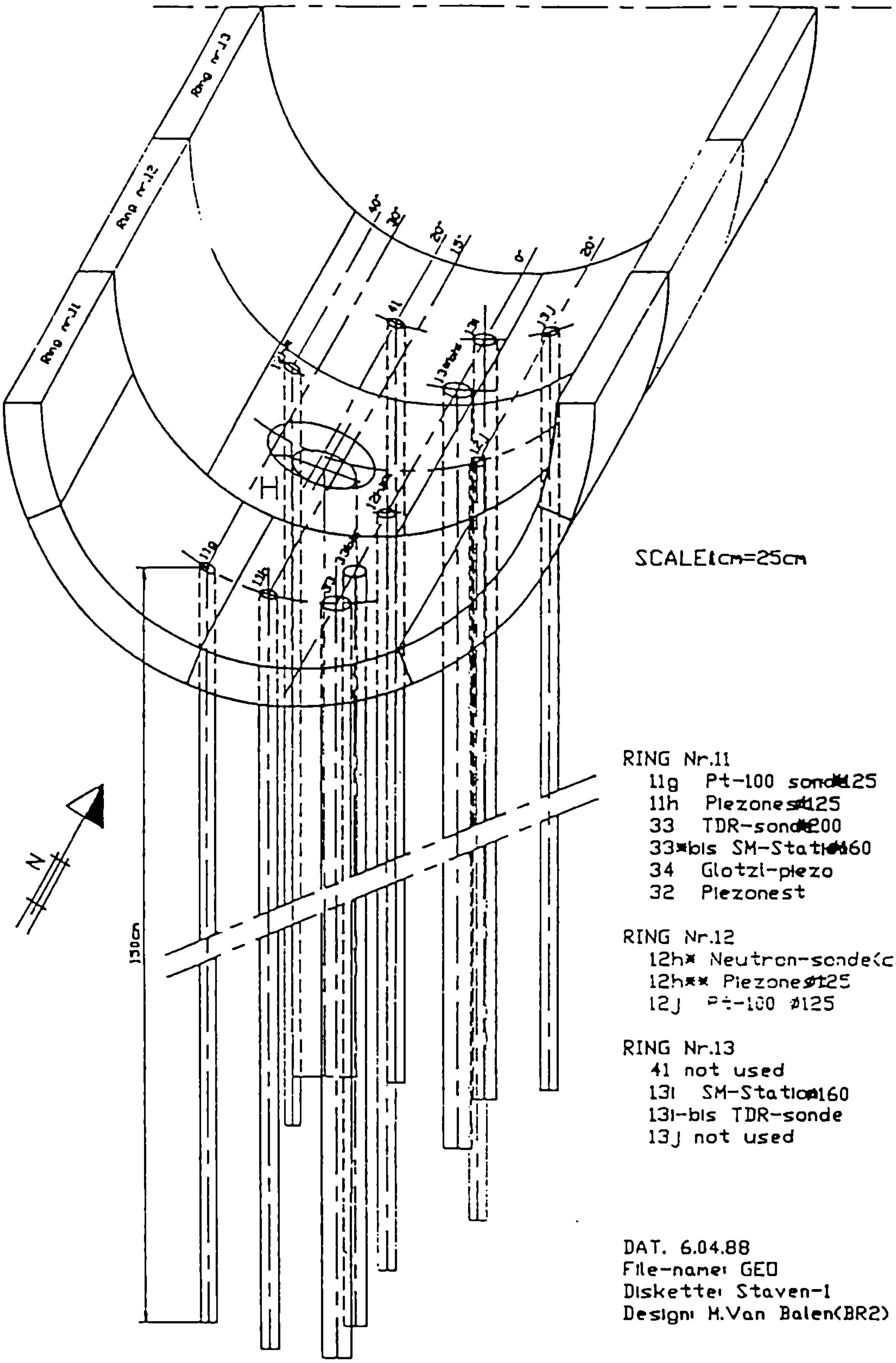


Figure 1 Clay host instrumentation - Location access holes in the lining

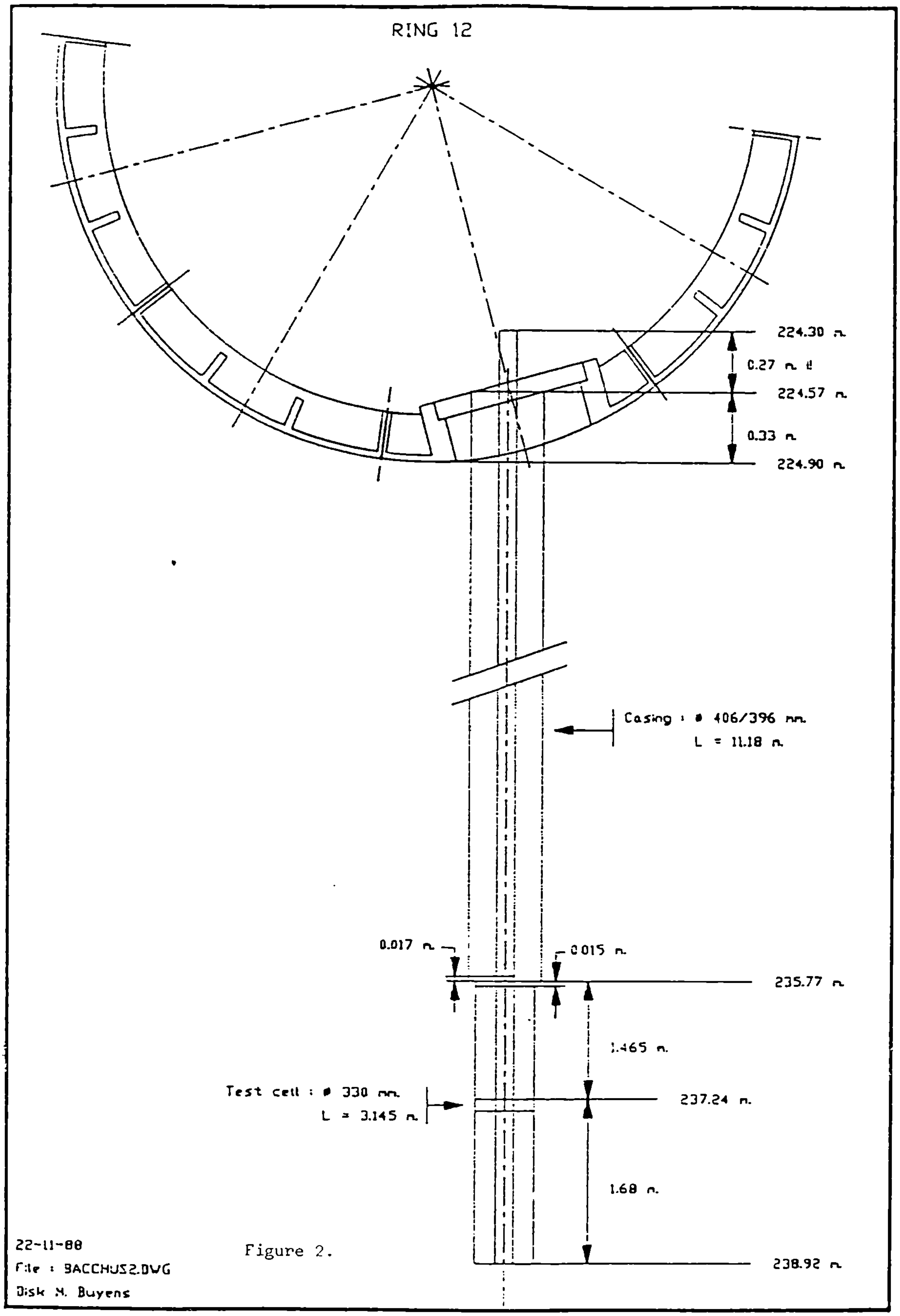

RING 12
224.30 m.
0.27 m.
224.57 m.
0.33 m.
224.90 m.
Casing : ø 406/396 mm.
L = 11.18 m.
0.017 m.
0.015 m.
235.77 m.
1.465 m.
Test cell : ø 330 mm.
L = 3.145 m.
237.24 m.
1.68 m.
238.92 m.
22-11-88
File : BACCHUS2.DWG
Disk M. Buyens

Figure 2.

FEASIBILITY STUDY FOR HIGH LEVEL RADIOACTIVE WASTE DISPOSAL IN DEEP BOREHOLES DRILLED FROM THE SURFACE

Contractor : ENEA, CRE CASACCIA, ROME (ITALY)
Contract No. : FI1W/0056-I
Duration of contract : January 1987 - December 1989
Period covered : January 1988 - December 1988
Project leader : E. Tassoni

A. OBJECTIVES AND SCOPE

A preliminary feasibility study has been carried out in the frame of a previous contract (255-80-7 WASI) on the disposal of high level and cladding hull waste in the Pliopleistocenic blue clay of Italy. Two preliminary repository models have been assessed :

- mined repository
- deep boreholes drilled from the surface

Of the two models, assessed for the same reference power programme (10 GWe), the deep borehole facility seems preferable if compared to the mined repository from several points of view but especially as regards total cost and flexibility. The deep borehole facility is also modular, it doesn't heavily interfere with local and use; even if inacceptable geological conditions are encountered in one borehole, operations can be moved to another part of the site with little cost penalty. The main goals of this project are the following :

- elaboration of a preliminary demonstration test of high level radioactive waste disposal in deep boreholes drilled from the surface;
- determination of the in laboratory and in situ behaviour of plugging and backfilling materials for deep borehole disposal.

B. WORK PROGRAMME

1. Feasibility demonstration test of deep borehole plugging.

1.1. Laboratory permeability tests, carried out both in small triaxial cells and in big oedometric cells, on samples formed by natural clay as well as on samples formed by clay sealed with different materials (cement, bentonite, etc.).

1.2. On site plugging test of a deep borehole, sealed with the material selected under item 1.1.

2. Project of a demonstration test for high level waste disposal in deep boreholes drilled from the surface.

C.PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Further laboratory tests, aimed at measuring the variations of permeability caused by central plugs of selected sealing materials within indisturbed clay samples, have been performed. The tested samples have been also examined by means of an electron microscope to study the nature cf the clay-sealing material interface.

Vertical electric resistivity sounding have been also carried out in the C.R.E. Casaccia area to know the stratigraphy of the site where the feasibility tests of geological disposal in a deep borehole drilled from the surface will be performed.

PROGRESS AND RESULTS

1.1. The permeability tests have been performed on clay samples after the emplacement of plugging materials (cement, bentonite). The tests have been carried out in a special cell which allows a central hole to be drilled in the clay sample and then filled with the plug material.
The further main conclusions obtained from these tests can be summarized as follows :

- the permeability of cement is lower than the permeability of clay;
- no adhesion has been observed between the clay and the different types of cement grout, with or without expansive additive, when cured under different conditions but without any application of load;
- when curing took place under load, good adhesion was observed between the clay and the cement grout;
- the flow of water in a sample consisting of a clay surrounded by remoulded clay is larger than in undisturbed clay.

1.2. To know the exact geological stratigraphy and the thickness of the clay formation situated under the C.R.E. Casaccia Centre a geophysical survey has been carried out. Eleven vertical electric resistivity soundings have been carried out in the Casaccia area. The geoelectric survey has located four different layers :

- an overburden layer (0-30 m) (electrical resistance 50+70 ohm-m) constituted by the permeable volcanic rock;
- a complex (30-250 m) (electrical resistance 15+20 ohm-m) constituted by the clayed tuff and the pleistocenic clayed sand;
- a complex (250-700 m) (electrical resistance 3 ohm-m) constituted by the pliocenic clay and miocenic flysh;
- a substratum constituted by the mezozoic carbonatic basement.

The exploration data are under examination for the project feasibility.

TRIALS AND CONTROL OF PLACING FILLING AND SEALING MATERIALS FOR DEPOSITS IN SCALE MODELS

Contractor : SOLETANCHE ENTREPRISE S.A.
Contract n° : FI 1W/0057
Duration of contract : February 1987 to March 1990
Period covered : January 1988 to December 1988
Project leader : D. GOUVENOT

A. OBJECTIVES AND SCOPE

The containers of radioactive wastes of high or low level activity will be disposed of in vertical shafts or horizontal galleries. In order to avoid any groundwater circulation in contact with the containers the void between the rock and the containers will be filled with sealing materials.

The materials, subject of this study, are cement based mortar formulations which are the results of previous studies and experiences of SOLETANCHE and C.E.A. These materials made of water, cement, clay and additives have to exhibit various mechanical characteristics as stability, imperviousness, durability, possible retention towards radioactive cations and above all easy placing in order to assure a good quality filling and sealing.

The first objective is to evaluate the hereabove characteristics of various mortar formulations, through lab tests, in order to select four formulations best suited to the filling and sealing.

The second objective is to improve and finalize the placing method of materials in scale models. To that aim, each of four scale models will be, one after each other, filled, cured at various temperatures, tested and dismantled for examination in order to define the best material placing technique.

B. WORK PROGRAMME

1. Laboratory studies

1.1 Theoretical studies. From the previous studies and experiences, choice of filling material formulations.

1.2 Preselection tests (based on the placing criteria). The rheological properties of the hereabove formulations are measured in fluid state (viscosity, shear strength, bleeding, workability limit).

1.3 Final selection of formulations. Measurements of characteristics of hardened materials (strength, shrinkage, water content, permeability, thermal conductivity, microstructure, retention and diffusion of caesium ions) cured at various temperatures (20, 50 and 80°C).

2 Experimental studies

2.1 Vertical scale models. Filling and quality controls of two successive vertical scale models.
- Study and improvements of materials placing and instrumentation.
- Controls of fresh and hardened materials in lab and in situ.
- Controls and tests of filling and sealing properties of materials in the scale models.

2.2 Horizontal scale models. Filling and quality controls of two successive horizontal scale models.
- Same tests and controls as vertical models.
- Finalization of materials placing technique. Particular specifications for horizontal scale models.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

After the first year (1988) dedicated to laboratory studies, the work in 1988 was mainly aimed at the experimental studies i.e. at the tests on scale models.

During the period, a first vertical model simulating a waste vitrified container in a shaft was prepared, sealed and tested after curing. This first model revealed some problems due to both design of the model and the technique of sealing. The results of tests were not as satisfactory as expected.

A second modified model was prepared according to the results of the first one. This model was sealed late 1988, and is now curing for 3 months. The sealing material used for the two scale models is a Portland cement based mortar. The formulation used for the second model was modified with regard to the first one, in order to improve the placing of the mortar in the annular space between container and shaft wall.

PROGRESS AND RESULTS

1. Laboratory studies

Laboratory studies were still going on early 1988 and new results concerning shrinkage and diffusion were known.

1.1 Shrinkage. The dimensional variations of samples cured at 20°C and in various moisture conditions showed the shrinkage is closely dependent of cement type. The following classification was found (from the best) :

CPA (Portland cement)
CLC (Portland + fly ash + slag)
CLK (Slag + Portland)

cured in water the samples exhibit swelling instead of shrinkage when cured in various moisturized air.

1.2 Diffusion. Diffusion tests of caesium 137 and tritium water (HTO) on 3 various sealing materials differing over the cement type have been in progress since one year at the CEA facilities. The stabilizations were not reached yet for most samples. However, a trend can be given concerning tritium water diffusion, as follows :

	Diffusion coefficient De (cm2/day)
CPA 55	1×10^{-3}
CLC 45	$< 4 \times 10^{-4}$
CLK 45	$2,4 \times 10^{-4}$

1.3 Selection of sealing materials for scale tests. The main criterion which was considered in order to select a sealing material was the shrinkage. Excessive shrinkage could lead to (i) cracks involving possible horizontal water circulation (ii) loose contacts between mortar and ground or between mortar and waste container involving possible vertical circulations of water, if any. These possible flaws could create water flow thousands of times higher than the flow due to intrinsic permeability of sealing material, ranging from 10-10 to 10-13 m/s.

For these reasons, a mortar formulation based on Portland cement was selected and used for the first vertical model.

2 Vertical scale models.

2.1 Design. The two attached figures show the design of the vertical scale model. A concrete cylinder 150 cm high and 90 cm in inside diameter is simulating the shaft. The bottom is plugged with a concrete layer and sealed with epoxy resin. A layer of filter sand put on the concrete plug allows leak tests to be performed. A filter fabric laid above the filter sand avoids sealing material to penetrate the sand during the filling operation.

A steel cylinder 50 cm in diameter and 100 cm high hermetically welded at the bottom, simulating the waste container, is placed in the middle of the shaft on the fabric.

The whole scale model is buried in the ground (fine silica sand); only the upper 40 cm of concrete cylinder is above the ground. The aim of the test is to check the quality of the sealing material placed in the annular space, and of the filling operation, and its behaviour to seal properly the waste container against possible water flow coming from the host ground.

Several successive designs were tested before getting the satisfactory present design. The steel container is filled with oil and heated by means of an electric heating element immersed in the oil and regulated at 80° C. Heavy steel ballasts put in the container avoid any displacement or floating at the time of filling the annular space with pseudo liquid sealing material.

2.2 Quality control equipments.

- Temperature : several thermocouples were placed in various locations in order to check the temperature variations in oil, sealing material and in the soil.
- Ultrasonic logging : 12 steel pipes placed inside the waste container, in the middle of annular space and outside the concrete cylinder, allow ultrasonic loggings to be performed, in order to check the quality of the contact between sealing mortar and walls of waste container and concrete cylinder.
- Leak test : two small metallic pipes fixed through the concrete cylinder at the level of filter sand allow water to be injected at the end of the curing period, in order to detect any flaws of the sealing along the steel or the concrete cylinders.
- Sampling : two diamond cored samples are recovered from the whole height of the sealing material at the end of the curing period, in order to measure the mechanical characteristics of the mortar cured at 80° C.
- Visual observation : the scale model is finally removed from the ground and cut in two parts, in order to check the overall quality of the scale test.

2.3 Filling with sealing material. After heating the steel container at 80° C, the annular space was filled with CPA 55 mortar. This mortar is made of : water
cement CPA 55
silica fume
clay
fluidifying agent
retarder

The placement of sealing material was carried out rather smoothly, even if the mortar was more viscous than expected. After filling completion, the mortar surface was covered with water and the whole insulated with fiber glass. After five months of cure at 80° C, the control tests were carried out.

2.4 Tests on cured scale model

2.4.1 Leak test. Water under a pressure of 50 kPa (0,5 bar) was applied at the bottom of the waste container through the water inlet and the filter sand. After about 3 minutes, the water came to the surface of the mortar along the concrete cylinder through a 2 cm long split and no thicker than 1/10 mm. The flow, very slight to start, slowly increased with he extension of the split. It seems the water pressure pushed the wall of concrete cylinder that cannot sustain the pressure without traction deformation. No leak appeared along the waste container.

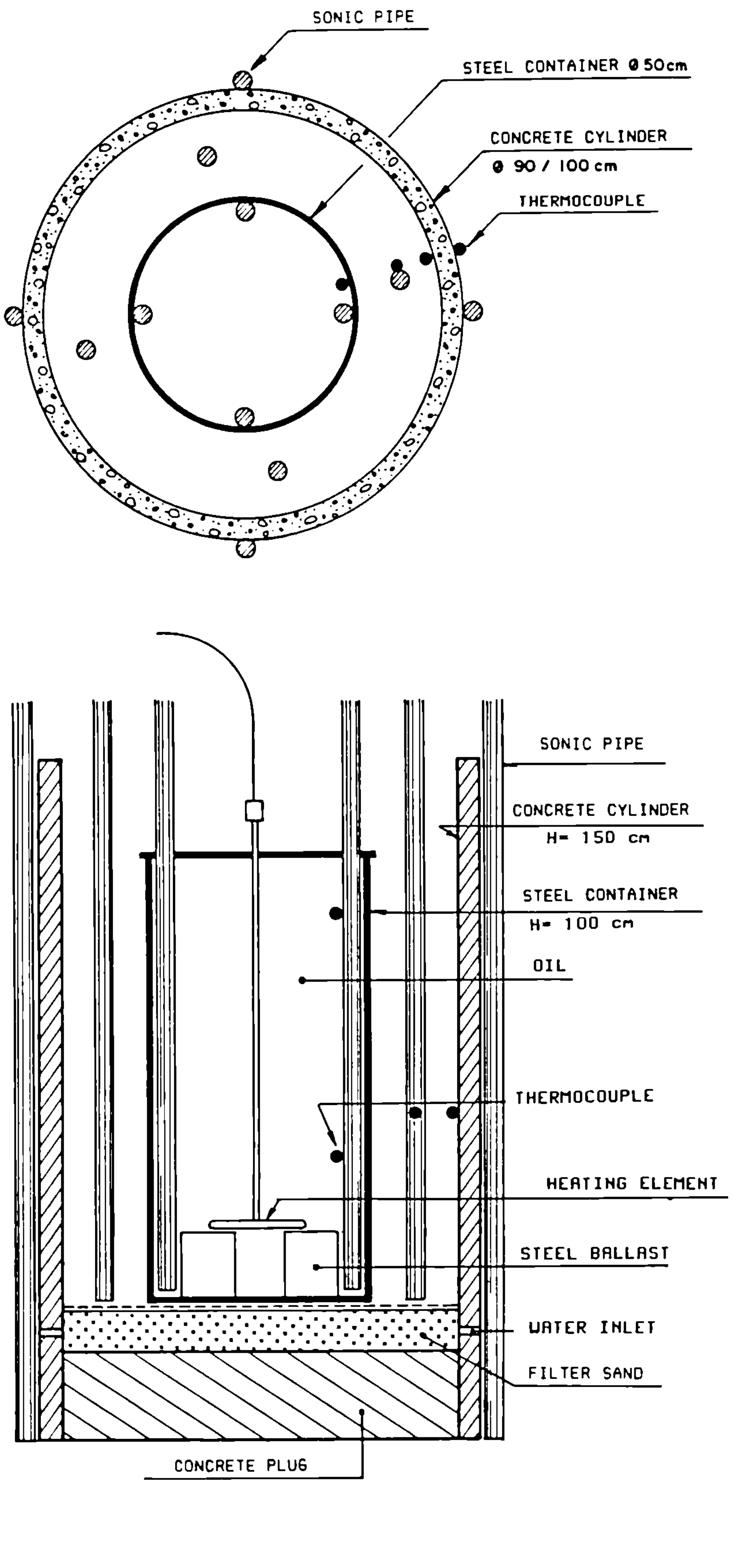
2.4.2 Ultrasonic loggings. Sonic tests were performed in the various pipes before the sealing and after the curing period. The comparison of the two series of results are shortly expected.

2.4.3 Visual observations. Two core samples were recovered in order to check the overall appearance of the mortar and to perform unconfined compression tests, and permeability tests as well. Presence of a few small air bubbles (<2 mm) was noted.

The model was then cut, by means of a diamond disc saw, in two parts along two opposite generatrices of the concrete cylinder. The visual observation highlighted the presence of some local flaws in the continuity of sealing mortar along the concrete cylinder : some kind of very flat lenses of air likely trapped during the placing phase, due to a too high viscosity of the mortar. These flaws, only localized along the concrete, should be avoided in the next scale model by using a more liquid mortar.

3. Next stages

The second vertical scale model was prepared and filled late 1988. Some slight modifications in the design were made to improve the placing of the sealing material. The formulation based on CPA cement was also modified to improve the initial viscosity and to reduce the possible shrinkage. After 3 months of cure at 80° C, the same series of tests will be performed. The leak test will be carried out more smoothly : the water pressure will be slowly increased, according to very small step, in order to better analyze the possible occurrence of leaks.



VERTICAL SCALE MODEL

Sealing of fractures and boreholes

Contractor : Commissariat à l'Energie Atomique, Fontenay-aux-Roses, France
Contract No : FI1W/0058.F
Duration of contract: October 1986 - December 1989
Period covered: January 1988 - December 1988
Project Leader: J.Y. BOISSON

A. OBJECTIVES AND SCOPE

Radioactive wastes disposal into deep cristalline rocks requires to carry out important underground works or geological investigations by the means of boreholes which might induce direct pathway or short circuits for water, between repository and ground surface. It is essential to use specific technics and methods allowing the sealing of these potential pathways.

Among several other possibilities, clay materials have been considered to fill the boreholes and shafts. One family of clays, the montmorillonites, are widely studied because of their qualities: good adsorption towards radionuclides, swelling properties in contact with water.

The aim of the study, is to determine, in laboratory and in situ, the behaviour of such clays, used as a plug in a borehole in contact with an hydraulic fracture, and to study the possible erosion of such a plug so as to be able to estimate the longevity of the sealing.

B. WORK PROGRAMME

2.1. Laboratory studies

2.1.1. Experimental erosion apparatus with bentonites are used, thanks to scale models simulating a fissure. Appropriate technics are carried out for the determination of the eroded clay mass by a water flow inside the model fissure.

Tests will be performed with different types of representative clays, i.e. those which are presentely investigated and could be used as sealing material in a real repository :

- the Na bentonite "Green Bond", from Wyoming (USA)
- the Na bentonite "MX 80", also from Wyoming (USA)
- one Ca bentonite (FoCa), from Fourges-Cahaignes (France).

The waters which will be used, will be of different types, either natural (from cristalline formations), or reconstituted with different percentages of relevant ions.

2.1.2. The behaviour study of this sealing material is completed with direct observations of the clay in the fissure, with granulometric analysis, and mineralogic X rays analysis.

We intend to ascertain the influence of the nature and velocity of the water flow. The experimental parameters will then be ion concentrations caracteristics and water flow rates.

2.2. In situ study

A fracture with a significative water flow, in a well known direction, will be selected, in situ, in a mine or quarry. The geometric position of this fracture will be identified thanks to boreholes, used themselves for hydraulic studies and sealed with bentonite at the fracture level.

The resulting water flow will be characterised by the means of tracers, water flow rates measurements at the free surface of the drift or on the face of the quarry.

After the experimentation, for a time depending upon the amount of the recovered clay mass, an over coring operation will be done, perpendicular to the fracture plane, both at the plug level, and around.

Modifications of the clay plug will be analysed in the laboratory, using the same technics as those for the scale model tests.

The clay, used for this test, will be one of those used in the laboratory.

2.3. Numerical model

The evaluation of the transported clay mass will be analysed and quantified both for the laboratory and in situ experimentations.

The results will be compared, and it is intended to establish a correlation between the two configurations.

This must lead us to provide a numerical model concerning the erosion behaviour and the durability of such bentonite plugs in boreholes in contact with water.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The first part of this research consists in laboratory tests, simulating the behaviour of a bentonite, sealing a fracture thanks to its hydration and swelling due to the presence of water, and submitted to erosion phenomena due to the action of the same water flow.

Tests have been performed under different conditions, considering two types of geometry for the simulated fracture, different types of bentonites at different physical stages of hydration. The measurements which are made aim at characterising and evaluating the longevity of the clay under different conditions.

PROGRESS AND RESULTS

2.1. Laboratory studies

2.1.a. As a first step, several free swelling tests with bentonites have been performed to evaluate and compare the behaviour of these bentonites under different conditions.

- Tests with highly compacted (1000 bars) cylindric samples ($\emptyset$ = 56 mm, h = 20 mm) of dry bentonites (W% init. = $\sim$ 10 %). Tests are run with two Na bentonites ("Green Bond" and "MX 80"), and one Ca bentonite ("FoCa"), and three types of waters for each sample (demineralised, granitic "Fanay", and volcanic "Volvic"). The free swelling curves show :

- More rapid free swelling for the first hours in the case of the Ca bentonite.
- Constant value ($\Delta h/ho \sim 150$ à 200 %) reached after 40-50 hours for the Ca bentonite.
- Very high swelling caracteristics ($\Delta h/ho > 400$ % after 85 days) in the case of the Na bentonite.
- Considering the influence of the water caracteristics, on the relative importance of the observed swelling it can be stated that :

Swelling (demineralised water)>Swelling (Fanay)>Swelling (Volvic) in any cases.

It can also be mentioned the strong difference in the clay mass behaviour between the Ca bentonite (which remains very homogeneous) and the Na bentonite wich presents after several days a zone of very soft gel. This shows the importance of the chemical ionic exchanges between clay and water and the influence on the erodability of the clay.

These chemical exchangesprocesses free static during swelling have been analysed for tests between Na bentonite and two waters (demineralised, and volcanic "Volvic") :

- Na, Ca, Mg and K ions are logically found in the demineralised water at the end of the tests.
- In the case of the Volvic water, it is shown that Ca, Mg and K ions are absorbed by the Na clay ; the amount of these ionic concentrations will be found to be very important concerning the erosion possibilities of the Na clay.

2.1.b. Erosion tests have been performed in a simulated vertical fracture (w = 5 mm) using a Na bentonite highly hydrated (W% = 400 to 1160 %) with waters at different ionic concentrations and different water flow rates.

In certain cases, it can be shown that the observed erosion phenomena (concentration of clay particules) during the first hours of the tests are more important for lower water flow rates. In the same conditions, this erosion appears to be non-existent or very low with certain ionic concentrations (cf. Fig.1). Depending on the nature and the concentration of ions incorporated in the different tested waters, it has been possible to establish an "erodability domain" for the Na bentonite (cf. Fig.2).

The lower the flow rates are, the more important is the development of different zones in the clay mass in contact with the water. The mineralogical and size particules caracteristics of these zones seem to be of very high importance for the erodability of the Na bentonite.

2.1.c. A second seriesof tests have been performed in a simulated horizontal fracture (w ≅ 2 mm) using cylindrical plugs (Ø 57 mm, h ≅ 100 m) of highly compacted samples (1000 bars) of dry (W% = 10 %) Na bentonite ("MX 80") and Ca bentonite (FoCa) in contact with demineralised water (cf. Fig.3).

The comparative tests, run for about four months yield the following constatations :

. The "radial swelling" of the clay in the fissure is in good agreement with what has been observed at the beginning of the test for the free staticswelling (cf. Fig.4) : higher swelling of the Ca bentonite at the beginning of the tests (∿ 40 days).

. After this period, the Na bentonite does not show the continuous swelling developped in static conditions and the observed swelling appears to be in the same order of magnitude for both bentonites. This point led us to conclude that the erosion phenomena are more active in the case of the Na bentonite.

. This constatation is confirmed by the mass concentration of clay measurements made on the flow which have been in contact with the plug. (cf. Fig.5). The average values for the MX 80 correspond to a mass between 25 and 50 mg/l for a flow rate of 10 ml/min.

. In the case of this Na bentonite, the development of the same zonation as the one previously mentioned, with dark particules and a light zone seems to be in direct connection with the relative fragility of this clay. At the opposite, the swelling part injected in the fissure during the test with the Ca bentonite appears to be homogeneous with no differenciation of colour or aspect.

2.2. In situ study

A suitable site has been identified in an uranium mine (COGEMA - Limousin), consisting in a gallery with a main fracture (45°) hydraulically active with a reasonable out flow rate. The complete identification and characterisation of this site will be done during the first months of 1989. The technical problems concerning the positionning of the clay plug in the fracture level, and the technic measurements to characterise the erosion phenomena are now under study.

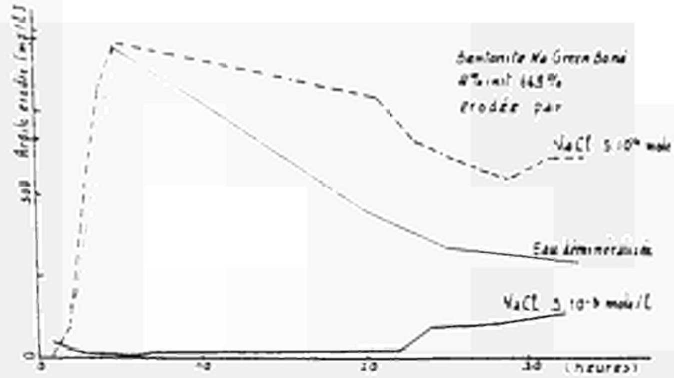


Figure n°1 : Variations de concentrations et masses cumulées de bentonite Na érodée en fonction du temps pour trois solutions

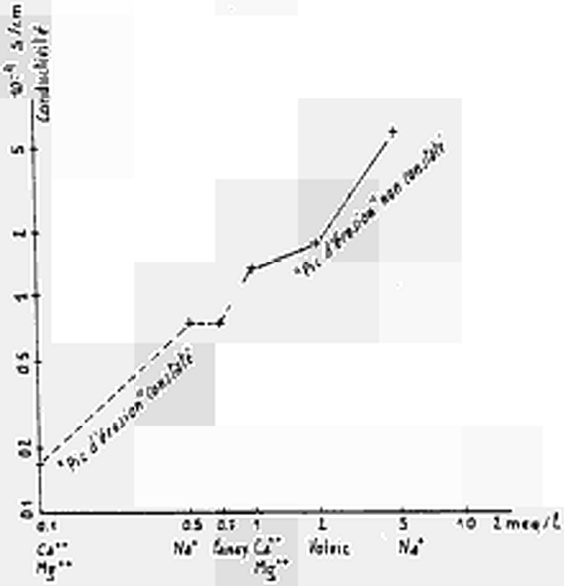


Figure n°2 : Relation entre conductivité et concentration ionique des solutions : érodabilité de la bentonite Na

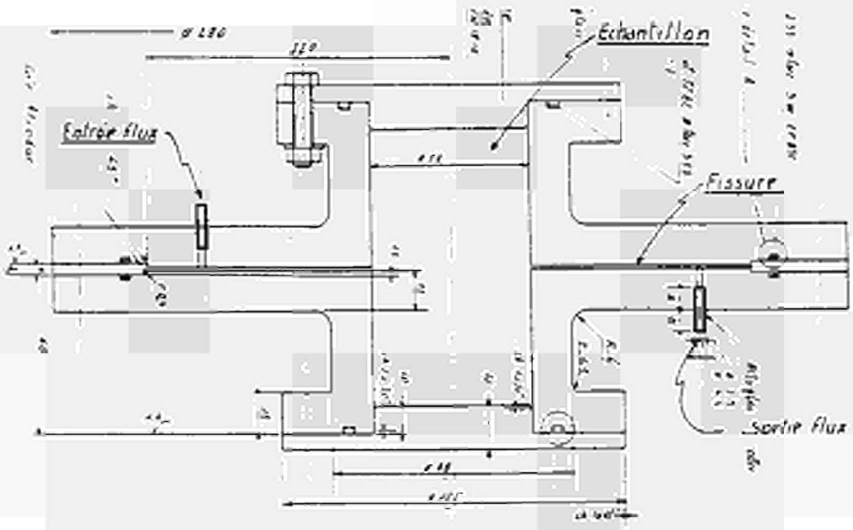


Figure 3 : Maquette fissure plane horizontale - Coupe.

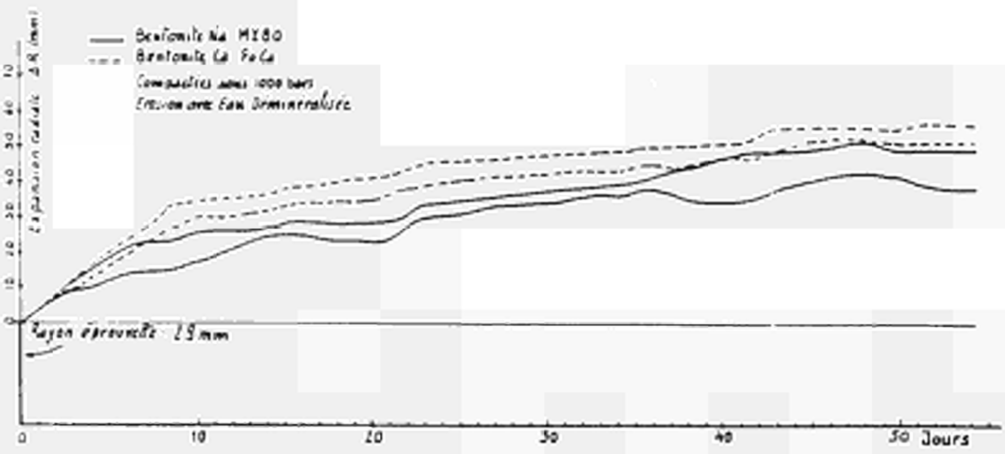


Figure n°4 : Gonflements radiaux en fonction du temps des bentonites Ca "FoCa" et Na "MX80" compactée, dans fissure horizontale - 1 mm

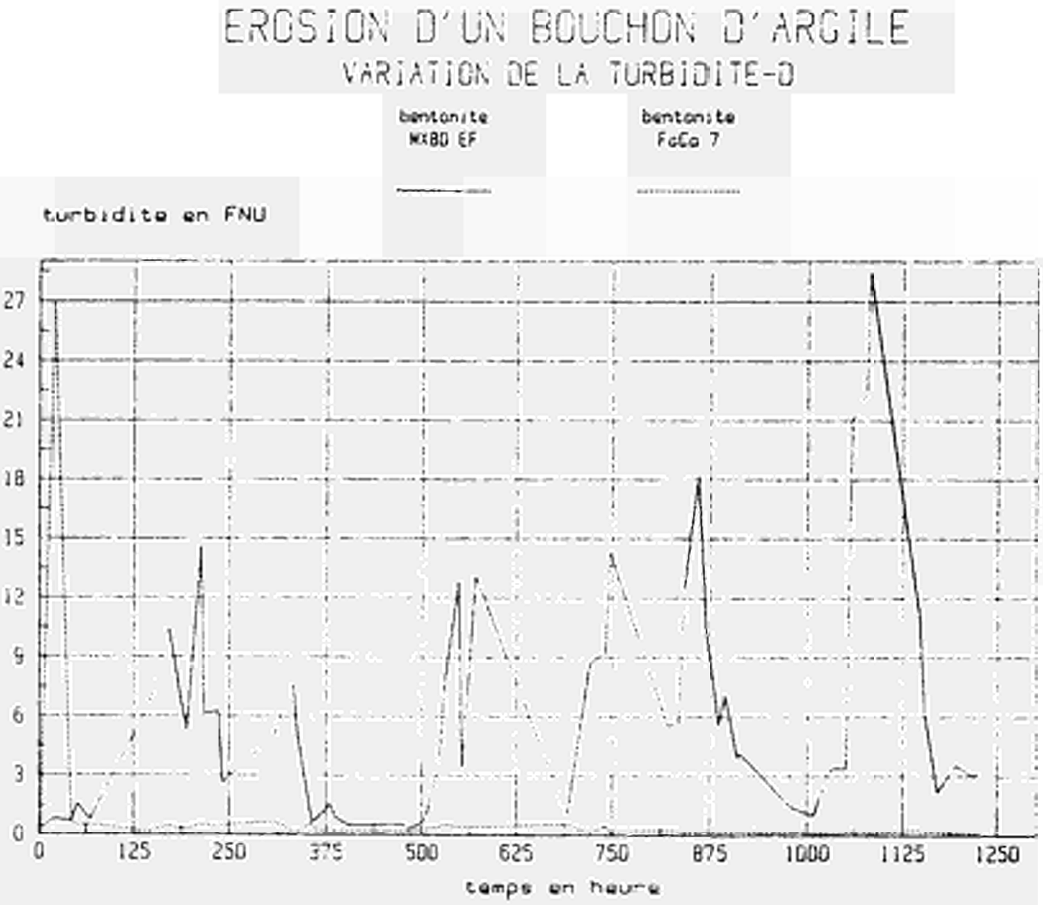


Figure n° 5 : Erosion des bentonites Na (MX80) et Ca (FoCa) en fonction du temps : mesure de la charge turbide en FNU

RESEARCH ON BACKFILLING AND SEALING OF ROOMS AND GALLERIES IN A REPOSITORY IN SALT

Contractor: GSF/Inst. f. Tieflagerung (IfT), Braunschweig, F.R.G.
Contract No.: FI 1W/0059/D
Duration of Contract: September 1986 - December 1989
Period covered: Jan. - Dec. 1988
Project Leader: M. W. Schmidt

A. OBJECTIVES AND SCOPE

The multibarrier concept for the final disposal of radioactive wastes provides backfill materials and seals as construction components with the purpose to prevent or to delay the transport of brines to the waste containers resp. to hinder radionuclides from reaching the biosphere via the brine as a consequence of an assumed incident.

Backfillings also have to act as mechanical stabilizers and to serve as a "filling agent", i. e. between waste containers.

Backfill materials have to take over a function as a barrier at a very early stage because so far there is no experience available on the long-term stability of constructive seal components.

In a first step technical concepts for backfilling of underground "repository rooms" with a so-called "reference backfill material" are to be developed and tested in the laboratory with supporting in situ tests.

B. WORK PROGRAMME

B.1.1 Soil mechanical laboratory research on backfill materials to investigate backfill characteristics
B.1.2 Geotechnical in situ measurements to determine the interaction between rock salt and backfill
B.1.3 Performance of geotechnical in situ measurements and rock mechanical laboratory tests on old, preconsolidated backfillings
B.1.4 Development of an evaluation matrix for selection and use of suitable backfillings in salt formations
B.2.1 Investigations in the laboratory on the mechanical and hydraulic parameters of sealing components
B.2.2 Surveying supervision of existing chamber and gallery sealing systems at the Asse salt mine
B.2.3 Development of an evaluation matrix for selection and use of suitable sealing components

C. PROGRESS OF WORK AND RESULTS OBTAINED

Some characteristics of backfill materials (salt grit) can be determined by using a newly developed test apparatus (Fig. 1). Salt grit can be heated up to 200 °C and compressed with an axial stress varying up to 103 MPa. After preparation breaking strength and deformation of these probes can be determined by using a KARMAN-machine.

The geotechnical control measurements in backfilled chamber 8a/532 m level and in the chamber itself did not show final results. The settling rates of the backfill range from 3 to 12 cm p. a., depending upon installation height and time of installation.

Automatic measurements of pillar deformation show a range of 0,8 to 1,3 mm p. a. Total extension in the eastern pillar was 6,3 mm in December 1988 (Fig. 2). Stress measurements in the southern supporting pillar exhibit an increase of pressure of up to 2,3 MPa in west-east direction. The north-south stress component is still at 0,5 MPa.

To determine the time-dependent deformation behaviour of backfill further extensometer measurements were carried out in the backfilled "double exploitation" chamber no. 3 at the 679/658 m level. In a horizontal borehole with a length of 38 m a divergence rate of 8 mm/month was measured.

A backfilled potash exploitation chamber at the 750 m level is being monitored with extensometers. The present deformation rate is 1,5 to 3,1 mm p. a. A convergence cross-section in close vicinity to this chamber in the main drift to the north show a horizontal convergence rate of 2,3 cm/a and a vertical convergence rate of 2,7 cm/a. Further measurements are no longer possible since the safety of the staff is hazarded by possible rock fall at this locality in the mine.

Potential building materials for the construction of underground sealing systems are salt concrete as well as salt bricks composed of salt grit with and without additives. One possible additive is clay. Crushed salt with a bentonite content of up to 20 % shows a higher density by 10 % without the influence of further compression of these mixtures.

In order to assess backfillings as well as construction components for sealing systems it is necessary to know details of their geochemical material properties. One basic matrix each was developed for the main properties of backfillings and sealings.

List of publications

SCHMIDT, M. W.; WALLMÜLLER, R., et al., Untersuchungen zum Versatz und Verschluß von Kammern und Strecken in einem Endlager im Salz. Abteilungsbericht IfT 10/88 (1988)

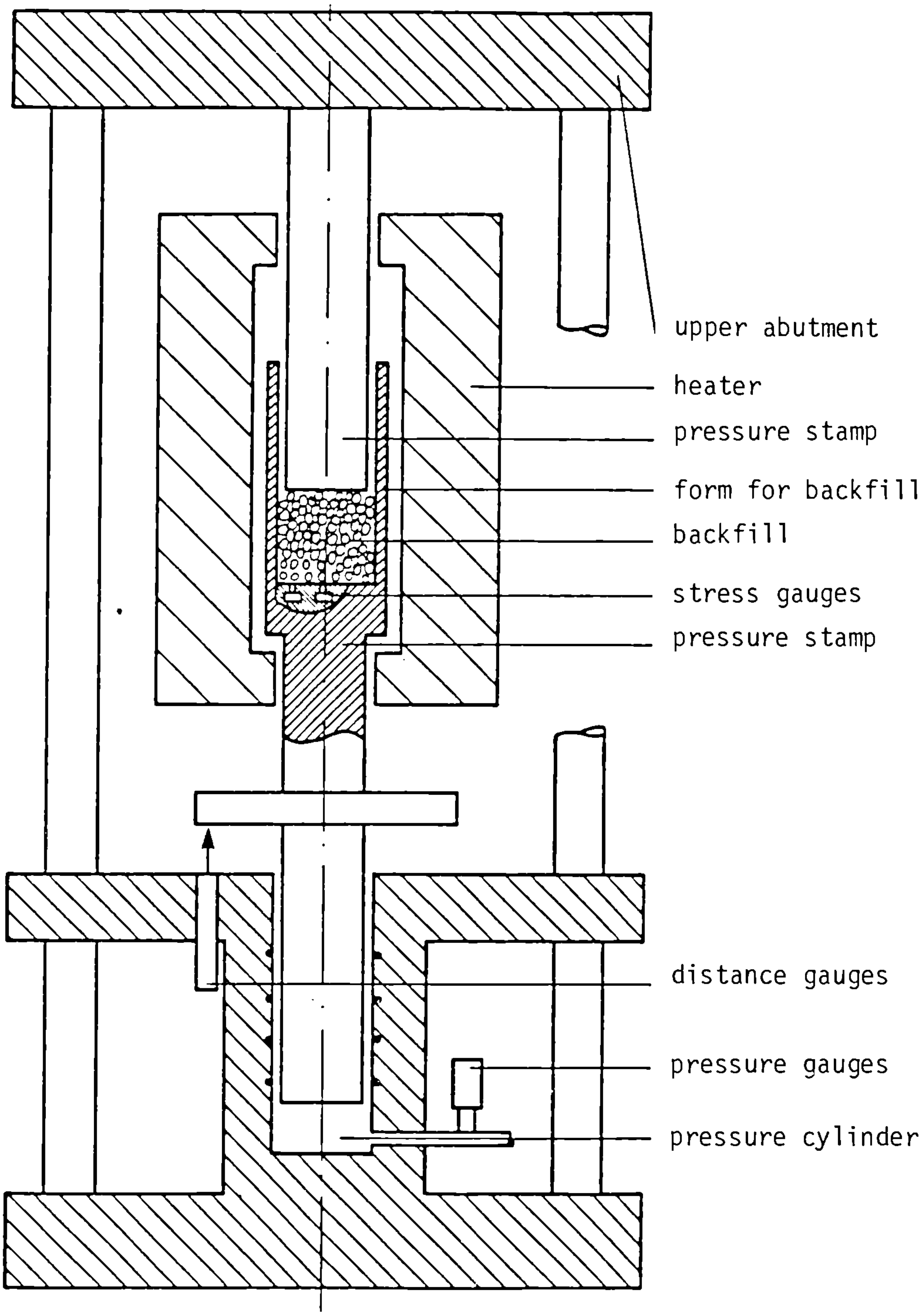


Fig. 1: Device for compaction of salt grit

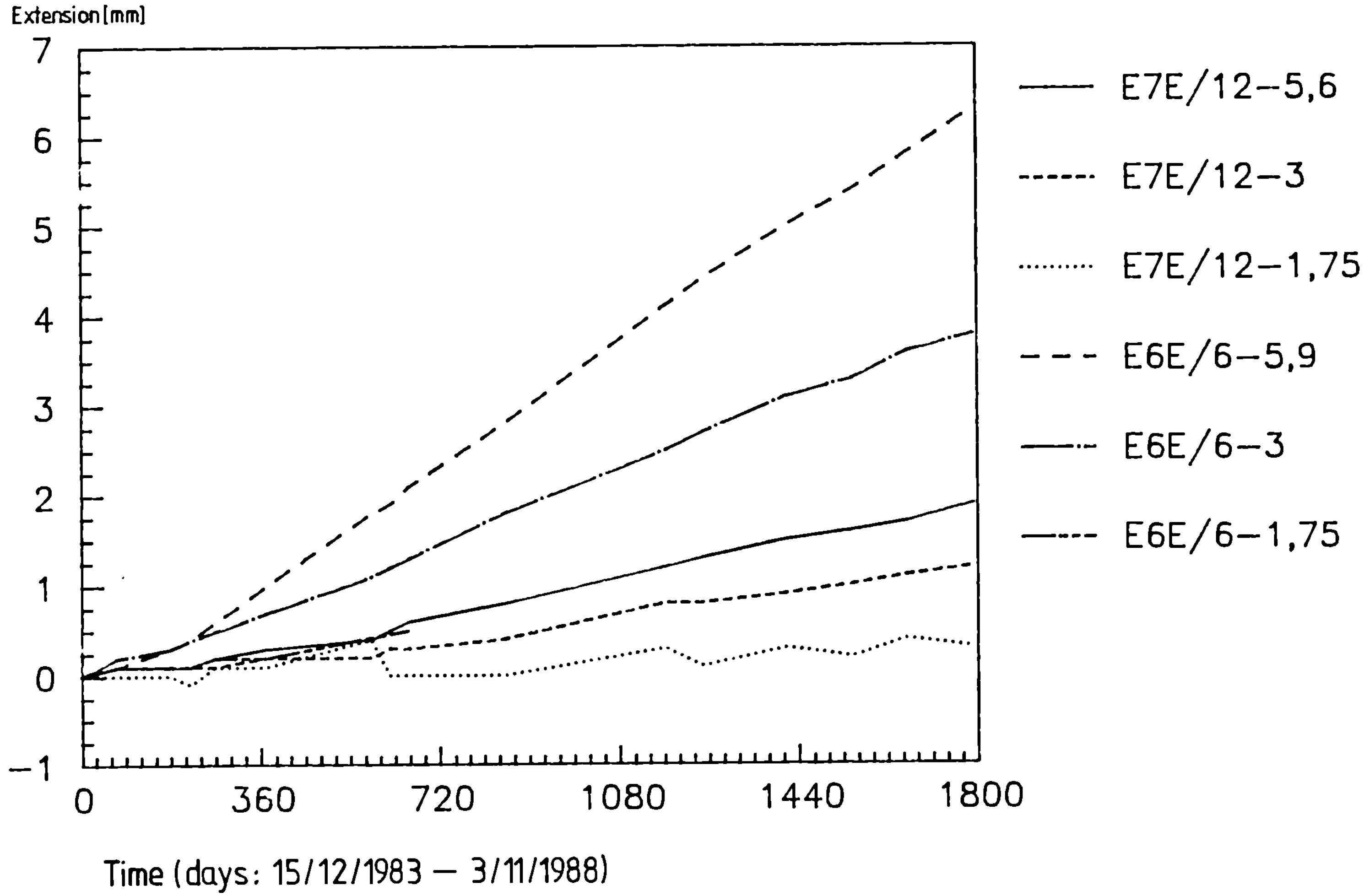


Fig.2: Measurement of deformation in the eastern pillar of Chamber 8a/532-m-level with extensometer

CRUSHED SALT BEHAVIOUR UNDER EFFECT OF A HEAT SOURCE IN BOREHOLES DRILLED IN A SALT MINE

Contractor	: ANDRA, PARIS, FRANCE
Contract n°	: F I1W/060
Duration of contract	: 24 months
Period covered	: 01/10/86 - 01/11/88
Project leader	: R. ANDRE JEHAN

A - OBJECTIVES AND SCOPE

The study concerns the final stage of a nuclear waste disposal in deep salt formations : the partial or complete closing of a site. Sealing of drifts and shafts must be performed as tight as possible to set up a barrier as similar as possible to the natural geological barrier.
As crushed salt backfilling corresponds to one of the considered concepts a research program is actually developped on the basis of the following axis :

- thermomechanical behaviour of crushed salt during thermal climax and during cooling,
- mechanical interaction between crushed salt and rocksalt,
- evolution of fluids, especially trapped air in crushed salt pores.

This study led to performing an in-situ experiment by heating crushed salt in several boreholes drilled in a salt layer in the Alsace Potash Mine (MDPA).

ARMINES (Association pour la Recherche et le Développement des Méthodes et Processus Industriels) is contractor for ANDRA to support the experiment which is conducted by LMS (Laboratoire de Mécanique des Solides) and the Engineering Department of MDPA (Mines de Potasses d'Alsace).

B - WORK PROGRAMME

1. Preparation of the test site
   Drilling of the boreholes :
   - six 240 mm diameter boreholes destined to be heated,
   - small measurement boreholes : four around each of the heated ones.

2. Instrumentation

2.1. Setting up of the computer controlled system developped by LMS for data acquisition and data transmission (from the test site to LMS).

2.2. Equipment of the borehole :
   - heated holes :
     + introduction of the framework supporting the electric wires and several transducers (for measurement of temperature, interstitial pressure of trapped air, total pressure of crushed salt, borehole closure),
     + backfilling of crushed salt in five of them, the sixth one leaving without backfill in order to check natural closure of heated salt and to compare its behaviour to the others'.

- Small holes : introduction of the temperature and extensometry measurement transducers.

3. Modelling

3.1. Preliminary calculations in order to define the following points :
   - thermal power to be supplied in each borehole to reach a wall temperature of 110° C,
   - geometrical shape and position of the heating sources,
   - heating process,
   - range of the transducers.

3.2. Interpretation of the in-situ results.

4. Laboratory experiments

The following experimental study has been conducted in order to determine mechanical and thermal properties of crushed salt :
   - physico-chemical analysis,
   - mechanical properties at ambient temperature,
   - mechanical behaviour under temperature (up to 100° C),
   - thermal study.

C - PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The technical difficulties occured at the beginning of heating in Borehole n° 1 were related in the former annual report. Here we just mention that they required the substitution of electric wires in the other boreholes and involved an extention of about six months for the duration of the experiment.

1988 was basically devoted to instrumentation of Boreholes n° 2 to 6 (fig. 1) and to analysis and interpretation of the first results. Heating was entirely stopped at the end of the year, in order to observe cooling until end of March 1989.

Progress and results

1. Analysis of the firts results

Analysis of the first results lead to the following observations :

- because of its variations between about 3° C and 28° C, ambient temperature in the galleries of the test site appears to be an important parameter, which must be taken in account ;

- in the heating boreholes :

   + a steady state temperature was established very rapidly in backfill materials, within less than 10 hours ;
   + temperature of backfill is homogeneous at the wall of the boreholes all along the heated zone ;
   + evolution of diameter closure, mesured in several directions, show a good axial symmetry of displacements.

- in the peripheral boreholes :
  + in spite of complex geometry of the site, temperature distribution in the rockmass appears to be isotropic inside a radius of one meter around the heating boreholes ;
  + security restrictions of the mine implied possibility of air convection in not backfilled Borehole n° 6 ; this resulted to uncontrolled heat input and thermal distribution ; therefore the measurements in this borehole cannot be considered as representative for the behaviour of rocksalt without backfilling.

2. Modelling

Modelling has been conducted at two levels, using finite elements codes : first thermal to determine temperature distribution in the rockmass, then thermomechanical, on the basis, of previous results, to evaluate in-situ stress distribution besides heating effects, boreholes closure in particular.

2.1. Thermal calculations

Thermal properties taken in account were defined as follows :

- for crushed salt : thermal conductivity was measured, using laboratory tests ; heat capacity was evaluated with respect to porosity,

- for rocksalt : the characteristics are based on laboratory tests as well as on published results ;

- all the characteristics are supposed to be constant ; this allows to use the linear law of Fourier according to which the resulting temperature field can be determined by adding initial temperature to temperature increase.

The calculations were performed in three steps :

a. Variation effect of ambient temperature in the galleries :

- Temperature in the galleries, measured since their excavation seven years ago, appears to be a sinusoid with a period of one year ;

- Cross sections of the galleries are represented in a mesh using plane assumption (fig. 2a) ;

- Initial temperature for the whole points of the mesh is considered to be equal to rock temperature at the depth of the site, that is 35° C ;

- Several cases of thermal exchanges at the walls of the galleries are considered, such as forced convection, imposed temperature ;

- Temperature applied to transient state calculations is :
  + average simusoidal evolution during the first seven years,
  + effective temperature measured in the galleries during the experiment.

- The following results have been obtained : a satisfying representation is given for measured temperatures in the peripheral boreholes sealed in the rockmass around Boreholes n° 2 to 5 between day 110 and day 260, that is before start of heating in these boreholes (fig. 2b).

b. <u>Thermal interaction of the heating boreholes</u>

- The boreholes are considered to be points in a mesh plane chosen normal to the borehole axis (fig. 3a), therefore they are assumed to have an infinite length ; moreover thermal exchanges due to the galleries cannot be taken in account : that is why only qualitative results may be considered.

- The average measured temperature at the wall of each borehole is applied to the representative points according to the time table shown in figure 1.

- Transient state calculations lead to the following results :
  + interaction of a hole on its neighbour one is significant (fig. 3b), but
  + the temperature field can be considered as of revolution inside a radius of more than one meter around each hole ; this is in agreement with the experimental measurements ;
  + in conclusion, it is possible to perform an accurate thermal study of a borehole using axisymmetric assumption.

c. <u>Thermal study of a borehole</u>

- The backfilled borehole is represented in an axisymmetric model, which dimensions are 50 meters radius and 100 meters hight (fig. 4a) ;

- The electric wires near the wall are modelised like a torus ;

- Initial temperature is supposed to be equal to zero for all the points of the mesh ;

- Increase of temperature is determined through this calculation : as shown on figure 4b there is a satisfying agreement between modelling and measurements.

2.2 Thermomechanical calculations

Interpretation of borehole closure and pressure measurements in the backfilled holes required to perform creep tests on rocksalt from the experimental site under various stress and temperature levels. The laboratory tests allowed to identify a constitutive law which parameters are introduced in finite elements computations ; modelling is undertaken in the following maner ;

- Stress field around the boreholes before heating is evaluated through a plane modelling of the galleries ;

- Initial stress field at the beginning of heating is obtained by axysymmetric modelling (fig. 5) ;

- Thermal field calculated previously is introduced into thermomechanical calculations using assumption of axial symmetry : mechanical behaviour of crushed salt is considered to be non linear elastic, characterized by pressure - void ratio relationship.

The results are in good agreement with measurements (fig. 6) ; an important pressure increase in the backfilling material is found without any significant effect on borehole closure. This phenomenon is explained by rheological properties of rocksalt, highly sensitive to temperature.

3. Future prospects

Analysis and interpretation of the measurements will be carried on especially to examine evolution and effects of cooling. On the test site the boreholes will be dismantled in order to observe the state of their wall, of the backfill and the instrumentation, especially the electric wires.

List of publications

/1/ CEC workshop on "Backfilling and Sealing of radioactive waste repositories" 2 nd meeting, january 18th - 19th, 1988 in Fontenay aux Roses (France)

/2/ French Comittee of Rock Mechanics (CFMR), december 8th, 1988 in Paris.

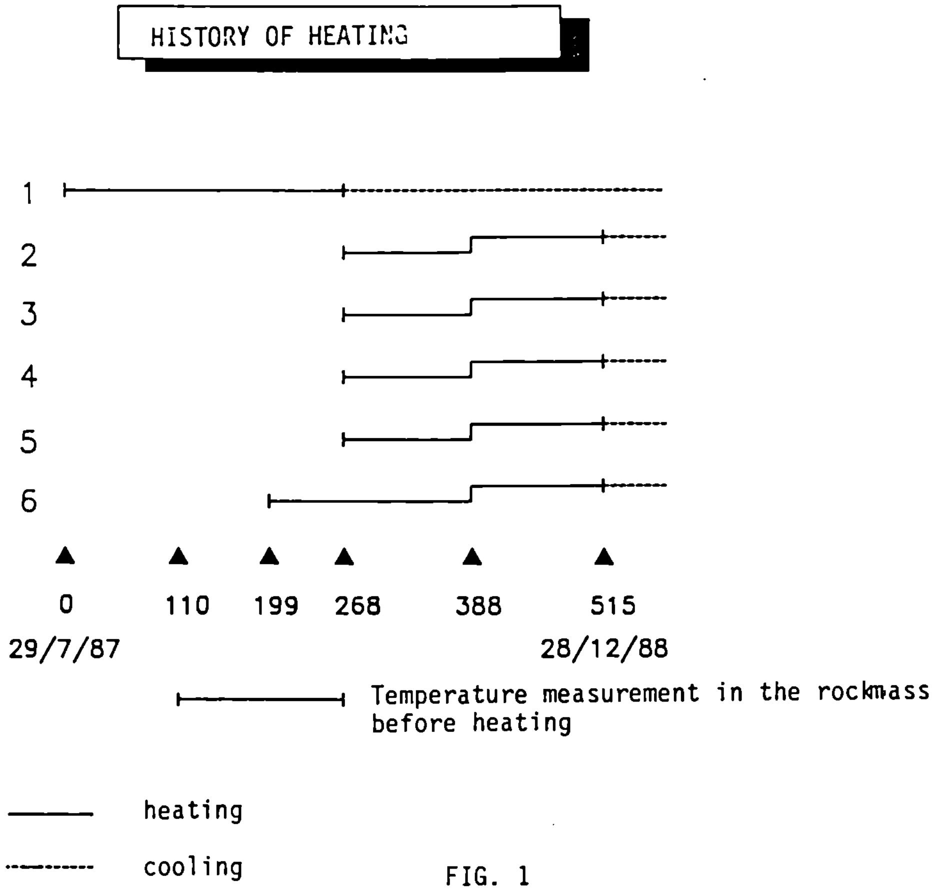
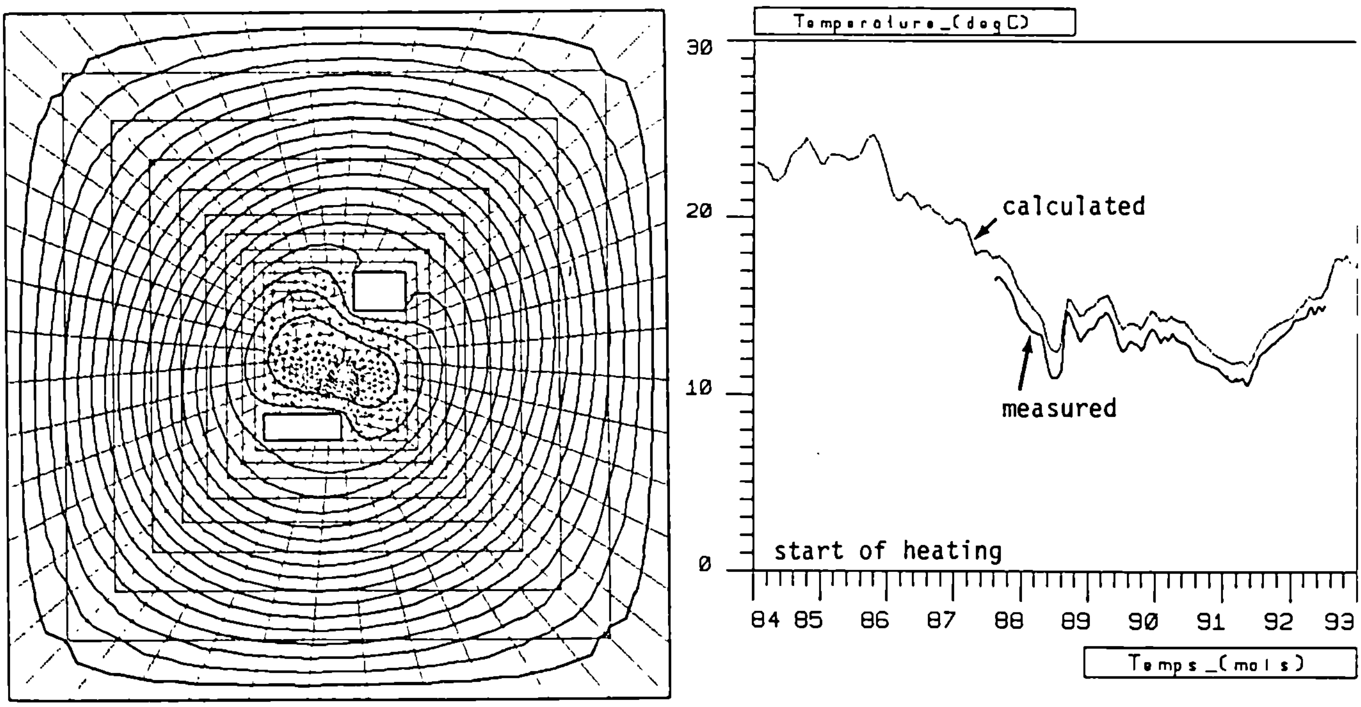


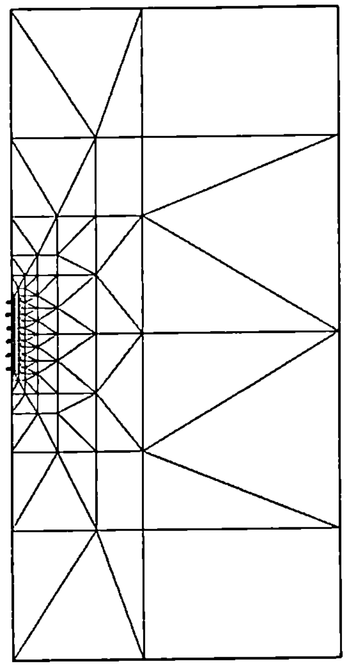
FIG. 1



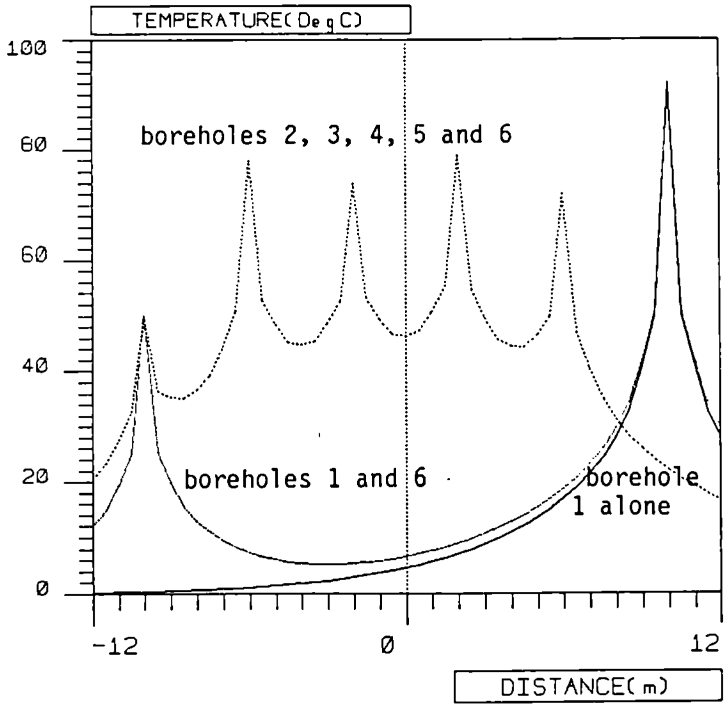
(a) temperature distribution in the rockmass around the galleries before start of heating
(isotherms from 22°C to 35°C)

(b) Comparison between measured and calculated temperature in the rockmass at a 75 cm depth from the inferior gallery

FIG. 2 : Variation effect of ambient temperature in the galleries

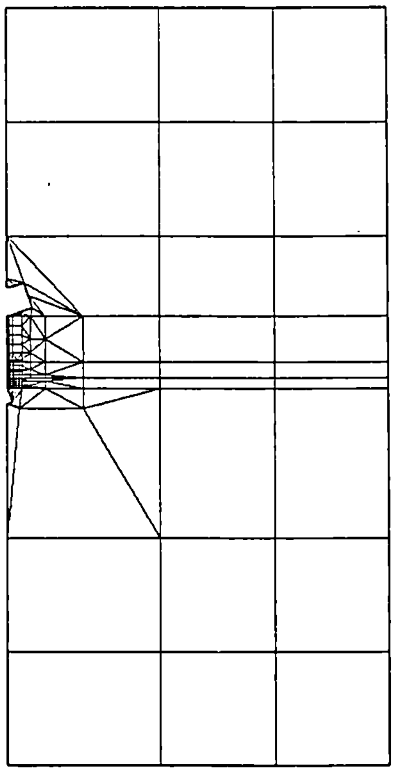


(a) plane mesh

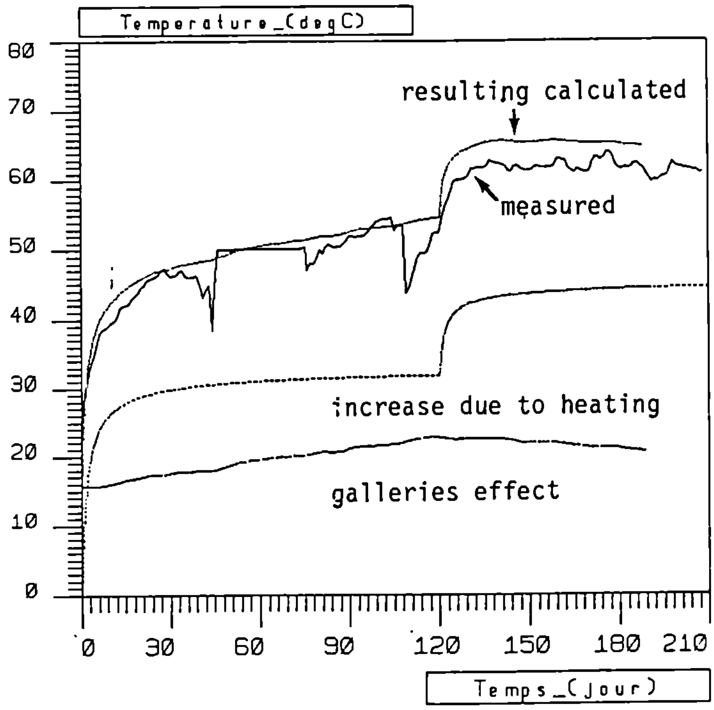


(6) Temperature distribution along the axis containing the boreholes

FIG. 3 : Thermal interaction of the heating boreholes



(a) axisymmetric mesh



(b) rockmass temperature at a depth of 126 cm from the inferior gallery

FIG. 4 : Thermal study of a borehole

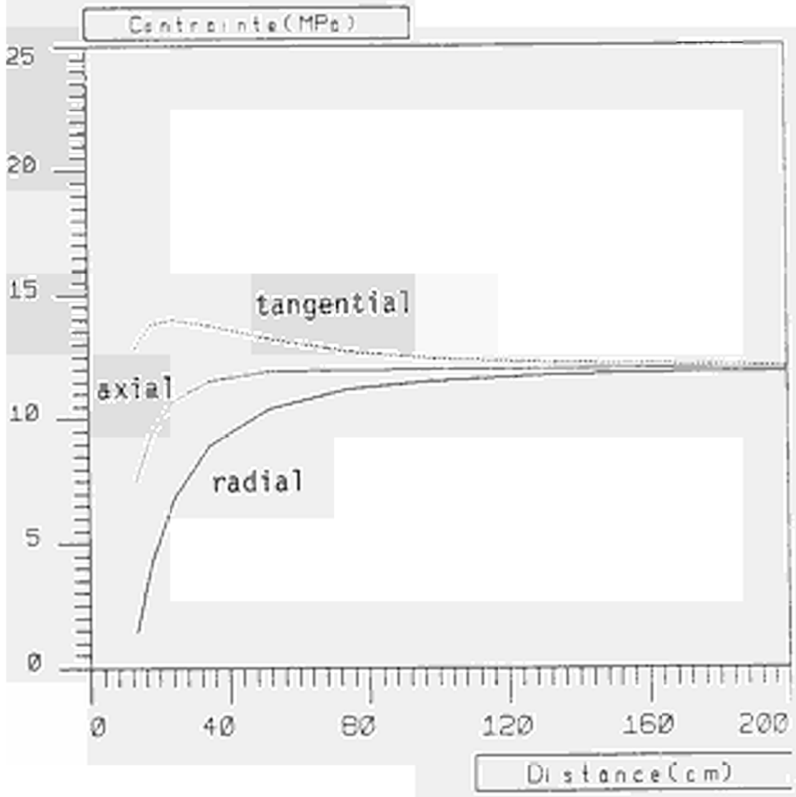


FIG. 5 : Stresses around a borehole at the beginning of heating

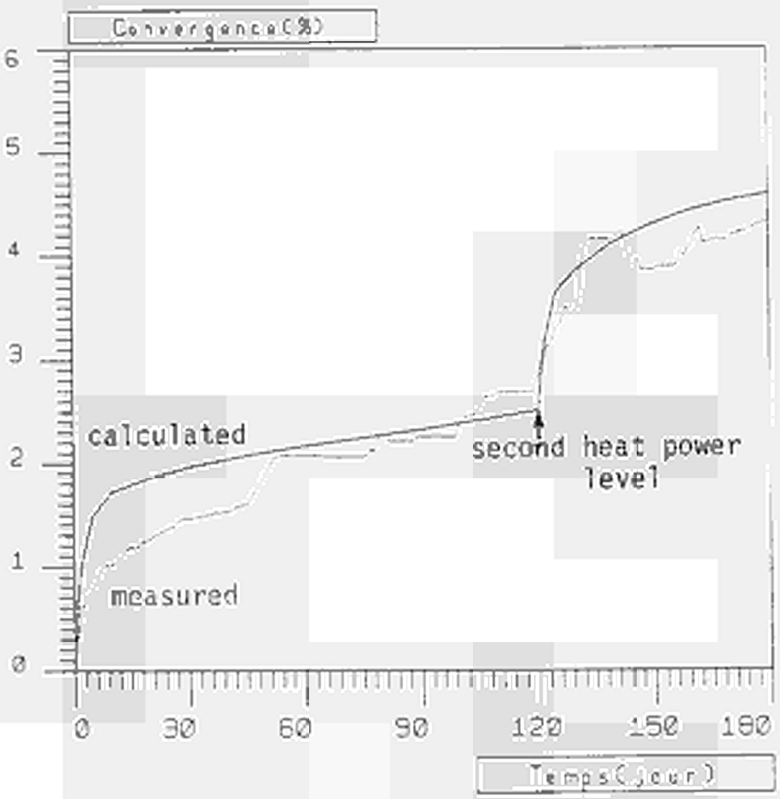


FIG 6 : Closure of borehole n°4 versus time (days)

Study of the thermal behaviour of clay-based buffer materials on reduced scale mock-ups and in an underground laboratory

Contractor : CEA, CEN Fontenay-aux-Roses
Contract n° : FI1W - 0061 F
Working period : July 1986 - December 1989
Period covered : January 1988 - December 1988
Project leader : M. DARDAINE

A. OBJECTIVES AND SCOPE

Clay materials could be used as components of engineered barriers placed between high-level waste canisters and host rock.

The purpose of this work is to compare the behaviour of different types of materials, both homogeneous and heterogeneous, simultaneously subjected to heat and humidity gradients. Only the initial stage of storage, the so-called "dried stage", is simulated.

The study, which involves instrumentation, first involves the desing and construction of an experimental device to reproduce the actual physical conditions of waste disposal : temperature and water content. Subsequently, in collaboration with CEN/SCK, an in-situ heat transfert experiment will be carried out in the Mol underground experiment facility. An electrical heater will be surrounded with buffer and backfill materials. The entire system will be instrumented with temperature, moisture and pressure sensors. The test will be supported by heat transfer modelling.

B. WORK PROGRAMME

B.1 Research and development work on water content sensors :
- . thermal conductivity sensor,
- . capacitance sensor.

B.2 Design and construction of an experimental heat transfert device. Experiments and modelling.

B.3 Properties of backfill materials, determined in the underground experiment facility at Mol (Belgium). In-situ experiments on heat transfer and modelling.

C. STATE OF PROGRESS AND OBTAINED RESULTS

State of progress

In situ determination of water content for different Engineered Barrier Candidates comes from the effective measurement of their thermal conductivity with thermal shock probes. A series of twenty probes, so called of second generation, has been made by a Manufacturer, specialised in miniature probe making. Twelve of them have been used as instrumentation of the BACCHUS in situ experiment.

The first step, laboratory study of a Engineered Barrier Candidate submitted to a thermal gradient, exclusively made of selected natural french clay 4a, has lasted for one and an half year. Taking into pieces the clay crowns, in order to analyse them (water content measurement, cracking amount) is planned for early 1989.

A first theoretical approach, transposition of an already developed pattern designed for studying the tranfers of heat-mass couples inside non-saturated porous environments, allowed to record relevant thermophysic parameters and to determine some orders of magnitude.

A simple experiment allowed to study the cracking coming from thermo mechanical stresses induced inside the material through a heat flux, and its filling up under hydration effect.

At last, a large part of the work was dedicated to realise the instrumentation and to built a mock-up, which is an element of the BACCHUS experiment. It has effectively been put in its place, inside the underground laboratory of Mol, during November. Heating up the mandrel is planned in early 1989. This experiment is made in cooperation with CEN/SCK (EC contract FI 1W/0055).

PROGRESS AND RESULTS

B.1 Research and development work on water sensors

Height of the twelve thermal shock probes, radially embedded into the middle crown of thermal bench, allowed the thermal conductivity measurement during the whole duration of the experiment, either at ambient temperature or with thermal gradients.

Twelve shock probes, of second generation, industrially manufactured, have been embedded inside the BACCHUS experiment. Important delays, due to the delivery of miniature thermistors, have not allowed to perform, up to now, a sufficient number of preliminary measurements.

B.2 Design and construction of an experimental heat transfer device. Experiments and modelling.

Laboratory study of the behaviour of a Engineered Barrier Candidate, submitted to a thermal flux without external water supplying has been continued over the whole year with a mandrel temperature of 130°C.

As an indication, figure 1 shows the respective temperature profiles plotted for different temperature values of heating mandrel.

Observed variations of thermal conductivity, but mainly radial samples made at regular time intervals, bring out a slow evolution of radial distribution of water content inside the clay. This can be due, on one hand, to water migration from hot part to cold one, but also and mainly, to evaporation at mandrel level.

A simple experiment consisted to create an orthocylinder of composite clay of 10 centimeters, with an electric resistor introduced in its center, and whose periphery was water-tight. Knowing the dilatation coefficient of clay versus temperature, this allowed to explain cracking phenomena which appear, either very quickly (due to thermomechanical stresses) or more slowly (due to hydromechanical stresses). A simple calculation allowed to determine their direction and intensity, as well as the induced deformations.

B.3 <u>In situ experiment on heat transfer and modelling BACCHUS experiment (figure 2)</u>

The BACCHUS experiment (BACkfill Control for Hlw Underground Storage) consists to study the behaviour evolution of a Engineered Barrier Candidate placed to the bottom of a 14 meters depth well, drilled in the old gallery of the underground laboratory of Mol (Belgium), which will be submitted to a heat flux given by an electric mandrel, and to a possible hydration of its surrounding field.

It is covered up by 4 crowns of BOOM clay, recompacted over 1.45 m height, then, in the tubed part of the drilling, by a stopper made of 50 cm of thermohardening resin and, at last, by a cement stuffing.

Instrumentatin of the experiment is formed of 4 Glötzl cells (stress measurement) embedded into two metal flanges bound up with internal tubing, of 34 Pt 100 ohms probes, and of 12 thermal shock probes, radially arranged in different directions on several levels.

All sensors are connected to a measuring frame through 25 meters cables grouped inside the supporting tube.

The drilling and instrumentation of surrounding field have been done by CEN/SCK. Effective installation of the whole equipment, wiring and connection checking to the measuring frame and to the computer, and first reference measurements, have been performed during the last quarter.

It has to be noted that, once installed to the bottom of the well, the natural convergence was not sufficient to restore a close contact between the experiment and the surrounding earth.

Accordingly, it has been decided to fill in, at best, the free space with powdery BOOM clay.

For a configuration including a well and a proximity engineered barrier, designed for a clay site, it is admitted the effective deposit of a vitrified block of High Level Waste (HLW) will only be done once there is a close contact between the barrier and the natural environment.

That is the reason why the power increase of the heating mandrel is only planned in early 1989. With natural convergence and BOOM clay addition, it is expected such a condition will be met.

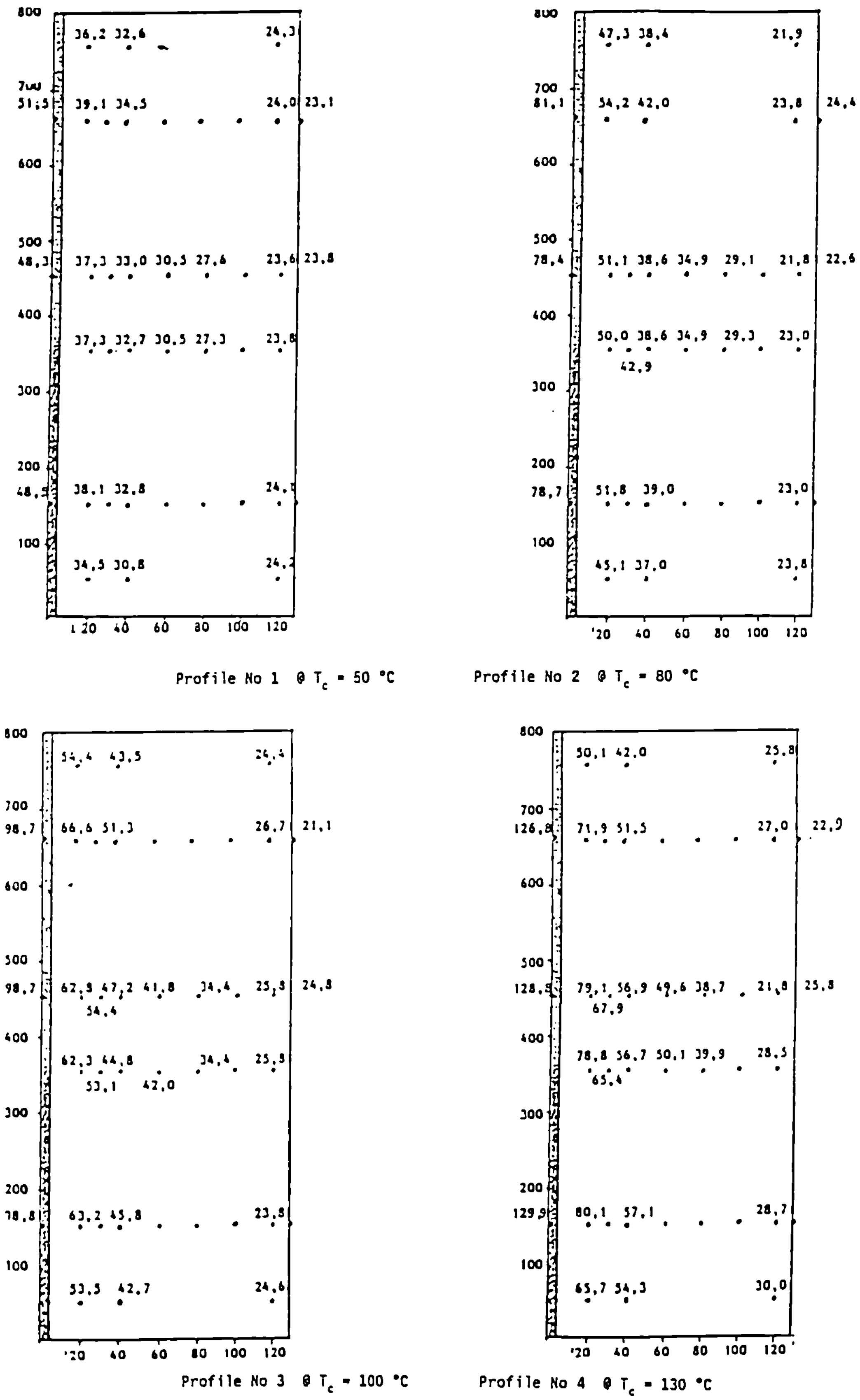


Figure 1 . Successive temperature profiles measured on thermal bench for crown temperatures T_c of 50, 80, 100 and 130 °C.

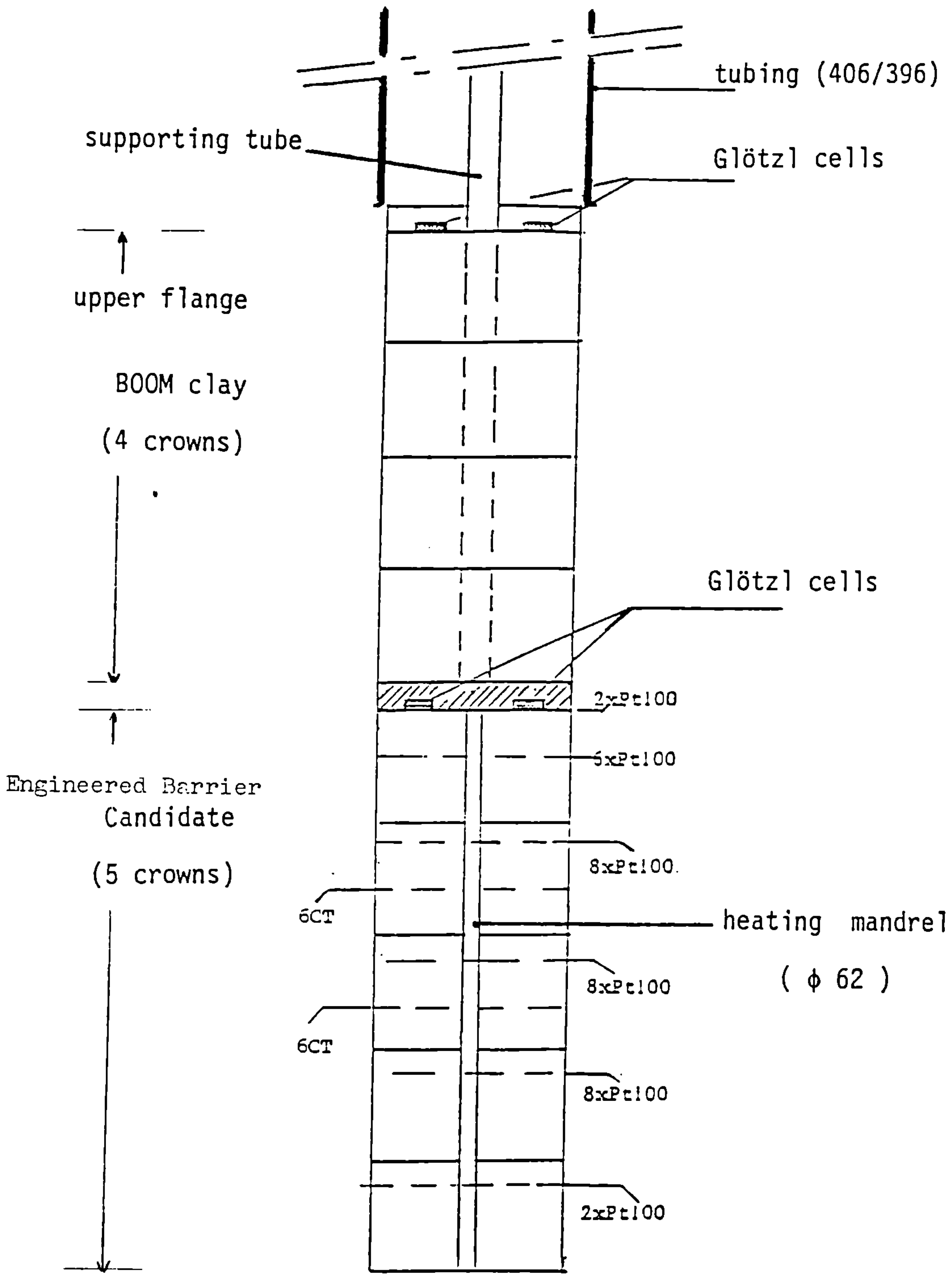


Figure 2 . BACCHUS experiment : sketch of the lower part (E.B.C.) and of the upper part (BOOM clay).

Development of effective concepts for attenuating the near-field effects of HLW in argillaceous rocks

Contractor : SCK/CEN, Mol, Belgium
Contract No : FI1W/00145-B
Duration of contract : From July 1987 to December 1989
Period covered : January 1988 to December 1988
Project leader : A. Bonne, L. Noynaert

A. OBJECTIVES AND SCOPE

In the earlier designs and concept developments of mined repositories of HLW in argillaceous formations, the main post-closure functions of the backfilling of disposal galleries are to provide mechanical stability of the system and to obstruct preferential pathways for radionuclide release.

In the recent years realistic and actual data about waste characteristics, waste arisings and potentially favourable argillaceous formation becoming available, and also in view optimization a need arises to develop more specific concepts which provide a limitation of the near-field effects and thus improve the performances of the individual structural and barriers components.

The aim of the study is to develop backfilling concepts specific for mined repositories in a stratiform argillaceous formation, which also participate in the attenuation of the effects of heat release, radiation, oxidation, wetting and corrosion, and provide all at the same time mechanical stability, chemical compatibility and emplacement feasibility.

B. WORK PROGRAMME

1. Definition and quantification of near-field effects around HLW-packages in a clay environment for selective representative emplacement configurations of mined repositories (in-gallery and in-floor concept).
2. Settling of disturbance allowances for the various components in the near-field, glass matrix, container material, concrete and in situ surrounding clay).
3. On the basis of the results of 1 and 2, definition of the attenuation factor to be achieved by engineered barriers with a particular attention to the thermal effects the radiation dose and the oxidation effects.
4. Definition of complementary requirements of these interface materials regarding their chemical compatibility with the waste packages and the clay environment, their mechanical stability, their permeability, water saturation level and their emplacement feasibility.
5. Selection of appropriate materials on the basis of heat transfer and shielding properties, radiation resistance hydraulic properties, chemical buffering, and radionuclide retention capabilities, mechanical characteristics, longevity, available methods of application and costs.
6. Design and dimensioning concept which meet the above set of requirements and etablishment of appropriate emplacement techniques and procedure for the selected emplacement configurations.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

A critical study of the two proposed configurations for mined high level waste repositories in argillaceous formation allowed the definition and the quantification of the near-field effects to be expected int he engineered barriers and the immediate surrounding host rock. This allows to formulate criteria for the selection of potential material to be used in the engineered barrier components.

PROGRESS AND RESULT

The materials considered in this study are the glass matrix, stainless steel canister, concrete, precompacted clays or clay based mixtures and the local Boom clay with modified properties due to mining and waste disposal.

The main results are for :

a) glass matrix :
   - thermal stresses exist into the glass matrix due to the thermal gradient created by its own heat production and due to difference between the thermal expansion coefficients of glass matrix and stainless steel. This may influence the lixivation surface,
   - the lixivation rate reaches a minimum value at pH = 7 ;
b) stainless steel canister :
   - the stainless steel container will fail as soon as the engineered envelope is saturated and hydrostatically loaded. This is due to the formation of a plastic zone (as soon as the outside pressure on the canister reaches 1.37 MPa) and due to the sensitivity of the material at stress-corrosion cracking ;
c) concrete
   - concrete with a low hydratation heat and hydratation rate poured in limited thicknesses are suitable to avoid thermal cracking,
   - concrete at its early age (< 1 month) is not suitable to withstand thermal load,
   - the best candidates concrete to be used in an argillaceous host rock are those without tricalcium aluminate (C_3A = 0 %) and with a molar ratio Al_2O_3/Fe_2O_3 lower than 0.64.
     SRPC is suitable as well,
   - the radiation disturbances allowances (1E+19 n/cm² and 2E+08 Gy) are not reached in a repository during the first 10,000 years ;
d) clay host rock
   - due to the high impermeability and the water mineral interaction of the clay, a clay temperature increase has as consequence a raise of the pore water pressure which take a long time to dissipate. Those increase is accompanied by a reduction of the effective stresses and the capacity of the clay to sustain shear stress. A modification of the water flux escaping the disposal formation is studied actually.

TITLE: Studies of Historic Concrete
Contractor Taylor Woodrow Construction Ltd
Contract No : FIIW/0158.UK
Duration of Contract: January 1988 to June 1989
Period Covered: January 1988 to December 1988
Project Leader: Mr. T.P. Lees

A. OBJECTIVES AND SCOPE

Durability of concrete materials, either as matrix for low and intermediate level waste, or as backfilling/sealing materials for repositories, is a major concern in the preparation of both design and safety assessments.

The objective of this study is to examine ancient concretes, and modern ones with proven durability; to identify the parameters which contribute to their durability. The work is an extension to that previously undertaken ('An Historical Examination of Concrete' (EUR 10937) reported March 1986) in which a number of of factors which appeared to be significant in relation to durability were identified. The results will be used to provide data to enable life predictions on modern concretes to be made.

B. WORK PROGRAMME

1. Sample retrieval

The following sources of samples will be investigated:

1.1 Ancient Materials

Preference will be given to obtaining samples from concretes containing natural pozzoloanic materials, where the sample is partially buried and from zones of the concrete where carbonation may be incomplete.

1.2 Medieval Materials (up to 1824)

Preference will be given to materials where a hydraulic binder is likely to have been used.

1.3 Modern Portland Cement Based Concretes (1824 - present)

Preference will be given to securing samples from large masses of underground concrete located in a moist environment and with a likely pozzolanic content.

2. Examination & Testing

The procedure set out below will be followed where appropriate, depending on the size and nature of the sample and the outcome of previous tests:

2.1 Visual examination
2.2 Assessment of depth of carbonation
2.3 Optical microscopy of thin sections taken at positions representing fully carbonated material, the carbonation front and uncarbonated material.
2.4 Scanning electron microscopy of polished sections and fracture surfaces in the same zones.
2.5 Individual phases identified above analysed by electron probe microanalysis.
2.6 Chemical and XRD analysis of portions representative of the different zones of the sample.

2.7 Pore structure analysis of similar portions.
2.8 Analysis of pore fluid expressed from the portions used in 2.7
2.9 Other tests may also be made to resolve issues arising from the results of the above procedure.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Additional funding for this project had been promised by the U.K. Department of the Environment. However, after a protracted period of negotiation, this offer was withdrawn. As a result very little progress was made with the study until October 1988. A sixth month time extension has been granted so the study will now be completed in June 1989.
A criticism of the work undertaken previously was that not enough ancient samples were studied, and that these samples were small and so possibly unrepresentative. Therefore it has been decided to concentrate on ancient concrete samples for this study.
A total of twenty eight samples have been obtained from sites in the United Kingdom, Italy and Austria. Apart from initial documentation, no analytical studies have been completed.

Progress and Results

1.1 Ancient samples obtained are detailed in Table I.
With the exception of the sample retrieved from Hadrian's Wall, coring has not been used as the method of retrieval. This is because either a) site directors were not prepared to risk unnecessary damage to ancient structures from core removal (It must be remembered that the samples have been obtained from ancient monuments) or b) coring was not practical in the excavations concerned.
As coring was not used the samples are not as large as intitially desired. The large majority of samples have been retrieved from the interior of structures as well as from the visible surface material.
1.2 Medieval samples obtained are detailed in Table II.
1.3 the Modern Portland cement based samples obtained aredetailed in Table III.
2.1 Visual assessment and photography of the samples has begun but is not yet completed.
2.2-2.9 No further analyses have been completed.

TABLE 1 : ANCIENT CONCRETE SAMPLES

Type	Approximate construction date	Origin
Core taken through rubble and mortar infill	120 A.D.	Hadrian's Wall, Northumberland
Interstitial mortar from wall of amphitheatre	? 100-300 A.D.	Roman Carnuntum; near Petronell, Eastern Austria
Waterproof cement bath lining	200 A.D.	Roman Vicus - a small town consisting mainly of bath houses; on Castelporziano estate 24km south-west of Rome, Italy.
2 x wall foundations	200 A.D.	
4 x interior rubble and mortar filled wall cavity of	150 A.D.	
1 x interior rubble and mortar filled wall cavity	200-300 A.D.	
Interstitial wall mortar	150 A.D.	
2 x wall mortar	100-300 A.D.	Roman baths at the junction of Huggin Hill and Upper Thames Street, City of London. (Recent excavation)
2 x floor mortar	100-300 A.D.	
Roof mortar	100-300 A.D.	
2 x wall mortar Foundation mortar	100 A.D.	Matrice Roman villa, 160Km east of Rome, near Campobasso, Italy
3 x mortar from wall	150-200 A.D.	Tor Paterno, a large Roman villa, probably imperial, on Castelporziano estate 24Km south-west of Rome, Italy

TABLE II : MEDIEVAL CONCRETE SAMPLES

Type	Approximate construction date	Origin
Foundations of apse Mortar from wall core	900-1100 A.D.	Rebuilding of Roman town of Vicus; for details see Table I
3 x wall mortar	800-1900 A.D.	Rebuilding of Tor Paterno villa; for details see Table I

TABLE III MODERN CONCRETE SAMPLES (1824 ONWARDS)

Type	Approximate construction date	Origin
Cores taken through foundations	marked on 1872 Ordnance Survey Map	Large warehouse in Thomas More Street, London E1 which has since been demolished

TITLE: The Development and Application of Mathematical Modelling approaches to interactive effects for concrete backfill in hard rock and argillaceous hosts

Contractor: Taylor Woodrow Construction Limited
Contract No: FI 1W/0159. UK
Duration of contract: April 1988 to June 1989
Period Covered: Period to December 1988
Project Leader: Dr G Storer

A. OBJECTIVES AND SCOPE

The main objective of this study is to develop a mathematical model utilising the ADINA CODE which can predict interactions between concrete backfill and host geologies for underground radwaste repositories.

B. WORK PROGRAMME

The work programme shall comprise:-

1. Literature review of recent international work to ensure that the mathematical models developed using ADINA represent a reasonable state of the art in terms of both analytical approach and representation of the physical phenomena.

2. Investigations of the newly available features provided in the latest release of the ADINA code and consideration of their appropriateness to modelling repository interaction behaviour.

3. Agreement, in consultation with Community partners, upon appropriate generic disposal scenarios in both hard rock and argillaceous hosts.

4. Study of mechanical and thermal interactions between the host material, the backfill and canistered waste, including long-term creep effects.

5. Assessment of the development of damage in and adjacent to the backfilling material, and how the damage may increase with time both short-term from early thermal effects arising from cement and waste and very long-term from lithostatic creep effects.

C. PROGRESS OF WORK AND OBTAINED RESULTS

Contractual agreement with the UK Department of Environment, who have agreed in principle to fund the balance of monies to complete the project, is now being actively progressed by their supervising agent, the Building Research Establishment (BRE). This follows a period of prolonged delay. It is hoped that a 1st March 1989 commencement date will be agreed with UK DoE. No technical progress within the project can be reported for 1988 therefore although some of the investigations into the ADINA code have been carried out independently.

QUALITY ASSURANCE ASPECTS OF WASTE EMPLACEMENT AND BACKFILLING IN ILW AND LLW REPOSITORIES

CONTRACTOR	Bullen and Partners Consulting Engineers 188 London Road Croydon CR9 1PT England
CONTRACT	FI 1W - 0161 - UK
DURATION OF CONTRACT	From February 1988 to July 1989
PERIOD COVERED	February 1988 to January 1989
PROJECT LEADER	Dr JA Allison

A OBJECTIVES AND SCOPE

The objectives of the study are :

- To critically examine existing conceptual design proposals for the deep underground disposal of ILW and LLW in water-bearing (non-saliferous) host rocks, with particular reference to the quality control/quality assurance aspects of waste emplacement and backfilling.

- To examine the extent to which existing proposals enable effective monitoring and remedial action to be achieved.

- To identify the parameters which are most effective in describing backfill material properties and waste unit characteristics, both before and after placement.

- To identify appropriate measurement techniques and the means of application of such measurements in the development of a coherent quality control system.

- To examine the ways in which repository design details, methods of waste unit/backfill placement and the sequence of operations involved may be adapted to ensure that an effective quality assurance system can be established.

B WORK PROGRAMME

1 **Phase 1** will comprise a brief review of the following aspects of the ILW and LLW disposal concepts developed in member states of the European Community :

- The types of waste units envisaged for deep underground disposal and the quality control/quality assurance systems currently adopted or envisaged for the pre-disposal stages.

- The waste emplacement systems currently proposed and the extent to which quality control/quality assurance procedures have been specified.

- The range of backfill materials under consideration, and the properties which relate most effectively to their intended functions.

Phase 2 will concentrate upon the generic deep-level repository design concepts for ILW and LLW disposal (including plutonium-contaminated wastes) which are incorporated within the current UK waste disposal strategy.

Consideration will be given to a range of backfill materials, reflecting the scope of current community research and development.

For these reference conditions, more detailed evaluation of quality control/quality assurance measures will be carried out, itemising the procedures and measurements required at each stage of development.

Where appropriate, the need for repository design modifications is to be considered as a means of improving the exercise of quality control in the waste emplacement/ backfilling processes.

The following are among the factors to be considered :

- The extent to which backfill materials can be prepared in solid 'fill block' form or as pre-batched 'fluid form' fills at off-site production centres, or at an adjacent surface facility.

- The scope for palletising waste units in appropriate spatial arrays, with partial interstitial filling, prior to emplacement and final sealing.

- The scope for exercising control over the geometry of waste unit arrays such that required full volumes are accurately known and directly comparable with volumes actually emplaced.

- Means of separate monitoring for groups or 'cells' of emplaced waste units/backfill, such that the origin of defects may be identified, and recovery or appropriate remedial action achieved.

- The extent to which the emplacement processes for different categories of waste (incorporated in a single repository) may require different approaches in exercising a consistent level of control.

3 The study will include recommendations concerning the development of quality control/quality assurance procedures for waste emplacement, backfilling and monitoring to complement and extend those which are being (or have been) developed in relation to the pre-disposal stages. Supplementary recommendations concerning the correlation of readily measured quality control parameters and specified performance properties will be provided.

C PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

A literature study covering the aspects outlined in Phase 1 has been completed. Limited information has been published and in consequence the study is being extended to cover non-member European states and North American countries.

- Several different types and sizes of waste unit have been identified and the quality control/quality assurance aspects of their production ascertained according to country of origin.

- Apart from the Swedish proposals concerning waste emplacement systems, detailed information of which is still being sought, the proposals of other countries are only known in outline. Waste emplacement systems are clearly under development, but details have yet to be published. Enquiries are in hand.

- Backfill materials, which are taken to include overpacks, buffering and repository linings in addition to the mass infill materials, are quite widely reported. Mass infill materials are found to be site specific and to vary according to host rock type, groundwater regime and depth of burial. Some further work is still to be completed prior to commencing Phase 2 of the study.

PROGRESS AND RESULTS

Waste Emplacement Systems

The waste emplacement systems proposed internationally are linked to site specific or host rock specific conceptual repository designs. As a result, the proposals of each country are being examined individually.

Quality control and quality assurance considerations relevant to waste emplacement and backfilling may need to include :

- ground stabilising measures
- preparation of engineered backfill materials
- preparation of disposal chambers prior to waste emplacement
- preparation of waste units prior to disposal
- placement of waste units
- sealing of disposal chambers
- sealing of access-ways and shafts
- monitoring performance of completed construction
- contingency plans for retrieval of waste units.

Reports defining specific quality control/quality assurance measures associated with the above aspects are generally lacking in the literature, with the exception of the outline proposals published by Sweden. Further enquiries are in hand.

Backfilling Materials

Repository backfill materials identified by various countries vary widely in composition, and include :

- reconstituted spoil
- cementitious-based material
- clay-based material
- compressed bentonite (pelletised or in block-form)
- bitumen
- zeolites
- magnesium oxide

The prime purposes of the backfill are to :

- inhibit the development of preferential groundwater flow paths
- buffer the pH/Eh regime of the repository near-field environment
- provide physico-chemical retention of radionuclides
- control mechanical interaction between the geological host and the enclosed waste
- control the dissolution of any gases generated within the vaults
- provide physico-chemical stability to the projected geochemical environment of the repository.
- provide a heat conduction medium in the vicinity of certain categories of waste.

The choice of backfilling material depends on a variety of factors, including :

- geological host formation
- hydrogeological features including groundwater perturbations during the construction and closure of the repository
- layout of the access-ways, shafts and vaults of the repository
- emplacement arrangements of the waste within the repository
- radionuclide inventory and its time-related impact on the geochemical environment
- heat generation characteristics of the wastes
- swelling and consolidation characteristics.

Hence, backfill materials must be designed to cover a variety of purposes leading to the development of 'mix designs' pertinent to their function within a repository. 'Mix designs' are site specific and many countries (eg France, Belgium, Sweden, Switzerland, US and Canada) are now employing the use of underground research laboratories within potential host rock formations. One of their purposes is to obtain information on the desired characteristics of the backfill material and to evaluate placement techniques: for example, injection with or without pressure, air entrainment, gravity filling, rolling, vibration, etc.

The civil and mining engineering industries have wide experience in placing materials to achieve specific performance characteristics. However, further development work is required in order to formulate quality control systems capable of meeting the more stringent requirements of repository backfilling and sealing.

A quality assurance programme in a repository backfilling operation must include :

- controls on the production of the backfill components, eg chemical composition, particle shape and size
- environmental controls on the storage of the individual components prior to mixing
- controls on mixing operations prior to emplacement
- controls to be exercised during the emplacement operation
- monitoring after completion of the emplacement operation.

Effective quality control procedures are likely to be based on index measurements which correlate with relevant engineering performance characteristics, e.g. particle size distribution, density, moisture content, porosity, void ratio, viscosity, etc. It is thought that simple mass-balance relationships will also provide a particularly useful means of regulation.

RESEARCH ON SWELLING CLAYS AND BITUMEN AS SEALING MATERIALS FOR UNDERGROUND REPOSITORIES FOR RADIOACTIVE WASTE

CONTRACTOR	Bullen and Partners Consulting Engineers 188 London Road Croydon CR9 1PT England
CONTRACT	FI 1W - 0162 - UK
DURATION OF CONTRACT	From February 1988 to July 1989
PERIOD COVERED	February 1988 to January 1989
PROJECT LEADER	Dr JA Allison

A OBJECTIVES AND SCOPE

Work previously carried out within the CEC's research and development programme indicates that swelling clay together with bitumen could be used to form a highly effective waste containment barrier for use in deep underground radioactive waste repositories.

This project seeks to identify relevant material properties and sealing mechanisms. Its objectives are :

- o To assess the potential behaviour of combinations of swelling clays and bitumen, and the potential effectiveness of synergistic combinations of these materials for sealing underground repositories against groundwater ingress, radionuclide release and gas release.

- o To assess the potential level of confidence in the long-term behaviour of such seals in the perspective of the quality assurance procedures that could be associated with the emplacement of backfill and seal materials.

B WORK PROGRAMME

The work programme consists of the following activities :

1. A review of available information on properties and behaviour of swelling clays (including bentonite and magnesium oxide) and information on bitumens as used in engineering structures.

2. Control tests on samples of swelling clays and bitumen to establish the range of compatibility between clay swelling properties and the rheological properties of bitumen.

3 Construction and operation of a test rig to monitor water uptakes and swelling pressures in combinations of swelling clay and bitumen.

4 Examination of combinations of the materials to establish whether seals are formed and the nature of the seals.

5 Calculation of the water, radionuclide and gas permeabilities of the seals: with particular reference to the radionuclides iodine, technetium and neptunium and to the gases hydrogen, methane and carbon dioxide.

6 Assessment of test results in the context of material properties and free swelling space that could be achieved in practice with current quality control systems in potential repositories.

C PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

A literature survey has been conducted into the physical and chemical properties of the candidate materials, with particular emphasis on the swelling pressures generated by confined compacted bentonite blocks, and the rheological behaviour of bitumen under sustained compression. Little information is available concerning the swelling properties of magnesium oxide and further enquiries are being made.

Samples of all candidate materials sufficient to satisfy the test programme have been obtained. Lack of detailed information about bentonite testing apparatus and discrepancies in reported test methods and results, has led to the manufacture and development of apparatus especially for this study. Control testing of compacted bentonite is approximately 50% complete.

Conventional tensile test methods for bitumen as used in the road pavement industry were found unsuitable. In consequence, casting and trimming techniques have been developed to produce true cylinders for compressive tests. Prismatic holes and slots have been successfully formed in bitumen specimens. Control testing of bitumen is approximately 20% complete.

A pilot test conducted on combined cylinders of bentonite and bitumen containing a preformed imperfection has been completed with promising results.

PROGRESS AND RESULTS

1 Literature Review

Bentonite - Differences occur in reported values of swelling pressure for similar bentonites and these are attributed to differences in the apparatus and method of pressure measurement. However, insufficient information is provided for the apparatus and test procedures to be replicated. The literature has also been researched to establish the influence of the following specimen variables on equilibrium swelling pressures :

- sample thickness
- dry density
- elastic and creep rebound movements after compaction
- direction of irrigation (top/bottom/both)
- irrigation water pressure
- free expansion movement permitted in the swelling chamber.

Due to scarcity of detailed published information, it has been found necessary to expand the control testing of bentonite to examine the influence of the above variables.

Bitumen - The use of bitumen as a building material, especially for road construction, is well documented and a series of tests are in commercial use to characterise its behaviour for these purposes. Unfortunately, these rely on penetration and tensile properties and are unsuitable for determining rheological behaviour and self annealing mechanisms in compression.

Magnesium Oxide - Indirect reference has been found to work conducted in Canada for KBS in 1979-80 using magnesium oxide in combination with silica sand as a potential repository lining. Further enquiries concerning the swelling behaviour of magnesium oxide are being pursued.

2 Control Testing of Candidate Materials

Bentonite - Samples of powdered natural sodium bentonite (MX-80), activated sodium bentonite (Fulgel) and natural calcium bentonite (Surrey Powder) have been obtained. Relationships between weight of material, compacted density and specimen thickness have been determined, to facilitate the production of 50mm diameter specimens of predetermined thickness and differing densities.

Fully confined swelling tests on highly compacted MX-80 sodium bentonite indicate maximum axial pressures of up to 30 MPa for samples of 15mm thickness. Fulgel (activated sodium bentonite) is found to generate similar pressures, whereas those generated by Surrey Powder are about 20% of those developed by the other two types.

The influence of time delay between compaction and commencement of irrigation has been examined. Instantaneous elastic rebound is found to be about 10% and creep rebound about 1% of sample height respectively. Creep rebound is negligible after 24 hours. Restraint of creep movement, after permitting elastic rebound, generates negligible loading.

The irrigation route (via top, base or both) was found to have negligible effect on the final swelling pressure. However, time to equilibration is significantly reduced where both ends of the specimen are irrigated. Increased porewater backpressure was also found to speed up equilibration without affecting the final swelling pressure. The influence of specimen height is currently being investigated, and tests to study the influence of the other variables are in progress.

Bitumen - Lack of published material on the rheological properties of bitumen in compression and its ability to anneal has necessitated the development of handling, casting and trimming techniques in order to obtain true cylindrical specimens of 38 and 50 mm diameters. Samples of bitumen grades R85/40, R105/35, R75/30, R85/25, R115/15, H80/90 and H100/120 (to British Standard 3690 Pt 2) have been obtained. Difficulties have been encountered in casting these into small cylinders free from entrapped air.

Temperature control is particularly critical and has to be sufficiently high to permit pouring without risk of overheating to cause chemical alteration due to loss of volatiles. As the small size of the specimens promotes rapid solidification, a progressive casting technique has been developed to overcome this difficulty. Removing specimens from moulds and casting in prismatic bars to form inclusions has required experimentation with different types of release agent.

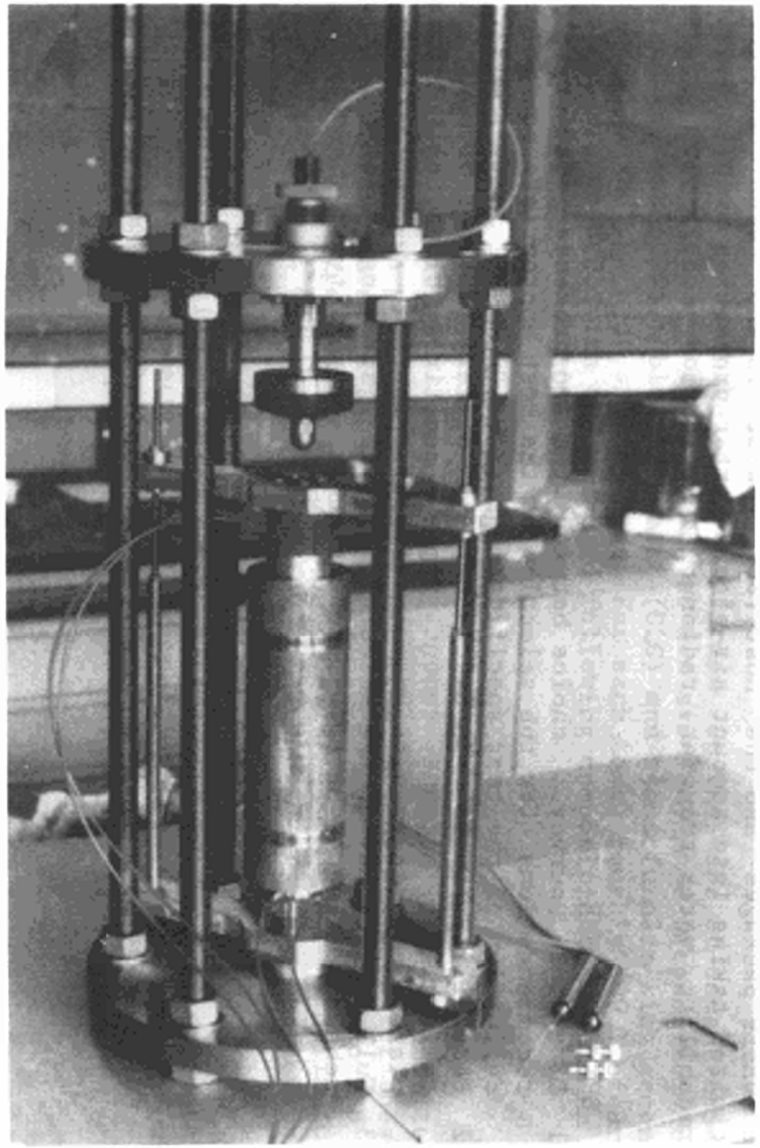
Creep tests using modified oedometers and unconfined compression tests on cylindrical specimens are in progress. These are being used to determine maximum loadings and load/deformation behaviour, to identify ductile or brittle failure modes, and to assist in the matching of bitumen and bentonite grades. Pilot annealing tests have just commenced on laterally confined specimens containing either triangular or long rectangular slots. The former represents edge damage to a bitumen block, whereas the latter simulates an imperfect vertical joint between adjacent blocks as built into a repository wall. Techniques have yet to be developed to identify closure of the imperfection.

Magnesium Oxide - The test programme on magnesium oxide has been held in abeyance until further enquiries are completed.

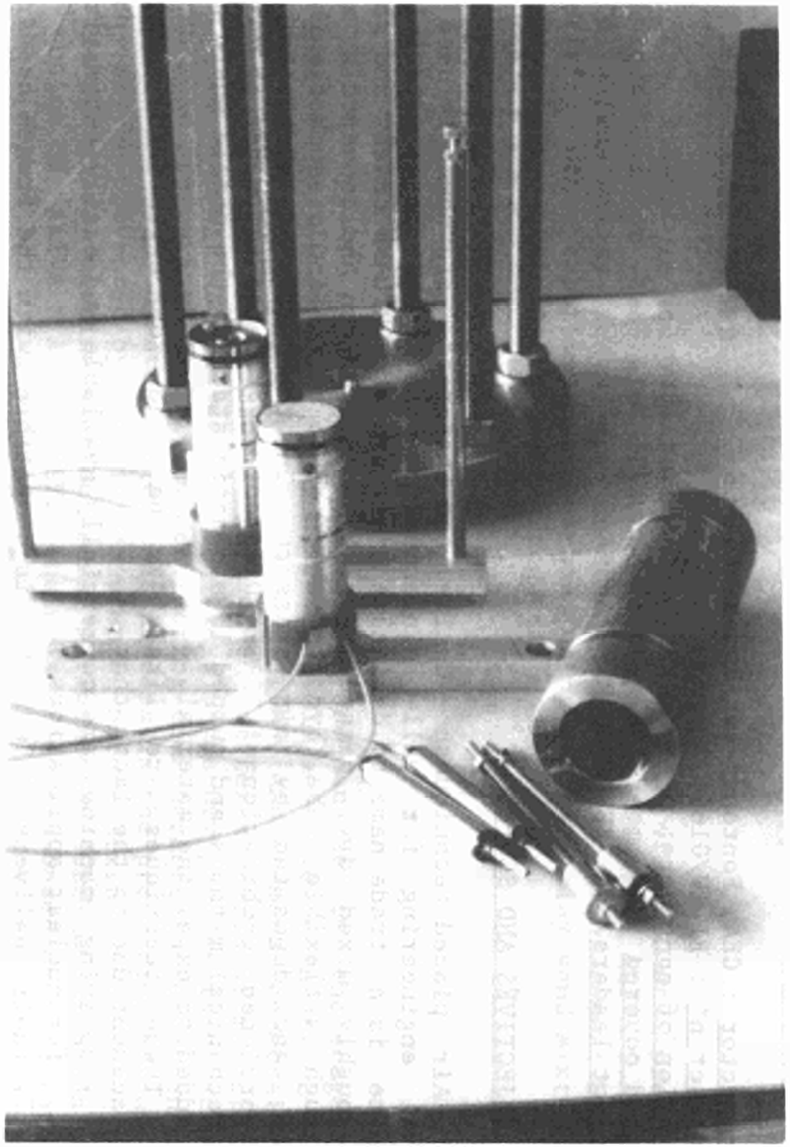
3 Construction and Operation of a Test Rig

Two test rigs have been constructed and are in use to determine the swelling pressures developed by compacted bentonite on irrigation. Water uptake was found to be instantaneous and occurs whilst de-airing is in progress. Release of air trapped in valves and fittings has also been problematic during testing, and modifications are in hand to overcome this problem. Manufacture/construction of two further rigs is in progress to bring the total number of rigs to 4. Photographs of a completed test rig are appended.

SPECIAL APPARATUS FOR TESTING BENTONITE AND COMPOSITE BITUMEN/BENTONITE SAMPLES



(A) ASSEMBLED APPARATUS



B) PISTONS, CONFINING CYLINDER AND DISPLACEMENT TRANSDUCERS (LOADING FRAME AT REAR)

EMPLACEMENT FEASABILITY OF OPTIMIZED AIR PLACED MORTARS

Contractor : CEA, Fontenay-aux-Roses, FRANCE
Contract n° : FI 1W/0166
Duration of contract : October 1987 - December 1989
Period covered : December 1987 - December 1988
Project leaders : A. BERNARD, R. ATABEK

A. OBJECTIVES AND SCOPE

Air placed techniques - Gunite and Shotcrete - are commonly used in civil engineering for wall reinforcement with cement based materials. Gunite is a trade name to designate a mixture of PORTLAND cement and sand thoroughly mixed dry, passed through a cement gun and conveyed by air through a flexible tube, hydrated at a nozzle at the end of such flexible tube and deposited by air pressure. In the case of shotcrete, a proportioned combination of PORTLAND cement, aggregates and water is mixed by mechanical methods and pumped in a plastic state to the nozzle where air is added to expel the material.

These techniques are likely to be used for engineered barrier emplacement due to the facts that :

- spraying machines are commercially available and easily automatized for nuclear applications,
- their delivery (≅ 10 m^3/h) is compatible with the french needs of gallery filling up.

The research program, developed within the framework of this contract, is devoted to test air placed mortar ability to fill up the voids between the waste packages and the host rock. Materials and techniques will be optimized taking into account air placed mortar properties such as density, permeability, water transfer, radionuclide retention, etc...

B. WORK PROGRAMME

2.1 Literature survey : choice between the two processes (dry or wet) ; recommendations for the selection of the most appropriate equipment ; definition of the test specifications.

2.2 Feasability tests : choice between different types of materials, taking into account cement types, additives (clays, silica fume), plasticizers, aggregate granulometry distribution.

2.3 Full scale study of the selected material : spraying cycle definition, rebound influence on air placed material homogeneity.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The literature survey allows the selection of the dry process technique and of two air placed machine manufacturers : ALIVA S.A and MEYCO. Feasability tests are undertaken, starting with cement-based materials. Other experiments are planned using swelling clay-sand mixtures and materials containing salt as aggregates.

Progress and results

2.1 Literature survey

The air placed techniques differ from one another in the stage at which water is introduced ; the mixture could be conveyed in dry or wet state (Table I). Taking into account the emplacement constraints (distance : 300 m, flow : 10 m^3 /h) and the different types of materials (hydraulic binders as well as swelling clays), the dry process is selected as the most appropriate technique, considering the characteristics given in Table II. The main disavantage of this process is the production of dust, which may be prevented by material moistening before mixing. The inventory of the air placed machine manufacturers, performed by SOLETANCHE, heads to the choice of ALIVA S.A and MEYCO equipments mainly based on a) a rotor machine (Figure 1), b) a compressor, c) metallic pipes and d) a nozzle (Figure 2).

2.2 Feasability tests

Feasability tests have been performed in the case of cement-based materials. Six different types of mortar are studied, made respectively of Portland cement (OPC), blended cement (CLC) and blast furnace slag cement (CLK), with and without silica fume. For each test, roughly 500 l of dry materials are needed, with the standard following composition :

Sand (0/4 mm)	≅ 1000 kg
Cement	≅ 300 kg
Silica fume	≅ 0 or 15 kg

MEYCO equipment with a rotor machine of 12 cells, a 50 mm in diameter and 20 m in length pipe and a compressor of 25 m^3/mn were used for these first feasability tests. The different mixtures were deposit into 150 l boxes (A.F.T.E.S type) specially designed to allow the core-sampling of four cylinders, 11 cm in diameter and 22 cm high. The nozzle flow was estimated between 1 to 2 m^3/hour.

The main properties of the air-placed materials (mechanical strength, permeability porosity, water diffusion coefficient) will be compared, leading to the choice of the best mixture for full scale experiment.

TABLE I : DIFFERENT TYPES OF AIR PLACED TECHNIQUES

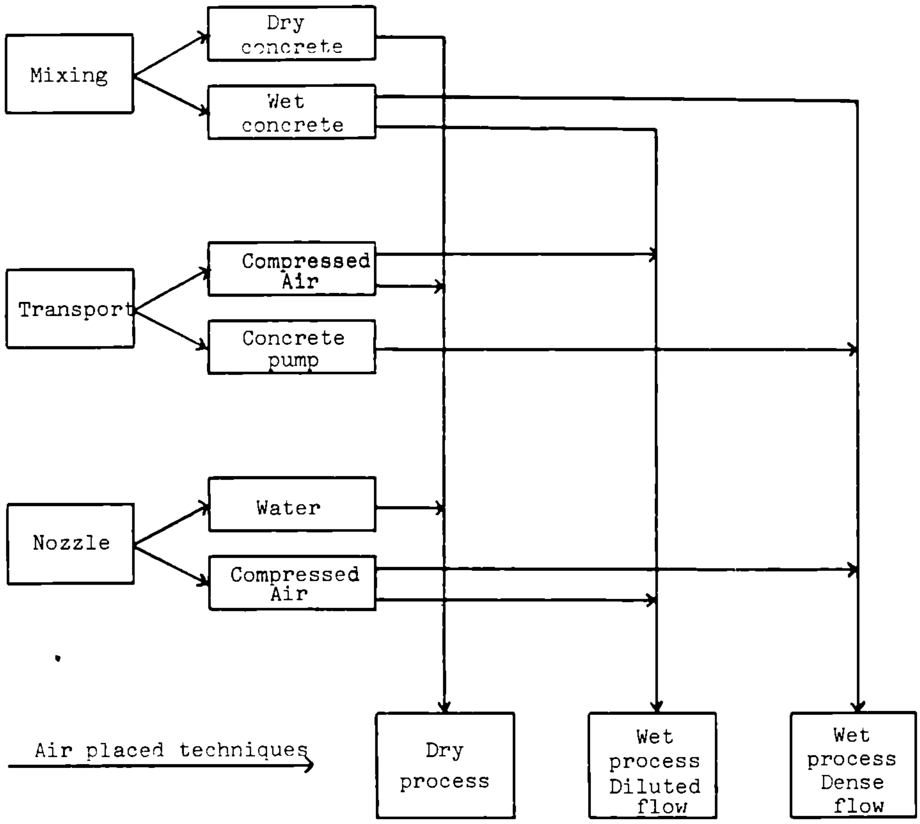


TABLE II : CHARACTERISTICS OF THE AIR PLACED TECHNIQUES

	Dry Process	Wet process Diluted flow	Wet process Dense flow
Transport distance	***	o	*
Delivery	***	o	***
Compaction	***	**	**
Dust	o (*)	**	**
Maintenance Cleaning	***	o	o

o Pass
* Average
** Fairly good
*** Good

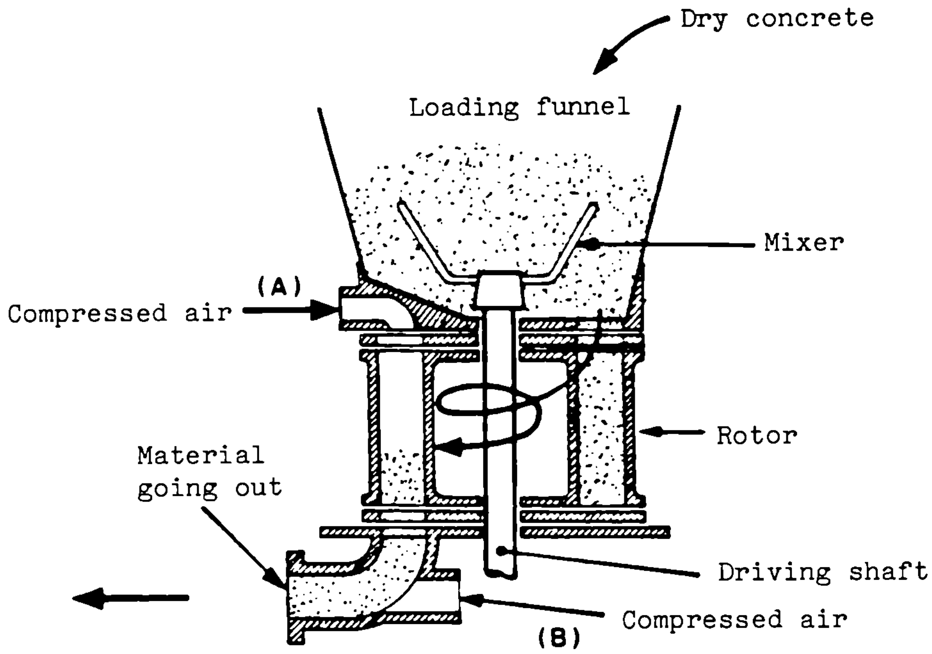


FIGURE 1 : Rotor machine
ALIVA equipment

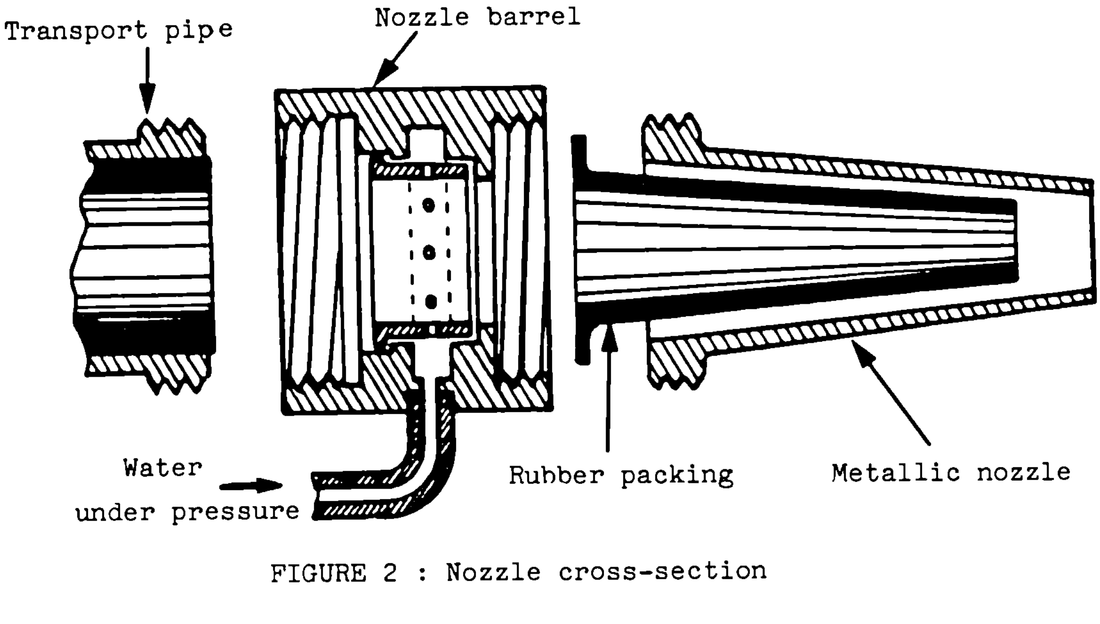


FIGURE 2 : Nozzle cross-section

UNDERGROUND LABORATORY VALIDATION OF VITRIFIED WASTE DISPOSAL IN A GRANITIC MEDIUM

Contractor : CEA, Fontenay-aux-Roses, FRANCE
Contract n° : FI 1W/0207
Duration of contract : August 1988 - March 1990
Period covered : August 1988 - December 1988
Project leader : M. JORDA

A. OBJECTIVES AND SCOPE

The objective of this research is the underground validation of the plugging back of a vitrified waste disposal pit in a granitic medium. The scenario selected, which is similar to that adopted in other studies carried out by the European Community, involves the stacking of twenty vitrified packages in a pit 30 m deep and 1 m in diameter.

The engineered barrier between the waste and the granite is composed of clay material, in the form of high density, compacted elements. Gaps are required to enable the lowering of the packages and of the barrier into the pit, but these gaps have the effect of reducing the density of the material introduced into the pit.

The uncertainty existing with regard to the definition of the minimum size of the residual gaps required for satisfactory installation can only be removed by performing an in-situ test. A demonstration pit will therefore be made in the mining region of Fanay-Silord, using state-of-the-art drilling technology, and then the pit will be plugged with the engineered barrier and the simulated glass containers.

B. WORK PROGRAMME

2.1 - Sinking of two demonstration pits, 1 m in diameter and 30 m deep, using the raise boring technique (one of the pits will be used as a reserve)

2.2 - Detailed characterization of one of the two pits bored, with verticality check and checks for surface condition, circularity faults and diameter variations

2.3 - Plugging back of the characterized pit using baskets filled with blocks of compacted clay, followed by testing of the installation of the glass containers.

C. PROGRESS OF WORK AND RESULTS OBTAINED

State of advancement

The barrier installation studies which form the basis of this research require the boring of a demonstration pit of 30 m depth minimum. Work authorisations have been granted by the local authorities of the mining region of Fanay and boring work proper will start in April 1989.

Improvements in manufacturing techniques for the uniaxial compacting of bricks of the type which make up the barrier should enable an increase in the pit filling rate.

Progress and results

2.1 Boring of the demonstration pit

The tests will be carried out at the mining center of Fanay, near Razes in the Haute Vienne Department (Silord Site). Figure 1 shows a detailed view of the underground site selected. The site is separate from the mine, with accesses via galleries a) to the upper level and b) to the lower level 40 m below the upper level.

The upper level where the CEA work will take place, is enlarged and arranged so as to enable :

- the boring of two holes of a diameter of 1 m. The holes will be spaced 4 m apart, center line to center line.
- about ten visitors to be received in order to monitor the installation tests under acceptable conditions.
- storage of about 20 dummy containers without hindering the free passage of personnel, the containers being 0.43 m in diameter and 1 m high approx.
- installation, over the holes, of a 3 tonne winch for the baskets and the containers.
- performance of the installation tests.

Primary aeration of the work gallery is provided by a 1 m dia. shaft leading to the surface. The distance between the surface (entrance to the shaft) and the test gallery (upper level) is 250 m. Figure 1 also shows the location of the niche which will be used by ENRESA for its own studies.

The last quarter of the year 1988 was given over the compiling of the administrative dossier, required to obtain the different authorisations for the boring work.

2.3 - Back plugging of the pit - fabrication of the compacted clay blocks

The compacted blocks are made from powdered material (of optimised particle size) and form the barriers which are installed using baskets. The technique used is compacting by uniaxial pressing. An assembly of six bricks (figure 2) forms a crown piece which does not require any further finishing work. Design of the mould used and of the compacting techniques have been improved, with

1) Modification of the rate of descent of the upper piston and de-aeration of the powder
2) Height check of the pressed parts
3) Improvement of the mould release techniques.

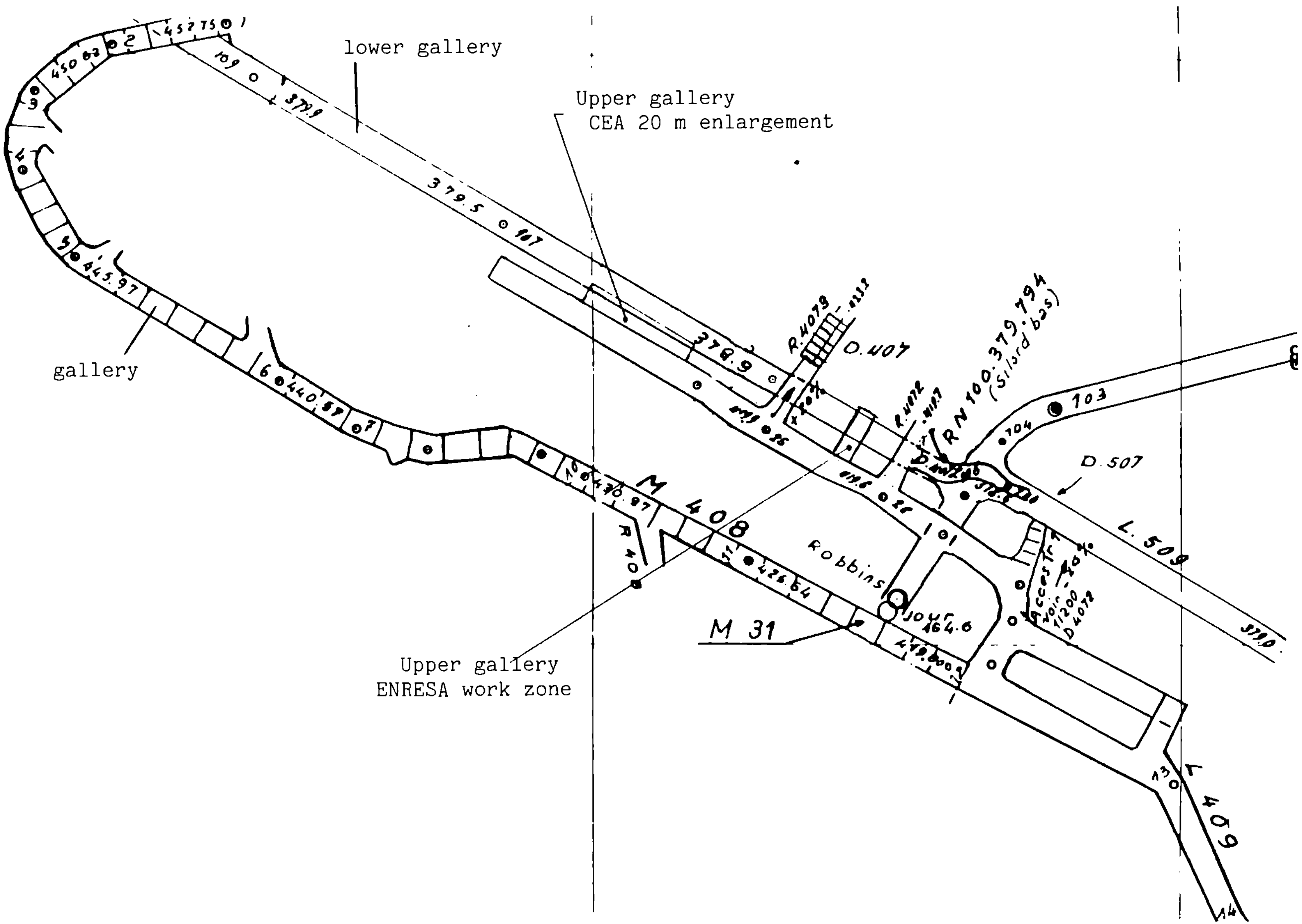
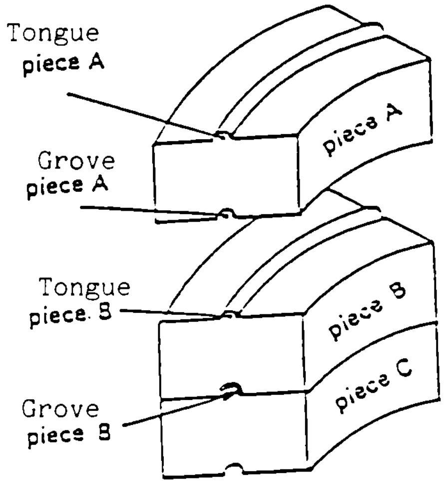


FIGURE 1 : SILORD site

FIGURE 2 : Assembly of compacted bricks



FIELD TEST FOR DEMONSTRATION OF EMPLACEMENT FEASIBILITY OF CLAY BUFFER MATERIALS AS ENGINEERED BARRIERS IN GRANITE

Contractor: ENRESA, Paseo de la Castellana 135, Madrid (E)
Contract No.: FI1W/0231
Duration of contract:
Period covered:
Project leader: C. del Olmo

Progress report not yet available.

4.3. RADIONUCLIDE MIGRATION IN THE GEOSPHERE (MIRAGE)

4.3.A. Actinide and fission product geochemistry in natural acquifer systems

Characterisation of the Boom clay and its multi-layered hydrogeological environment with a view to radionuclide migration

Contractor : SCK/CEN, Mol (B)
Contract n° : FI1W/0055/B
Duration of contract : October 1986 to December 1989
Period covered : January 1988 - December 1988
Project leader : A.A. Bonne

A. OBJECTIVES AND SCOPE

The effectiveness of the geological barrier is a key function for the long-term safety of the disposal of nuclear wastes. The assessment of its barrier performances can only be done by modelling, provided that reliable in situ data is available.

The underground experimental HADES-laboratory in the Boom clay at Mol allows to sample the clay according to various procedures and to perform experiments aiming at characterizing the clay in situ and at determining in situ migration parameters. The hydrogeological observations and sampling network around the site enable to verify if the local site data is in agreement with the regional groundwater flow regime and hydrochemistry. Research previously carried out has shown that organic matter and compounds in clay are of prime importance for trapping various radionuclides in the argillaceous barrier. The characterisation of the organic substances in the Boom clay, their specific retention capabilities and stability with regard to irradiation are therefore of particular interest in the near-field. A collaboration in this matter with the University of Louvain (KUL, Prof. Cremers) is on-going.

The research is backed by migration tests simulating in situ conditions (diffusion experiments on reconsolidated clay plugs) and by hydrogeological studies, both not being part of the contract, but the results of these will be made available for sake of confirmation or completion.

B. WORK PROGRAMME

1. Study of the organic substances in the Boom clay, with emphasis on their affinity for Eu and Tc.
2. In-situ short-term migration experiments in the Hades-URL
3. Application of isotopic techniques and hydrochemistry for the characterisation of a multi-layered aquifer system

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The characterisation of the organic substances in the Boom clay and their retention specificities continuing on the line of the research on that matter already started in the previous multi-annual programme. For the in-situ migration experiments, emphasis was on a precise understanding of the very local hydraulic conditions of the test. For the various possible experimental set-ups the mathematical formulations have been worked out. Several in-situ migration experiments are running and a few have been completed. Carbon-14 groundwater dating is continued on the line of the previous work.

Progress and results

1. Study of the organic substances in the Boom clay, with emphasis on their affinity for Eu and Tc.

Since no method could adequately be devised to measure the diffusion parameters of small humic molecules in the Boom clay pore liquid, measurements were carried out on substitute molecules like sucrose and lactose ($M.W^t$ 343), phenylalanine ($M.W^t$ 169) and triiodothyronine varying the consolidation pressure applied to the clay. The general conclusion is that a flux of actinides carried by the small humic acetors could be characterized by a product RD_a equal to 5.10^{-8} cm^2sec^{-1} in Boom clay under in situ conditions.

In parallel, migration tests including forced convection, were conducted from the underground laboratory, obtaining results for Eu and Sr. For the last element the influence of convection cannot be detected in spite of a Darcy velocity exceeding the natural conditions by a factor 12000. For Eu, two successful tests have clearly indicated an irreversibility in the exchange of Eu between mobile and immobile humic complexing molecules.

The possibility of exchanging Eu between small and large humic molecules has been the object of a separate laboratory investigation, and has led to the conclusion that weeks may be needed to reach an equilibrium distribution of Eu in an homogeneous solution. The kinetics of exchange may be less favourable still in a compacted clay. In conclusion, "accelerated" migration tests reveal features of the retardation mechanisms but do not directly provide migration parameters suitable for safety assessments.

The Tc reduction by Boom clay porewater and the subsequent distribution of Tc on the humic material has been investigated by Gel Filtration Chromatography. In absence of a solid phase the reduction step of Tc is extremely irreproducible. However tow facts have emerged :

- the organic complexation is not quantitative ; inorganic Tc (not TcO_4^-) remains in the solution ;
- the fraction of Tc present in the porewater shows a definite preference for the smallest humic molecules.

In the long run though, the humic complexation may well not constitute the ultimate sink for Tc. A brief examination of the system FeS_2 - HA - Tc indicates that ultimately all the Tc could well end up on the surface of the micropyrite in the clay. TcO_2 is excluded as too soluble. The formation of TcS_2 on th pyrite surface is however thermodynamically favourable.

2. In-situ short-term migration experiments in the HADES-URL

Two types of in-situ migration experiments are actually performed in the underground laboratory : percolation experiments with labelled clay cores emplaced in self-sealing boreholes and direct injection of tracers in the clay formation.

The main purpose of the percolation experiments is to verify the laboratory data for a few isotopes in conditions closely approaching a repository far-field environment. The availability of real porewater constitutes a definity advantage for this type of "in-situ" experiments.

Results obtained with Sr-85 are in close agreement with those from purely diffusive laboratory experiments in consolidation cells ($D = 6{,}9 \times 10^{-8}$ cm²s^{-1} versus $4{,}6 \times 10^{-8}$ cm²s^{-1}) /1/. A very small fraction of europium is mobile under the conditions of the test and is almost not retarded ($D = 1{,}5 \times 10^{-7}$ cm²s^{-1}) /2/ while the bulk of the tracer behaves as a concentration source. Laboratory studies have been initiated in order to check the existance of a mobile europium species. Problems associated with experiments in self-sealing boreholes are related to :

- time lag between emplacement and porewater flow ;
- complexity of the experiments : drilling of a borehole for emplacement of the experiment and overcoring for its retrieval.

In view of overcoming the above-mentioned problems, a borehole has been equipped with a stainless steel receptacle for retrievable (re-changeable) labelled clay cores. Testing of this much simplified concept is actually underway.

Due to the strong retardation and the low hydraulic conductivity of the Boom clay, migration experiments with adsorbed tracers are necessarily limited to very short distances in the clay. For the validation of the transport model in the Boom clay at greater distance, large scale 3-dimensional in-situ experiments with non-retarded tracers have been started. On 20.01.1988, one gigabecquerel of HTO has been injected in filter No. 5 of piezonest CP1. The purpose of this experiment is to monitor the migrating tritium by periodic sampling at the neighbouring filters situated at one meter intervals from the point of injection. After a period of several years the tracer is expected to be detected in the two adjacent filters on both sides of filter 5. At this time the tritium will have diffused in a sphere with a radius of 2 m (volume = 33,5 m³), which means an upscaling of 10^6 compared to laboratory experiments run on clay cylinders with a diameter of 3,8 cm and 3 cm in height (volume = 34 cm³).

3. Application of isotopic techniques for the characterisation of Boom clay porewater

The dating of clay porewater by carbon-14 measurements is progressing very slowly but this is inherent to this type of work. Preliminary results obtained on porewater samples abstracted from piezometer SSW situated in the silty layer at -257 m gave positive ^{14}C values as measured by LSC (Liquid Seintillation Counting). In view of establishing a ^{14}C profile as a function of depth, a series of improved piezometers were installed at different horizons in the clay. After developing a procedure for the preparation of small amounts of CO_2 from porewater, several samples were sent to ETH-Zürich for ^{14}C measurement by TAMS.

The preliminary positive ^{14}C results for SSW could NOT be confirmed and also the samples from the higher horizons in the clay yielded ^{14}C-contents below the detection limit. The initial high values obtained from SSW are probably due to contamination induced during drilling and piezometer construction. During drilling of a borehole the clay is inevitably exposed to modern carbon ! All recent available data disprove the existance of a measurable carbon-14 content in the clay porewater.

Sampling is actually in progress for ^{36}Cl measurement by TAMS at ETH-Zürich. A procedure has to be developed for the preparation of pure AgCl samples. The sulphur content in these samples should not exceed a few ppm for reason of interference with the isobar ^{36}S. This problem of potential sulphure contamination is very important for Boom clay porewater as its chlorine content (± 25 ppm) is lower than the DOC (± 100 ppm) containing several percent of sulphur. An analytical method for controlling the ^{36}S content in AgCl has to be found before the samples can be analysed for ^{36}Cl by TAMS.

PUBLICATIONS

P. HENRION, M. MONSECOUR, A. FONTEYNE, M. PUT and P. DE REGGE
Radioactive Waste Management and the Nuclear Fuel Cycle
Volume 6 No. 3-4, pp. 313-359, (1985)

M. MONSECOUR, A. FONTEYNE, M. PUT, H. YOSHIDA
In-situ experiments in the Boom clay at Mol
12th Int. Symp. on the Scientific Basis for Nuclear Waste Management, Berlin, Oct. 10-13, (1988)

The Role of Organics in the Migration of Radionuclides in the Geosphere

Contractor: Risø National Laboratory, DK
Contract N°: FI1W/0066
Working Period: july 1986 - december 1989
Project Leader: Lars Carlsen

A. Objectives and Scope:

A review on the possible role of organics species in the ground water on the migration behaviour of radionuclides in the geosphere is required. Considerable amounts of data are available. They are, however, rather scattered throughout the literature.

An experimental study as well as theoretical considerations on the influence of organic complexing agents on the sorption, and hence migration behaviour of radionuclides are of fundamental interest in attempts to evaluate the possible transport of released radioactive waste with ground water.

Characterization of naturally occurring organics, e.g. humic and fulvic acids is of general interest, due to the omnipresence of these polymeric species in the terrestrial environment and to their known complexing abilities towards metal ions.

B. Work Programme:

B.1. Review of available literature on the influence of organics compounds including experimental and theoretical data obtained at Risø.

B.2. Batch-type experiments to elucidate the influence of organics on radionuclide migration.

B.3. Column-type experiments to elucidate the influence of organics on radionuclide migration.

B.4. Theoretical study to elucidate the effect of complex formation on radionuclide migration.

B.5. Characterization of humic acids partly within the frame of the joint european programme on humic acid characterization.

C. Progress of Work and Obtained Results

Summary.

The review paper on the role of organics on the migration behaviour of radionuclides in the terrestrial environment has been completed and will be published by the Commision in due course.

The theoretical study on the influence of complexation on radionuclide migration has been completed. The work has been extended compared to the originally proposed, and the results will be published separately in *Waste Management*. The results are furthermore incorporated in the above mentioned review paper.

The laboratory has during 1988 invested considerable effort in participating in the joint european intercomparison and characterization study of humic acids.

Progress and Results

1. Review (B.1.)

The review report summarizes factors of importance concerning the role of organics on the migration of radionuclides in the terrestrial environment.

Following some introductory remarks (*chapter 1*), *chapter 2* describes the occurrence of organic compounds in the geosphere, taking both naturally occurring as well as artificially introduced compounds into account. The behaviour of organic compounds in the terrestrial environment is summarized (*chapter 3*) with special emphasis on sorption and persistence in the environment. Both chemical and microbiological degradation reactions are discussed. *Chapter 4* describes the fundamentals of complex formation in relation to migration. Stability constants for the interactions between relevant metal ions and low molecular weight ligands as well as with humic- and fulvic acids are summarized. *Chapter 5* is devoted to an evaluation, based on theoretical considerations, of the influence of organics on the migration behaviour of radionuclides in the terrestrial environment. Additionally, the behaviour of two special elements, *i.e.* technetium and iodine, is discussed. An overall summary is given in *chapter 6*.

The report contains a comprehensive list of references.

2. Theoretical work (B.4.)

The results of the theoretical investigations has been in detail been reported in the paper *The influence of complexation on radionuclide migration. A theoretical study.*

The paper describes a theoretical evaluation of the influence of complexation on metal ion, *e.g.* radionuclide migration in environments containing an excess of complexing agents, *i.e.* the equilibrium between the free and the complexed metal ions can be regarded to follow pseudo first-order kinetics. It is shown that as long as the rate of interconversion between the free and complexed metal ions is rapid relative to the residence time in the system studied, the two species will migrate with the same speed, controlled by an "effective retention factor". It is clearly demonstrated that approaching zero complexation the effective retention approaches that of the more retarded species (the free metal ion), whereas the effective retention approaches that of the less retarded species (the complex) for increased complexation. The implication for the distribution of radionuclides in the terrestrial environment is discussed.

The described calculations unambiguously demonstrate the influence of complexation on the migration of metal ion in the terrestrial environment. Increasing the apparent stability constant of the complex, either as a results of high true stability of the complex or by increased ligand concentrations, significantly decrease the effective retention of the equilibrium system. Hence, it is concluded that the naturally occurring organic ligands, i.e. humic- and fulvic acids, which form strong to very strong complexes with metal ions, apparently must be expected to exhibit a pronounced influence on the migration behaviour of radionuclides, leading to an increased migration speed. Similarly the possible presence of artificially introduced potent ligands, as e.g. EDTA, forming stable complexes with most polyvalent metal ions, certainly should be avoided, due to the dominating role such compounds, even in very minor concentrations, will play in controlling the migration of the metal ions.

3. Humic Acid Characterization (B.5.)

The work in this area has been concentrated on the study of europium - humic acid complexation. The complexation of europium with different humic acids has been studied based on the conventional ion-exchange technique. The originally proposed dialysis-approach appeared less advantageous due to the fact the the different humic acids contained low molecular weight fractions, which could pass through the dialysis membrane.

Humic acids of different origin were studied: a) commercially available (Aldrich) humic acid and b) Humic acids isolated from Gorleben ground water. Further humic acids from Fanay Augeres and possibly from Mol will be studied.

The studies were carried out in 0.1 molar NaCl solutions. Surprisingly it was found that nearly pure 1:2 complexes were formed, in contrast to the expected 1:1 complexes, as reported by other groups. The two studied humic acids both form rather stable complexes with europium. The logarithmic interaction constants were found in the order of 5, the ligand concentration being expressed in g/L.

The work on humic acid characterization based on isotope-exchange has not yet been initiated.

ACTINOIDE MIGRATION PHENOMINA IN GROUNDWATER: COLLOID GENERATION AND COMPLEXATION WITH NATURAL ORGANICS

Contractor: Institut für Radiochemie, Technische Universität München

Contract No.: FI 1W/0067

Duration of contract: Sept. 1986 - Dec. 1989

Period covered: 1. Jan. 1988 - 31. Dec. 1988

Project leader: J.I. Kim

A OBJECTIVE AND SCOPE

Important geochemical processes that govern the migration of actinides in deep geological aquifer systems are: hydrolysis reaction, redox reaction, complexation with inorganics as well as organics and colloid generation. The colloid generation and complexation with natural organics, e.g. humic substances, appear to be the most significant geochemical phenomena with regard to actinide migration in a variety of aquifer systems.

The contract research deals, therefore, with the colloid generation of representative actinides in different groundwaters and the complexation of Am and Pu with natural organics, particularly humic substances. The results are expected to give an insight into the migration mechanisms of actinides in the geosphere.

B WORK PROGRAMME

B 1. Actinide colloid generation in groundwater

- Characterization of colloids
- Generation mechanisms of actinide pseudocolloids
- Quantification of colloid generation in a migration medium for actinides

B 2. Actinide complexation with natural organics

- Characterization and complexation study
- Humic substances as organometallic colloids
- Mobility of complex species and colloids in aquifer systems
- Quantification of actinide mobility

B 3. Interlaboratory comparison exercise on complexation with natural ligands (COCO-group: TUM, CEN/SCK, KUL, CEA-FAR, Risø, BGS, JRC-Ispra and other new members)

- Intercomparison of characterization methods
- Separation and production of natural humic acids present in the reference sites
- Intercomparison of stability constants

C PROGRESS OF WORK AND OBTAINED RESULTS

Statement of advancement

For the complexation study in the frame work of COCO-Club programme, a commercial humic acid from ALDRICH Co. (Aldrich-HA(Na^+)) has been purified, protonated and characterized, which is then taken as a reference humic acid (Aldrich-HA(H^+)) in the programme. The humic acid extracted from one of Gorleben groundwaters (Gohy-573), chosen as the first site specific humic acid (Gohy-573-HA(H^+)), is also purified and characterized. These two humic acids are distributed to the COCO-Club members for the interlaboratory exercise of characterization and complexation. From 400 mL pore water concentrate from Boom-Clay in Mol (CEN/SCK), humic acid is extracted, purified and characterized. This is taken as the second site specific humic acid (Boom-Clay-HA(H^+)) in the programme.

The complexation of Am(III) with different humic acids has been studied by UV-spectroscopy, Laser-induced photoacoustic spectroscopy (LPAS) and ultrafiltration. For the purpose of comparison, the humic acid from the Lake Bradford, Florida, USA (Bradford-HA(H^+)) is also included in the complexation study, since this humic acid has been used already by many laboratories.

The colloid quantification has been investigated, using electron micrography as well as light scattering measurements by LPAS. Groundwaters from Grimsel and Gorleben are included in this investigation. The work is still in progress.

Progress and results

1. Characterization of humic acids

A part of results is already given in the first report [1] and further results are to be found in the second report [2]. The results of interlaboratory exercise are presented already in the review paper of the MIRAGE plenary meeting 1988 [3]. In order to facilitate a gross overview, the elemental compositions of investigated humic acids are summarized in Table 1, together with the ranges of values known in the literature [3]. The elemental compositions of Aldrich-HA(H^+) and Gohy-573-HA(H^+) resemble very closely, whereas the values of Boom-Clay-HA (H^+) differ considerably from the two other humic acids.

2. Complexation

The complexation study of Am(III) has been carried out with Aldrich-HA(H^+) and Bradford-HA(H^+) by UV-spectroscopy, LPAS and ultrafiltration. The investigation with Gohy-573-HA(H^+) and Boom-Clay-HA(H^+) is for the moment in progress. The important features observed in this study are as follows: a tridentate complexation of Am(III) with the functional groups of humic acid is predominant, 1:1 or 1:2 complexation is not evident, the complexation constant does not appear varying with

Table 1: Elemental composition of humic acids (in percent) normalized to 100 % of organic components

Element	Aldrich $HA(Na^+)$	Aldrich $HA(H^+)$	Gohy-573-$HA(H^+)$I	Gohy-573-$HA(H^+)$II	Boom-Clay-$HA(H^+)$	Lit. Values [4]
C	48.93 (55.08)	55.23	56.25	57.32	62.36	50 - 60
H	5.13 (4.48)	4.48	4.52	4.76	6.05	4 - 6
N	0.29 (0.33)	0.32	1.69	1.77	2.94	2 - 6
O	43.40 (37.64)	37.64	35.80	35.72	26.98	30 - 35
S	2.20 (2.47)	2.33	1.73	0.43	1.66	0 - 2

( ): Values normalized to the hydrogen and oxygen content of Aldrich-$HA(H^+)$; Gohy-573-HA (H^+)I and Gohy-573-$HA(H^+)$II are two different charges separated from the same groundwater in the different time span.

Table 2: Complexation constants of Am^{3+} with humic acids under different experimental conditions

Humic Acid	pH	I(M)	L	Method	log β (L/mol)
Aldrich-$HA(H^+)$	6.0	0.1	(0.651)	LPAS	6.45 ± 0.03
Bradford-$HA(H^+)$	6.0	1.0	0.518	UV-Spectroscopy	6.28 ± 0.34
Bradford-$HA(H^+)$	6.0	0.1	0.650	Ultrafiltration	6.53 ± 0.30
Bradford-$HA(H^+)$	6.0	0.1	0.650	UV-Spektroscopy	6.20 ± 0.36
Bradford-$HA(H^+)$	5.5	0.1	0.400	UV-Spektroscopy	6.24 ± 0.28
Bradford-$HA(H^+)$	5.0	0.1	0.190	UV-Spektroscopy	6.13 ± 0.35

(L): Loading capacity of humic acid for Am(III); (I): Ionic strength ($NaClO_4$)

increasing pH as indicated in the literature [4] and a loading capacity of humic acid is limited in a given solution but increases with increasing pH and with decreasing ionic strength. The complexation constants of Am(III) determined for Aldrich-$HA(H^+)$ and Bradford-$HA(H^+)$ are given in Table 2.

3. Quantification of groundwater-colloids

The quantifiaction of groundwater-Colloids i.e. size distribution and population, is for obvious reasons an important subject for the migration study of actinides. Much effort has been concentrated on the development of qualification methods, expecially, to establish a non-perturbing spectroscopic method. A new determination method is introduced using laser-induced photoacoustic spectroscopy, which facilitates the measurement of scattered lights directly on a piezocrystall transducer. The time difference in signal propagations between photoacoustic waves and scattered lights enables the quantification of colloid population, provided an average size of colloids is known. This method is further in development. The preliminary quantification results for Grimsel water and a number of Gorleben groundwaters are shown in Table 3. The calibration has been made by measuring Latex solutions of different concentrations with various particle diameters (40 nm ∿ 300 nm). The density of colloids is assumed to be one.

Table 3: Determination of colloid population in groundwaters by LPAS

Groundwater	P.A. Signal (μV/mJ)	Colloid content (ppm)	Population (N/L) (I)	Population (N/L) (II)
Grimsel water •	0.166	0.095	1.7×10^{10}	4.2×10^{11}
Gohy-1011	0.176	0.101	1.8×10^{10}	4.4×10^{11}
Gohy-214	1.445	0.830	1.5×10^{11}	3.6×10^{12}
Gohy-S105	3.175	1.824	3.3×10^{11}	8.0×10^{12}
Gohy-S104	7.645	4.393	7.9×10^{11}	1.9×10^{13}

(I): evaluation based on the average diameter of colloids at 200 nm
(II): evaluation based on the average diameter of colloids at 100 nm

The electron microscopic scanning has been also applied for counting colloid population in Grimsel water. The groundwater is filtered by a Amicon XM50 filter of ca. 3 nm norminal pore size (from Dr. Degueldre, PSI). From counting areas of 2431 μm^2 and 2244 μm^2 in 25 sigments, the colloid population is evaluated to be

$$(0.6 \sim 1.5) \times 10^9 \text{ particle/L}$$

with an average diameter of ca. 100 nm. In comparison with the LPAS determination (cf. Table 3), the electron microscopic counting gives a considerably lower colloid population. The reason is still to be understood. The electron microscopic scanning requires the manipulation of groundwater, i.e. filtration and dryness, and counts a relatively small area of the filter. Such methodical handicaps may result in the low accuracy of measured values. The LPAS method includes measuring of very small sizes of colloids which are for technical reasons neglected in the electron microscopic counting (limitation at < 40 nm Ø). Further comparison of the two methods is still in progress.

A typical size distribution determined by electron microscopic counting is shown in fig. 1. The shaded area represents the total area (2431 μm^2)

of counting and the black columns are from counting of one segment area (187 μm^2) which is then magnified 10 times for particle numbers in this figure. The counting limitation arises in < 40 nm, so that the particles less than this size-limit are neglected in counting. The emission of the smaller particles in this method may be the reason why the colloid population is counted much less.

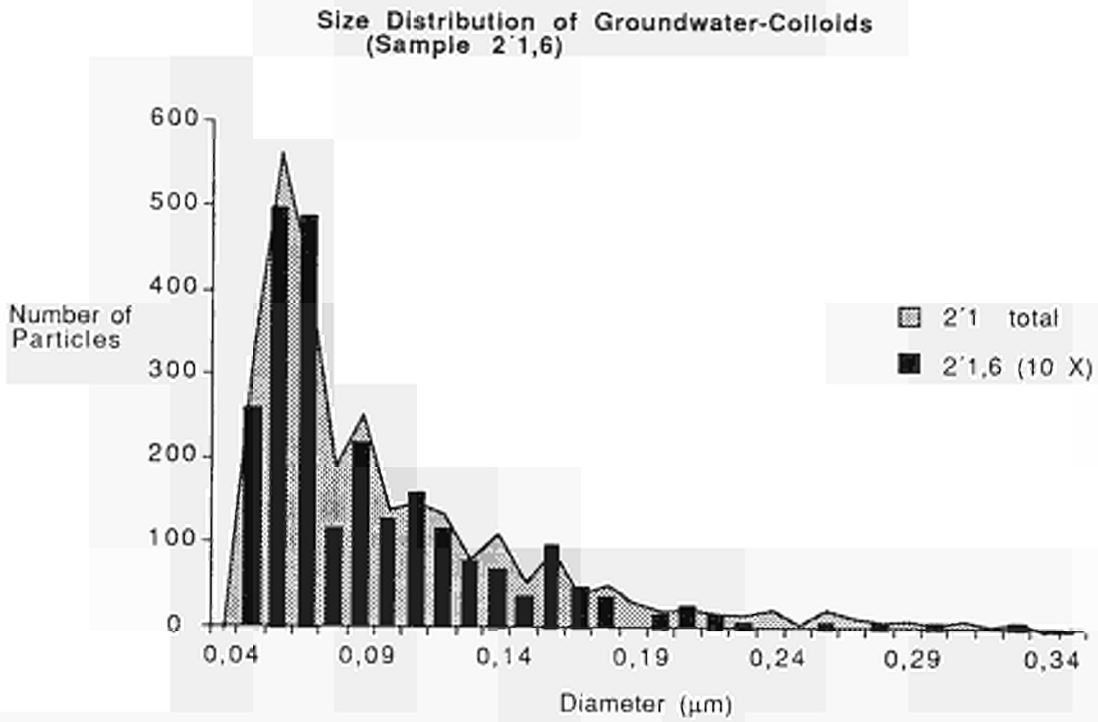


Fig. 1. Size distribution of colloids in Grimsel water determined by electron microscopic scanning.

<u>List of Publications</u>

[1] J.I. Kim, G. Buckau: "Characterization of Reference and Site Specific Humic Acids: Part I", RCM-01588 (1988)

[2] J.I. Kim, G. Buckau: "Characterization of Reference and Site Specific Humic Acids: Part II", RCM-02188 (1989)

[3] J.I. Kim: Geochemistry of Actinides and Fission Products in Natural Aquifer Systems" in EUR 11589 EN (Ed. B. Come) (1988)

[4] Ref. [3] fig. 2

[5] J.I. Kim, G. Buckau, H. Rommel, B. Sohnius: "Transuranium Elements in Gorleben Aquifer Systems: Colloid Generation and Retention Process", Mat. Res. Symp. Proc. (1989) in print

STUDY OF THE INTERACTIONS BETWEEN ORGANIC MATTER AND TRANSURANIUM ELEMENTS

Contractor : CEA-IRDI/DERDCA/DRDD/SESD/SCPCS - FONTENAY-AUX-ROSES - FRANCE

Contract n° : FI1W/0068

Duration of contract : From 1985 to 1989

Period covered : 1988

Project leader : Mme MOULIN, Mr BILLON

A. OBJECTIVES AND SCOPE

The main objective of this programme is to study the importance of natural organic ligands (humic and fulvic acids) as complexing agents of radionuclides in their migration or retention behaviour in the geosphere /1-2/. A second part of this programme is to emphasize the role of the natural colloids (inorganic and/or organic species) present in deep or shallow aquifers, which could have a major importance in the transport of radioelements./2-3/. The goals of this research programme are :

- to define the characteristics and properties of the natural colloids including, particularly the humic substances,
- to determine the formation conditions of organic complexes with radioelements and the interactions with the natural colloids (sorption, complexation, ...).

B. WORK PROGRAMME

B.1. Isolation and Characterization of humic substances

B.1.1. Concentration of humic materials from natural groundwaters.

B.1.2. Characterization of humic materials by physical and chemical methods.

B.2. Interactions betwen humic substances and actinides

B.2.1. Development of the chromatographic method selected for the complexation studies.

B.2.2. Determination of interaction constants by this method. Comparison with other methods, particularly spectroscopy (application and development).

B.3. Studies on natural colloids

B.3.1. Isolation of colloids from natural waters by ultrafiltration : sampling, water analysis, development of the technique.

B.3.2. Characterization of the colloids : size, particle population, composition. Use of Scanning Electron Microscopy.

B.4. Intercomparison exercice (CoCo group)

C. PROGRESS OF WORK AND RESULTS OBTAINED

State of advancement

The isolation of humic materials of Fanay-Augères from a granitic water is finished : around 20 grams of humic and fulvic acids have been obtained. Their characterization by physical and chemical methods has been nearly achieved.

Complexation studies between humic materials and americium (III) by chromatographic and spectrophotometric techniques have been performed and formation constants values are presented.

The interlaboratory exercice on the isolation and characterization of natural colloids from Grimsel water /4/ has been performed in the frame of the CoCo exercice, with good results : ultrafiltration on membranes of different porosities coupled with Scanning Electron Microscopy has been used to determine the colloid population, their size and composition.

The general work progress status is as follows : B.1.1. is finished, B.1.2.is nearly achieved ; B.2., B.3. and B.4. are progressing normally.

Progress and Results

The concentration of humic substances from a natural granitic water (Fanay-Augères/ Massif-Central ; a borehole at 260 meters of depth) on an anionic exchange resin DEAE-cellulose /5/ conducted to the isolation of 8 grammes of humic acids and 10 grammes of fulvic acids. A second step in the purification of Fanay-Augères humic acids has been reconsidered due to their high ash content and has been performed with success : 5 grammes of humic acids have been obtained after this procedure. The quantities obtained (representative of a very long work in mine and laboratory due to the rather low organic carbon content in Fanay-Augères water) are suffisant for the characterization studies (in particular for the intercomparison exercice CoCo) and interactions studies with actinides. These natural humic materials are considered as representative of a granitic site and hence, have been distributed to the participants of the CoCo group for the intercomparison work.

The humic substances in Fanay-Augères water represent around 42% of the total organic carbon (sorbed on the resin) with 19% of humic acids and 23% of fulvic acids. These results are quite in good agreement with those obtained in the literature /6/. The characterization of the Fanay-Augères water in order to have a better understanding of the "origin" of the organic materials is still in progress and development : water analysis, application of geochemical codes, datation, ...

The characterization of Fanay-Augères humic compounds by different physico-chemical techniques (elementary, mineral, spectroscopic (UV-Visible, IR), chromatographic and titrimetric analysis) showed that these natural substances present the same characteristics of other aquatic humic materials (elementary composition), a non negligeable mineral charge, an aliphatic stucture more pronounced for humic than for fulvic acids (infrared spectroscopy) and a low humification state for fulvic acids.

Focus on the size is undertaken : development and comparison of the results obtained by chromatography, ultrafiltration and photon correlation spectroscopy /7/. This characterization study is still in development for the "purified" humic sample, particularly for the size and charge properties. Some properties for the fulvic acids from Fanay-Augères are presented in Table I.

The results obtained in the frame of the CoCo exercice on the intercomparison of humic substances (from different sites) characteristics will be presented together in a next report.

The study of the interactions between humic materials and Am(III) by a chromatographic technique /5, 8/ is under progress : a typical chromatogramme obtained with a Sephadex gel G15 is presented on Figure 1 where the two peaks, a positive (complexed Am) and a negative (deficient Am) one, appear (proof of the complexation). In order to understand the non-resolution of the two peaks (non-equilibrium or molecular size of the entities), the same experience has been performed on a Sephadex gel G25 (Figure 2) : the peaks are separated. The values of the formation constants determined by this technique are given in Table II. These values can be compared with the formation constants obtained by spectrophotometric method /9/ : they are quite in good agreement (Table II). The application of this chromatographic technique to different humic materials is under investigation, in the same pH range, and the study in alcaline pH range (in carbonate medium) is under development.

The spectrophotometric study of the system Fanay-Augères humic substances-Am(III) has been performed and the interpretation is nearly achieved. The comparison with other ligands will be made.

The characterization of Grimsel colloids in the frame of the intercomparison exercice CoCo on colloids has been carried out by combining ultrafiltration (in situ and in laboratory) of Grimsel water, and Scanning Electron Microscopy (SEM). The ultrafiltration has been done on Amicon and Millipore membranes of different porosities (450, 220, 100, 15 and 3 nm). The results obtained with the CEA membranes are presented in Table III. The observations of PSI membranes and AECL concentrate (production of membranes in the laboratory) have also been made. The water analysis (cation/anion content, U and organic carbon content) has also been investigated. A common report with all the results will be published.

List of Publications

MOULIN V., ROBOUCH P., VITORGE P. and ALLARD B.
"Environmental Behaviour of Americium(III) in Natural Waters"
Radiochimica Acta, 44/45, 33-37 (1988)

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/1/ CHOPPIN G.R. and ALLARD B., Handbook on the Physics and Chemistry of Actinides, Eds A.J. Freeman and C. Keller, North Holland Publ. Corp., Amsterdam, 3, Chapter 11 (1985)

/2/ McCarthy J.F., Wobber F., DOE/ER-0331 (1986)

/3/ KIM J.I., Handbook on the Physics and Chemistry of Actinides, Eds A.J.

Freeman and C. Keller, North Holland Publ. Corp., Amsterdam, 4, Chapter 8 (1986)
/4/ DEGUELDRE C., LONGWORTH G., MOULIN V., VILKS P., ALEXANDER R., BRUETSCH R., DEARLOVE J., WERNLI B., PSI Report, TM-453-88-04 (1988)
/5/ MOULIN V., BILLON A., THEYSSIER M. and DELLIS T., CCE FI1W/0068 Report, September-December 1987
/6/ PAXEUS N., ALLARD B., OLOFSSON U., BENGTSSON M., Mat. Res. Soc. Symp. Proc., IX, 525-532 (1985)
/7/ RAMSAY J. and BILLON A., APR 1988, Contract FI1W/0204
/8/ MOULIN V., Rapport CEA-R-5354 (1986)
/9/ MOULIN V., ROBOUCH P., VITORGE P., ALLARD B., Inorg. Chim. Acta, 140, 303 (1987)

	Fulvic Acid
Elementary Composition	
%C	47.5
%H	4.6
%O	43.1
%N	1.3
%Ash	3.5
Mineral Content (in ppm)	
Al	20
Si	1200
Fe	40
Mg	110
Ca	2200
K	1700
E4/E6*	17.5

Table I : Some characteristics of the fulvic acids obtained from Fanay-Augères water
* absorbance ratio at 460 and 660 nm at 50 mg/L, pH8 , Tris 0.1M

	Formation Constant (lg K ; K in L/g)
Chromatographic Technique	
G15 500µl [HA] 0.5g/L	4.75 ∓ 0.15 s=0.02
G15 200µl [HA] 0.5g/L	4.85 ∓ 0.15 s=0.03
G25 200µl [HA] 0.25g/L	5.0 ∓ 0.2
Spectrophotometric Technique	4.65

<u>Table II</u> : Formation constants values obtained for Aldrich humic acids at I = 0.1 M $NaClO_4$ in the following conditions :
o chromatographic technique on G15 :
pH 5, [Citrate Buffer] = 2.5 10^{-3} M, [Am] = 5 10^{-7} M
o chromatographic technique on G25 :
pH 5, [Citrate Buffer] = 10^{-3} M, [Am] = 10^{-6} M

o spectrophotometric technique :
pH 4.65, [Acetate Buffer] = 2 10^{-3} M, [Am] = 3 10^{-5} M, [HA] from 0 to 50 mg/L

s = standart deviation

Membrane Cut-off (in nm)	Particle Population (in particle/L)
⩾ 3*	4.5 10^9
⩾ 15*	3.8 10^9
	4.4 10^9
⩾ 100	6.0 10^8
⩾ 220	1.8 10^8
⩾ 450	2.6 10^7

* : in reality ⩾ 80 nm due to the detection limit with SEM

<u>Table III</u> : Grimsel colloid population obtained by ultrafiltration and SEM

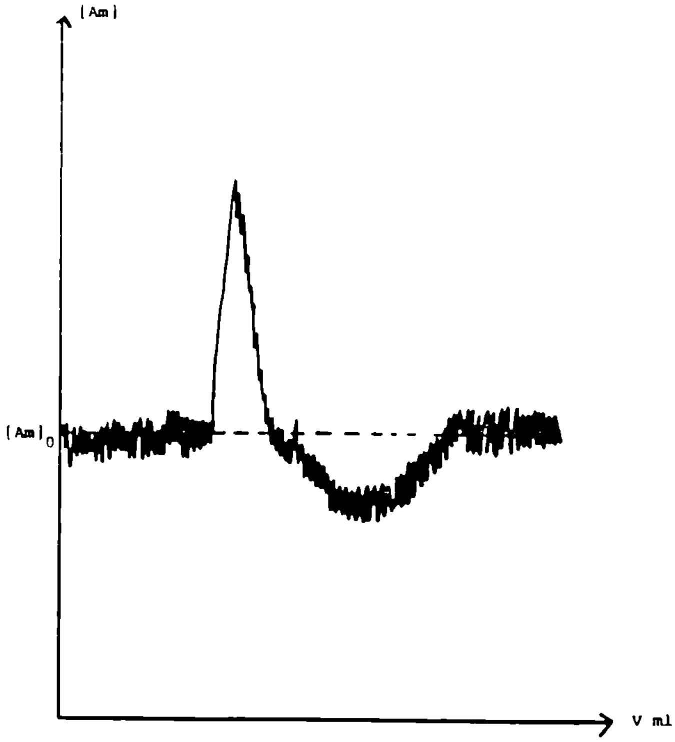


Figure 1 : Chromatogramme obtained on G15 with the system Am(III)-Aldrich humic acids (same conditions as in Table 2)

Figure 2 : Chromatogramme obtained on G25 with the system Am(III)-Aldrich humic acids (same conditions as in Table 2)

DIFFUSION, SORPTION AND STABILITY OF RADIONUCLIDE - ORGANIC COMPLEXES IN CLAYS AND CLAY-ORGANIC COMPLEXES.

Contractor : ICST, London, United Kingdom
Contract No. : FI1W/0147
Duration of contract : January 1988 - June 1990
Period covered : January 1988 - December 1988
Project leaders : L.V.C. Rees

A. OBJECTIVES AND SCOPE

Diffusion is likely to be the principle mechanism of the transport of radionuclides in clay. In order to select clay formations suitable for the disposal of radioactive waste, prior to site specific investigations, it is necessary to understand the factors upon which the diffusion of radionuclides in clay may depend.

Several factors may influence the mobility of ions in clay systems. We have chosen to concentrate on the following : clay mineralogy ; ionic strength of the electrolyte solution ; the nature of the exchangeable cations on the clay surface ; and the presence of organic ligands both in solution and adsorbed on or complexed with the clays.

As the clay system is modified both the degree of sorption of an element, and its mobility in both the solution and the adsorbed phases will vary. In order to elucidate the mechanism of diffusion it is necessary not only to measure the diffusion coefficient of the chosen ion, but also to assess the contribution of each species present, adsorbed or in solution, its oxidation state, degree of complexation, hydrolysation etc.

The element chosen for this study is neptunium, in the form NpO_2^+, because of the considerable hazard posed by this element, and the stability of the + 5 oxidation state in aqueous solution. The effect of simple organic ligands such as EDTA, citrate and saccharic acid and of humic acids will be studied.

B. WORK PROGRAMME

1.1. Preparation of Ca and Na forms of the three chosen clays ; St Austell kaolinite (two layers, nonswelling), Fithian illite (three-layer, nonswelling) and Wyoming montmorillonite (three-layer, swelling).
1.2. Measurement of d-001 spacing using X-ray diffraction.
1.3. Diffusion coefficient of a non-sorbed ion, $^{36}Cl^-$, which does not form organic complexes and the actinide, $^{237}Np(V)$, through columns of the different clays has been measured. Pore water composition covered a range of ionic strengths.
2.1. Preparation of clay organic complexes with simple ligands such as EDTA, citrate and saccharic acid, and with humic acids.
2.2. Measurement of d-001 spacing using X-ray diffraction.
2.3. Measurement of the rate at which the above radionuclides diffuse in the complexed clays.
2.4. Measurement of the self-diffusion rates of the simple organic ligands in the clays.
3.1. Preparation and characterisation of actinide-organic complexes starting with the EDTA, then citric and saccharic acids, and finally humic acid.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The Na and Ca forms of each of the clays have been prepared, with a background electrolyte of the perchlorate salt, molarity 0.01 M. The montmorillonite and kaolinites have also been prepared with a background electrolyte solution of molarities 0.001 M and 0.1 M. Cells suitable for the measurement of the tracer diffusion coefficient of ^{36}Cl ($^{36}Cl^-$ in perchlorate solution) using a pulse labelling technique, and of the diffusion coefficient of ^{237}Np using a half cell technique have been prepared and tested. Clay organic complexes of the calcium forms of montmorillonite and kaolinite have been prepared using the following ligands, EDTA, citrate and Aldrich humic acid at two levels of ligand concentration. The diffusion coefficients of ^{36}Cl and ^{237}Np have been measured in each clay preparation. The time dependant and concentration dependant sorption of ^{237}Np on each clay has been measured. The adsorption isotherms and self-diffusion coefficients of the ligands EDTA and citrate have been obtained for each clay.

The characterisation of the radionuclide organic complexes is about to commence.

PROGRESS AND RESULTS

1. The diffusion coefficient of ^{36}Cl is required to calculate the liquid phase impedance factor of an ion in solution, f_L. Unless further impeded by geometric or electrostatic interactions this factor should be the same for each species in solution. In order to minimize any interference from the complexes $CaCl^+$, $CaCl_2$ and NaCl both in solution and adsorbed on the clay surface, the isotope was purchased in perchlorate solution. It was not possible to obtain $^{36}ClO_4^-$.

Moist clays were packed into Perspex cells of internal diameter 0.95 cm and length 1.8 cm. They were allowed to come to equilibrium on a moisture suction plate at a tension of 150 kPa uniform packing density and moisture tension. Deposition of a trace amount of ^{36}Cl was achieved by placing a nucleopore filter containing a solution of ^{36}Cl, dried to the same moisture tension to avoid moisture flux, on the surface for a short period (< 1 % diffusion period). The clays were then sealed and placed in an incubator for a period of 4-6 hours. They were then removed and sections of 370 µm progressively extruded and removed. The slices were extracted into 0.01 M $CaCl_2$ solution and the activity in aliquots of the supernatant solution measured and from the concentration profile the diffusion coefficient calculated. The results are shown in Table II. The clay mineralogy and exchangeable cation influence the value of D(Cl), but the effect is not great.

The adsorption of NpO_2^+ by each of the clay minerals both in the Na and Ca forms at each electrolyte concentration has been measured. No change in concentration of the supernatant solution was found over a period of several weeks ; the sorption was thus not time dependent. This strongly suggest that fixation of Np by the clay minerals or penetration into slowly accessible interlayer sites in the clay mineral does not occur. The distribution coefficient, R_D was measured at a clay : solution ratio of 1 : 20. The sorption was measured at a range of concentrations of Np $3 \times 10^5 - 7 \times 10^6$ CPM dm^{-3}. The adsorption isotherms were found to be linear, R_D obtained from linear regressions on these isotherm are shown in Table II. These values were used to calculate the proportion of the element in solution as a time average, and hence along with f_L, the calculated contribution of the liquid phase to the measured diffusion coefficient. This term, denoted by D_{Liq} would be the value of D expected if diffusion occurred in the liquid phase only. A value of D_L, the diffusion coefficient of NpO_2^+ in an ideal solution was taken to be $6 \times 10^{-10} m^2 s^{-1}$.

The diffusion coefficient of Np was calculated from the fraction of the isotope having diffused from an initially labelled half-cell to a half cell which did not initially contain Np. The labelled clay was prepared by mixing dry clay with a solution containing ^{237}Np, the unlabelled clay was otherwise identical. Both half cells were equilibrated on a moisture suction plate as for Cl diffusion. When equilibrated the half cells were clamped together, and stored in an incubator for an appropriate diffusion period. The ratios D/D_{Liq} are plotted in Figure I. For the kaolinite samples D is greater than D_{Liq} which might be taken to indicate that surface diffusion of the adsorbed species occurs. However the reverse is observed for the montmorillonite samples. There are two explanations for this, firstly that the value of D_L is not accurate, which invalidates all calculated values D_{Liq} or that the liquid phase diffusion pathway of NpO_2^+ differs considerably from that of Cl^- due to its greater size so that f_L (Cl) may not be applied to NpO_2^+.

2. The clay-organic complexes were prepared by mixing air dry clay with a solution made 10^{-3} or 10^{-5}M in the organic compound, and 10^{-2}M $Ca(ClO_4)_2$.

The procedures for the measurement of D (^{36}Cl), D (^{237}Np) and R_D were exactly as described for the uncomplexed clays. The results are presented in the same way in the same Tables and Figure.

It should be noted that the formation of clay-organic complexes did not have a significant effect on the liquid phase impedance factor of Cl^-; as the moisture content and the packing density were unchanged this is not surprising. The adsorption isotherms of Np in the complexed clays were linear, and R_D not significantly modified. The presence of organic complexing ligands thus did not modify the adsorption of neptunyl on the three clays studied.

A definite change in the measured value of the diffusion coefficient of Np is observed, particularly in the clays complexed with humic acid. Given the uncertainty surrounding the value of D_{Liq} it is unfortunately not possible to attribute this to a change in the proportion of surface diffusion or to greater impedance in the liquid phase due to the different size and charge of the complexed forms of neptunium in solution. A more thorough interpretation of these results may be possible when the neptunyl-organic complexes have been characterised, in the presence and absence of clay.

Table I

Distribution ration of Np (v) for each clay preparation

		Montmorillonite	Kaolinite	Illite
Na	0.001 M	68.35 ± 5 %	18.37 ± 6 %	
Na	0.01 M	82.79 ± 12 %	5.02 ± 16 %	64.39 ± 24 %
Na	0.1 M	79.55 ± 6 %	4.05 ± 28 %	
Ca	0.001 M	143.38 ± 8 %	3.48 ± 94 %	
Ca	0.01 M	81.81 ± 13 %	1.74 ± 61 %	21.96 ± 15 %
Ca	0.1 M	18.84 ± 15 %	1.40 ± 64 %	
EDTA	10^{-5} M	65.12 ± 12 %	0.92 ± 96 %	15.33 ± 6 %
EDTA	10^{-3} M	59.25 ± 5 %	5.34 ± 28 %	12.72 ± 6 %
Citrate	10^{-5} M	55.58 ± 16 %	2.06 ± 166 %	13.96 ± 9 %
Citrate	10^{-3} M	77.37 ± 14 %	32.38 ± 14 %	34.13 ± 12 %
Humic Acid	10^{-5} M	68.15 ± 9 %	3.75 ± 59 %	20.98 ± 14 %
Humic Acid	10^{-3} M	92.04 ± 20 %	20.06 ± 26 %	36.31 ± 16 %
Average of Ca 0.01 M including organic ligands		65.43 ± 6 %	2.65 ± 28 %	16.54 ± 10 %

Table II

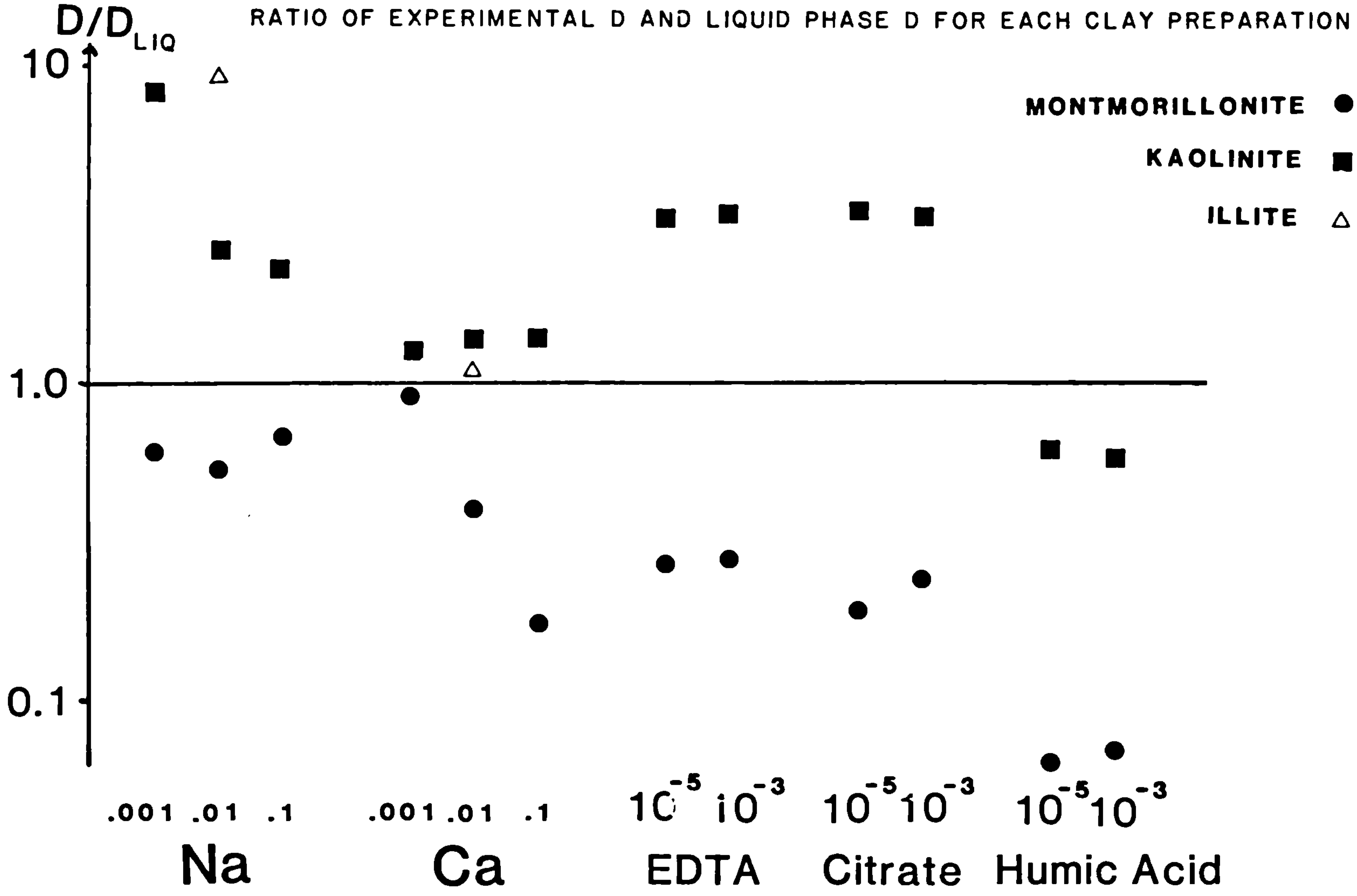
Diffusion coefficient of ^{237}Np and ^{26}Cl in each clay preparation

	Montmorillonite		Kaolinite	
	$D(^{237}Np)$ 10^{-13} m^2s^{-1}	$D(^{36}Cl)$ 10^{-10} m^2s^{-1}	$D(^{237}Np)$ 10^{-11} m^2s^{-1}	$D(^{36}Cl)$ 10^{-10} m^2s^{-1}
Na 0.001 M	14.89 ± 10%	7.14 ± 2%	2.94 ± 3%	4.66 ± 7%
Na 0.01 M	11.58 ± 24%	5.67 ± 2%	3.57 ± 37%	4.65 ± 6%
Na 0.1 M	15.75 ± 14%	6.68 ± 10%	2.95 ± 12%	4.49 ± 9%
Ca 0.001 M	4.80 ± 16%	4.15 ± 6%	3.00 ± 21%	5.34 ± 20%
Ca 0.01 M	8.14 ± 3%	5.80 ± 3%	3.08 ± 2%	4.94 ± 5%
Ca 0.1 M	11.60 ± 16%	5.61 ± 4%	3.27 ± 25%	5.11 ± 5%
EDTA 10^{-5} M	5.89 ± 6%	6.26 ± 16%	7.95 ± 8%	5.29 ± 6%
EDTA 10^{-3} M	6.39 ± 37%	6.75 ± 3%	8.24 ± 12%	5.31 ± 10%
Citrate 10^{-5} M	4.21 ± 29%	6.36 ± 9%	6.16 ± 2%	4.03 ± 9%
Citrate 10^{-3} M	5.65 ± 31%	6.69	5.96 ± 9%	4.09
Humic Ac. 10^{-5} M	1.05 ± 20 %	4.80 ± 16%	1.37 ± 30%	4.47 ± 4%
Humic Ac. 10^{-5} M	1.34 ± 31 %	5.48 ± 1%	1.28 ± 3%	4.47 ± 5%

Table II cont.

	$D(^{237}Np)$ 10^{-11} m^2s^{-1}	$D(^{36}Cl)$ 10^{-10} m^2s^{-1}
Na 0.01 M	3.34 ± 14%	3.36 ± 13%
Ca 0.01 M	0.447 ± 16%	3.72 ± 2%

FIGURE 1



D/D_{LIQ}
RATIO OF EXPERIMENTAL D AND LIQUID PHASE D FOR EACH CLAY PREPARATION
10
1.0
0.1
MONTMORILLONITE
KAOLINITE
ILLITE
.001 .01 .1
Na
.001 .01 .1
Ca
10^{-5} 10^{-3}
EDTA
10^{-5} 10^{-3}
Citrate
10^{-5} 10^{-3}
Humic Acid

EFFECTS OF NATURAL ORGANIC SUBSTANCES ON THE GEOCHEMISTRY OF A RADIOACTIVE WASTE REPOSITORY

Contractor : UKAEA, Harwell Laboratory, UK
Contract No : FI1W/0156
Duration of Contract : January 1988 - December 1989
Period Covered : January 1988 - December 1988
Project Leader : F T Ewart

A OBJECTIVES AND SCOPE

A significant proportion of the dissolved organic carbon in natural groundwaters consists of humic and fulvic acids. There is evidence that these may form complexes with some radionuclides which may result in increased aqueous concentration either due to increased solubility or decreased sorption on to surfaces. Previous work in this laboratory has involved the completion of a literature review /1/, an investigation of possible methods of characterisation and some preliminary studies of the effect of humic acid on the sorption of americium on cementitious materials. This programme is for a continuation of that research in those areas relevant to a radioactive waste repository.

There are four main objectives in the programme. Limited humic characterisation forming part of the "CoCo" intercomparison exercise will be made on groundwater samples obtained from geological structures which are typical of reference repository sites. The effects of humic acids on the solubility of Pu, Am and Np in waters representative of a cementitious repository will be investigated and the influence on sorption studied for these elements. A study will be made of the feasibility of including the effects of natural organic compounds in geochemical modelling codes.

B WORK PROGRAMME

B.1 Characterisation of humic substances
B.2 Effects of humic acids on Pu, Am and Np solubilities.
B.3 Effects of humic acids upon the sorption of Pu, Am and Np on cement grouts.
B.4 Modelling studies.

C PROGRESS OF WORK AND OBTAINED RESULTS

Statement of advancement

In the period covered by this report the sample of reference humic acid supplied as part of the CoCo club intercomparison exercise has been characterised by ultracentrifugation and gel permeation chromatography. The molecular weight was determined to be between 2,600 and 4,000. Calcium at concentrations greater than 10^{-3}M has been shown to cause flocculation of humic acid extracted from Boom clay. This concentration is comparable with those found in waters equilibrated with cement formulations. Humic acid has been shown to maintain high levels of americium at high pH in solutions defined by filtration through 0.45µm filters. Some enhancement of americium solubility may also be observed in sodium hydroxide solutions after filtration through 30000 molecular weight cut-off filters.

The current state of the programme is as follows:

B.1 Work for the intercomparison exercise completed, other work progressing normally.

B.2 Progressing normally.

B.3 Scoping experiments in progress, slightly delayed from original programme.

B.4 Not started, current experimental data insufficient.

PROGRESS AND RESULTS

Characterisation of humic substances (B1)

As part of the CoCo club intercomparison exercise we have determined the molecular weight of a sample of the reference purified humic acid (ex-Aldrich) by ultracentrifugation. All determinations were made on samples of humic acid dissolved in sodium hydroxide solution. Sedimentation velocity experiments yielded a weight-average molecular weight, M_w, of 4,000 whilst sedimentation equilibrium studies gave values of 2,600 for M_w and 2,800 for M_z the z-average molecular weight. Gel permeation studies under alkaline conditions show some polydispersity to be present and give an average molecular weight of approximately 3,000.

The flocculation of humic acid by calcium ions has been studied; these experiments give an indication of the behaviour of humic acid in the near-field of a cementitious repository. Humic acid was extracted from samples of Boom clay by contact with 0.1M sodium hydroxide solution under nitrogen and the resulting leachates filtered through a 0.45µm filter. The leachates contained up to 200 $\mu g.ml^{-1}$ Total Organic Carbon (TOC) and were 2.5×10^{-4}M in calcium. Portions of the leachates were equilibrated with added levels of calcium for seven days and then filtered through 0.45µm or 0.22µm filters prior to TOC analysis. The results are shown in Table I and show that flocculation occurs at calcium concentrations above 1×10^{-3}M. This value can be compared with a concentration of approximately 4×10^{-3}M found in water equilibrated with a 9:1 Blast Furnace Slag/Ordinary Portland Cement mix.

Effect of humic acid on americium solubility (B2)

Initial studies of the solubility of amercium were made in the presence of humic acid extracted from Boom clay with sodium hydroxide solution as for the flocculation studies above. The solubility was approached from oversaturation using inventories of americium-241 of 4×10^{-8}M or 4×10^{-6}M and the concentration of the radionuclide determined by gamma counting or alpha spectrometry. Solid/liquid separation was achieved by filtration through 30,000 molecular weight cut-off (MWCO) or

0.45μm filters and the pH was 12.7. Results are presented in Table II for a range of humic acid concentrations. Most of the americium is in solution as defined by 0.45μm filtration for both inventories when humic acid is present. Solubility is enhanced by a factor of 1,000 for 30,000MWCO filtrates at the higher americium inventory. No similar enhancement is seen with the lower inventory of radionuclide.

Solubility measurements in solutions of Boom clay humic acid in 9:1 BFS/OPC equilibrated water have also been made. The humic acid concentration is controlled by the calcium level and the maximum attainable is therefore lower than for sodium hydroxide extracts. Only one americium inventory of 4×10^{-8}M has been used. Table III shows that no trend is discernible in the 30,000MWCO filtrates but that the inventory of americium is in solution for the 0.45μm filtrate at a humic acid level of 13 $\mu g.ml^{-1}$ TOC.

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TABLE I: EFFECT OF CALCIUM ON HUMIC ACID CONCENTRATION

[Ca]/M	TOTAL ORGANIC CARBON/$\mu g.ml^{-1}$	
	0.45μm FILTRATE	0.22μm FILTRATE
9.4×10^{-2}	25	-
9.7×10^{-3}	20	-
8.3×10^{-3}	24	38
5.3×10^{-3}	31	40
4.3×10^{-3}	70	63
2.3×10^{-3}	98	97
1.2×10^{-3}	160	-
3.4×10^{-4}	180	-
2.5×10^{-4}	165	-

TABLE II: AMERICIUM CONCENTRATIONS IN SODIUM HYDROXIDE EXTRACTS OF BOOM CLAY HUMIC ACID

HUMIC ACID/ $\mu g.ml^{-1}$ TOC	[Am] IN 30,000MWCO FILTRATE/M	[Am] IN 0.45μm FILTRATE/M
(a) Inventory $^{241}Am = 4.1 \times 10^{-8}$M		
241	9.1×10^{-10}	3.8×10^{-8}
24	2.5×10^{-9}	3.8×10^{-8}
2.4	4.5×10^{-9}	3.7×10^{-8}
0	2.7×10^{-9}	3.4×10^{-8}
(b) Inventory $^{241}Am = 4.1 \times 10^{-6}$M		
240	1.0×10^{-8}	3.8×10^{-6}
24	3.8×10^{-9}	3.9×10^{-6}
2.4	1.4×10^{-9}	3.5×10^{-6}
0	9.2×10^{-12}	2.1×10^{-9}

TABLE III: AMERICIUM CONCENTRATIONS IN 9:1 BFS/OPC WATER EXTRACTS OF BOOM CLAY HUMIC ACID

HUMIC ACID/ $\mu g.ml^{-1}$ TOC	[Am] in 30,000MWCO FILTRATE/M	[Am] in 0.45μm FILTRATE/M
13	7.4×10^{-11}	3.1×10^{-8}
1.3	1.4×10^{-10}	4.0×10^{-9}
0.13	1.7×10^{-10}	8.3×10^{-12}
0	4.1×10^{-11}	1.7×10^{-10}

SIMULATION OF RADIONUCLIDE EXCHANGE BETWEEN AQUEOUS AND MINERAL-ORGANIC PHASES.

Contractor : Université de Nantes, Republique Française
Contract No : FI1W-0197-F
Duration of contract: January 1988 - December 1989
Period covered : January 1988 - December 1988
Project leaders : Prof. J. PIERI,

A. OBJECTIVES AND SCOPE

Humic and fulvic acids are important transfer vectors of radionuclides in the geosphere, particularly in the porous sedimentary systems. Such systems are obliged ways between the geological formation of the nuclear waste storage sites (salt or clay options) and the biospher. In the case of the granitic formation, weathering materials of fractures or mined cavities have a similar behaviour. Indeed these acids ,always present in the underground waters, have complexation properties which can induce high mobility species in the water-table.

These are the goals of the project:

- measurments of humic acids molecular weight by means of ultracentrifugation and liquid chromatography (intercalibration exercise).
- fractionation and isolation of humic and fulvic species.
- measurements of the competition between the different species in the sorption of 156 Eu, 99 Tc, 241 Am, 238 Pu by means of a specially-built dialysis display.

B. WORK PROGRAMME

1 - Construction of a polydialysis reactor, derived from an existing model. The chosen material is P.T.F.E.
2 - The tested samples originate in Boom (Mol, Belgium), Gorleben (F.R.G.), and Fanay-Augères (France).
3 - Estimation of the competition at equilibrium conditions is obtained by sorption cinetics.
4 - Taking part in the intercalibration exercise on the humic and fulvic acids.

C. <u>PROGRESS OF WORK AND OBTAINED RESULTS</u>

<u>STATE OF ADVANCEMENT</u>

The aim of the fourth item, in the main outline of the intercalibration exercise in which the Co-Co-Club associated Laboratories took a part, was to give data on the molecular weights of some polymers (humic and fulvic acids). The natural tested sample originated in the experimental deposit site of Boom (Belgium). A humic acid, commercialized by the Aldrich firm was also studied.

Two metods were used: ultracentrifugation on preformed sucrose gradients, and gel-filtration low-pressure chromatography.

<u>PROGRESS AND RESULTS</u>

1 - <u>ULTRACENTRIFUGATION ON PREFORMED SUCROSE GRADIENTS</u>

1.1 - METHODOLOGY

The determination of the Svedberg's sedimentation coefficients have been carried on by the method of the linear preformed gradients (sucrose 0-15%, BUCHLER Auto Densi-Flow II C) in a high gravitation field (110,000 g, 8 hours. BECKMAN L5-75B Ultracentrifuge, SW 41-Ti rotor). The gradients are then divided into 25 fractions (Fraction collector LKB Superrac 2211) and the optical density measured for the following wavelenghts: 254, 465 and 665 nm.

The Svedberg's coefficient is calculated from the classical formula:

$$S = \frac{1}{\omega^2 . x} \cdot \frac{dx}{dt}$$

in wich x represents the migration of the fraction which has the higher optical density. From GOSH and SHNITZER (1980) /1/, the structure of the humic and fulvic acids own a fibrillar, more or less fold up ("spherocolloids"). Therefore, the estimation of the molecular weight has been estimated by an interpolation using two globular proteins: the Catalase (MW = 230,000, S = 11.3) and the horse spleen Ferritin (MW = 440,000, S = 17.6). The solvent used to make the gradients was a sodium hydrogenocarbonate - sodium perchlorate buffer ($NaHCO_3$ 0.01 N 15%, $NaClO_4$ 0.01 N 85%, pH 9).

1.2 - RESULTS

1.2.1 - The Boom Humic Acid

The initial concentration of the studied sample is 0.35 g/l. After the run, a maximum optical density is found for the 5th fraction (figure 1 B). The sedimentation coefficient is $2.62.10^{-13}$ sec. and the extrapolated molecular weight is 54,000. The examination of the figure 1 C shows a soft decrease of the E4/E6 ratio (except for the pellet-fractions 22 to 25 ,containing insoluble substances) indicating a separation of the humic acids from the fulvic acids whose molecular weights are less (CHEN <u>et al.</u> (1977) <u>in</u> STEVENSON (1982) /2/).

1.2.2 - The Aldrich Humic Acid

This substance is sold by the Aldrich firm in a salt-form. In order to remove the excess of sodium salt, a dialysis of a 1 g/l solution has been made, in a sodium hydrogenocarbonate - sodium perchlorate buffer. The result is a loss of little molecules, indicated by the variation of the E4/E6 ratio (from 2.52 to 2.28). As it is shown in the figure 2 B, the final distribution of the studied acid is mainly located in the 4 - 7 fractions. The calculated Svedberg's coefficient and the estimated molecular weights values are contained in Table I.

Fraction number	S sec	MW g
4	1.32 x 10-13	27,000
5	2.62 x 10-13	54,000
6	3.94 x 10-13	81,000
7	5.23 x 10-13	107,000

Table I. Svedberg's coefficients and associated molecular weights of Aldrich Humic Acid

This sample appears hence very polydispers. The E4/E6 ration evolution proves a high separation of the fulvic species on the top of the centrifuge tube (figure 2 C).

1.3 - DISCUSSION

The linear preformed gradients technic is proving effective in the determination of the Svedberg's coefficient. The lack of usable molecular structure data obliges us to do an analogy with well known proteins (Catalase and Ferritin) in order to estimate the molecular weights. The results are yet in harmony with those obtained by ultrafiltration (KIM, 1988) /3/. On the other hand, if the hypothesis of a fibrillar, partially coiled structure is retained, the WALES and VAN HOLDE formula /4/ will be applied:

$$M_{S,K} = 9.2 \times 10^{24} \times \eta^{3/2} \times \bar{V}_2 \times \left(\frac{S_o \times K_s}{1-\bar{V}_2 \times \rho_1}\right)^{1/2} \qquad \text{with } K_s = 1.66.[\eta]$$

in wich:

η_1	= 1.7558 x 10	poises	: solvent viscosity
$[\eta]$	= 0.5 to 5	poises	: intinsec viscosity (VISSER, 1964) /5/
$\bar{V}_2$	= 0.75	ml/g	: partial specific volume
ρ_1	= 1		: solvent density
S_o	= 2.62 x 10	sec	: sedimentation coefficient

The molecular weights range calculated in this way stretches from 14,000 to 43,000. The E4/E6 ratio shows the condensation degree of the aromatic woof (CHEN <u>et al.</u>, 1977) /6/.

2 - GEL-FILTRATION LOW PRESSURE CHROMATOGRAPHY

2.1 - METHODOLOGY

The selected ges was Sephacryl S-300, in spite of increased ionic interactions. The packing and eluting solvant was a sodium hydrogenocarbonate - sodium perchlorate buffer ($NaHCO_3$ 0.01 N 15%, $NaClO_4$ 0.10 N 85%, pH 9) added with 0.05% sodium azid. The column was 60 cm high, with a 0.9 cm inner diameter, the bed height was 58 cm, the flow rate was 8 ml/h. The column calibration has been made with the substances caracterized in Table II.

Substance	MW	abs. nm
Dextran Blue	2,000,000 (V0)	660
Aldolase	158,000	280
Albumin	67,000	280
Chymotrypsinogen	25,000	280
A-Ribonuclease	13,700	280
K-Hexaferrocyanate	329.3 (Vi)	420

Table II - Physical characteristics of column standards

The MW determination is obtained by the classical formula:

$$KD = \frac{Ve - V0}{Vi - V0} \quad \text{where } Ve : \text{elution volume}, \; V0 : \text{void volume}, \; Vi : \text{inner volume}$$

and a linear correlation between log(MW) and KD.
The samples were centrifugated before deposit and the only supernatant was chromatographied.

2.2 - RESULTS

2.2.1 - The Boom Humic Acid

One adsorption peak is displayed at 20 ml elution (figure 3 A), corresponding to a 0.251 KD value and to a 123,000 molecular weight value. Between this peak and the Vi volume, the acid concentration decrease to a threshold, beyond wich small molecules with high ionic interactions are eluted. The evolution of the E4/E6 ratio indicates a humic enrichment in the content of the latest eluted molecules, and the ionic-interacting molecules (figure 3 B).

2.2.2 - The Aldrich Humic Acid

The sample has been first dialyses as shown in 1.2.2. The chromatogram present a single peak (figure 4 A) for a 31 ml elution volume (KD = 0.926, MW = 7,000) corresponding to a monodisperse solution. The E4/E6 ratio indicates very clearly an enrichment in humic species with increasing elution volumes (figure 4 B). Interactions with the gel bed are lesser than those described in 2.2.1.

2.3 - DISCUSSION

The general sight of the elution curves agrees with the one given by DUBACH et al. (1963) /7/. The elution curves have always a single maximum and an asymmetrical shape. Thje tailing is very important in the case of the natural Boom sample (with large interaction effects), in spite of a high pH value (9). A separation of the humic and fulvic species is always observed.

3 - CONCLUSION

The two methods give similar results only in the general behaviour of the molecules. The estimated molecular weights are widely different because of the fundamental analogy made on the molecular shapes (ultracentrifuged gradients) and the various molecular adsorption (gel-filtration chromatography). Nevertheless, both methods are usable for size division and purification.

Further experiments in gel-filtration are in progress to define a more adequat elution buffer.

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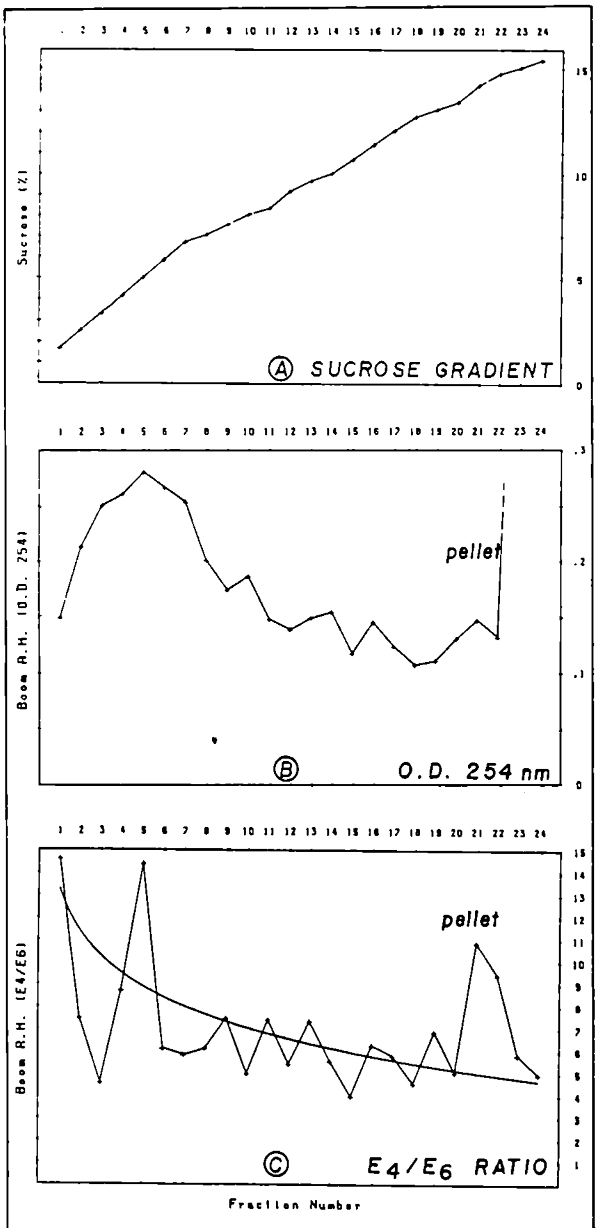


Figure 1 - Ultracentrifugation of the Boom H.A.

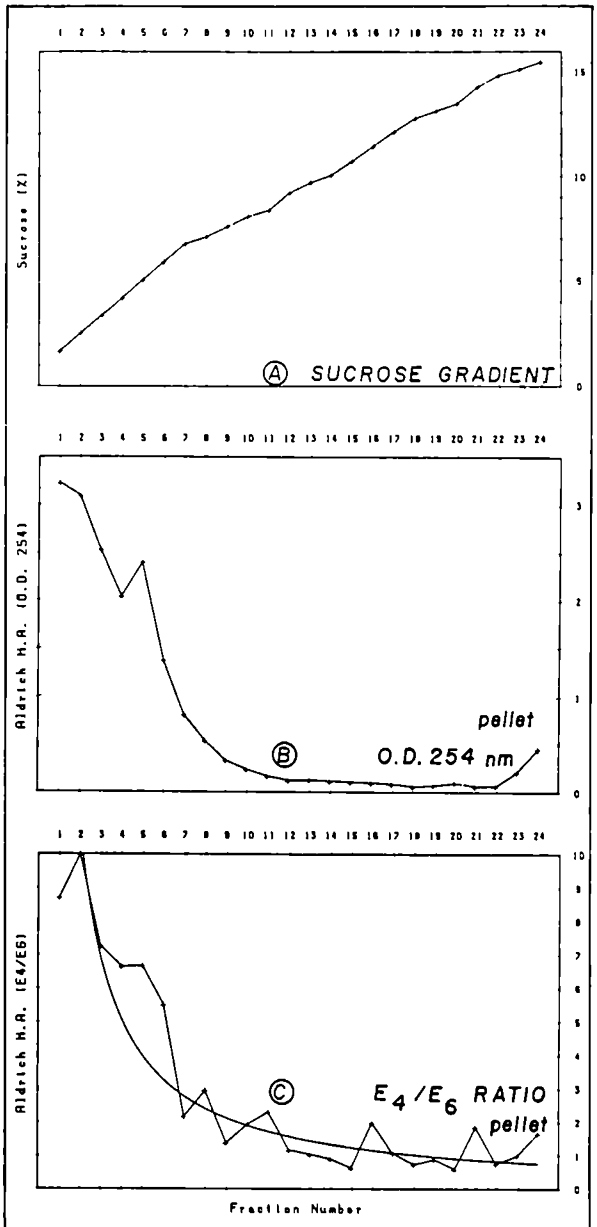


Figure 2 - Ultracentrifugation of the Aldrich H.A.

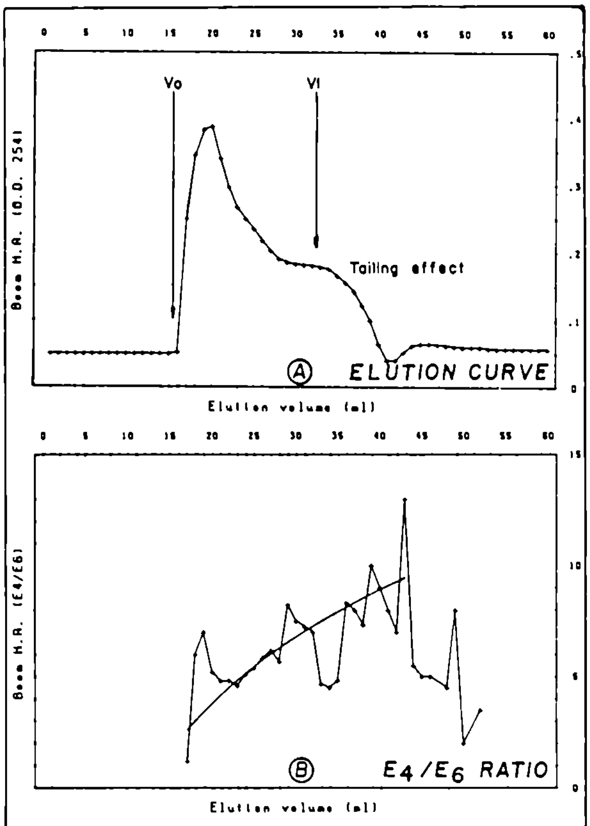


Figure 3 - Gel-filtration of the Boom H.A.

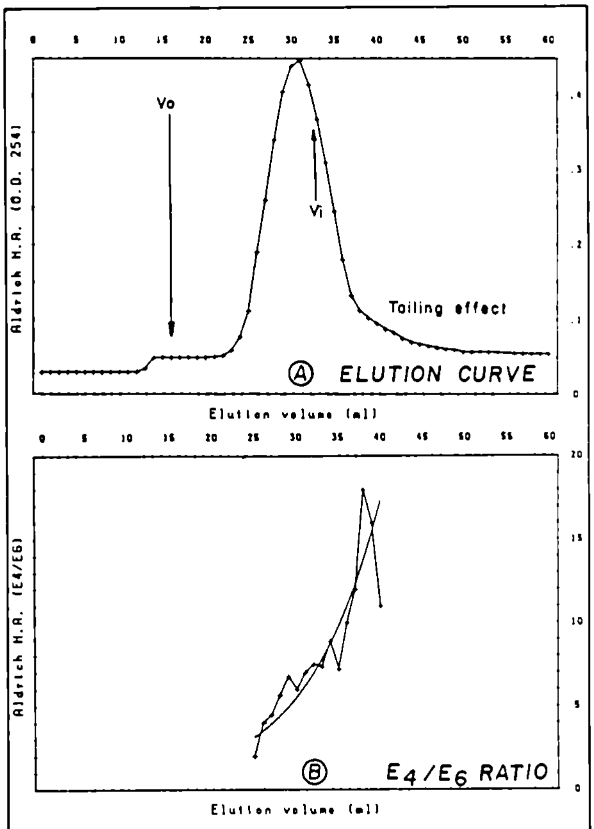


Figure 4 - Gel-filtration of the Aldrich H.A.

ASSESSMENT OF BASIC MIGRATION PHENOMENA OF ACTINIDES: COMPILATION, VERIFICATION, DETERMINATION OF THERMODYNAMIC CONSTANTS AND SPECIATION IN NATURAL AQUIFER SYSTEMS

Contractor: Institut für Radiochemie, Technische Universität München

Contract No.: FI 1W/0202

Duration of contract: Oct. 1988 - Dec. 1989

Period covered: 1. Jan. 1988 - 31. Dec. 1988

Project leader: J.I. Kim

A OBJECTIVES AND SCOPE

The first part of the research is to obtain reliable thermodynamic constants of hydrolysis reaction and carbonate complexation for actinides of different oxidation states, whose data are not well known or known with large uncertainties. This concerns Am, Pu, Np, Th and U ions. The species assumed for the evaluation of thermodynamic data will be verified with the help of spectroscopic or other methods.

The second part of the programme aims at the speciation of actinides in various aquifer systems. The conditions would be those of the Gorleben aquifers as a MIRAGE reference site. The elements to be studied are Np, Pu and Am. The speciation will be carried out using laser-induced photoacoustic spectroscopy (LPAS) with the help of ultrafiltration and ultracentrifugation. The indirect chemical speciation will be included for extremely low concentrations: $< 10^{-8}$ molar. A new spectroscopic method using laser-induced fluorescence (LFS) will be tested for actinides of tri- and hexavalent state with an expected speciation sensitivity up to 10^{-12} mol/L.

B WORK PROGRAMME

B 1. Compilation, verification, determination of thermodynamic constants of hydrolysis reaction and carbonate complexation

- Critical assessment and compilation of thermodynamic data of An(III), An(IV), An(V) and An(VI)
- Experimental determination and verification of uncertain thermodynamic data
- Verification of species assumed for the evaluation of thermodynamic data

B 2. Speciation of actinides in natural aquifer systems from the MIRAGE reference site (e.g. Gorleben)

- Speciation under various geochemical conditions of different aquifers
- Comparison of experimental speciation with theoretical speciation based on thermodynamic data
- Development of a new spectroscopic method

C PROGRESS OF WORK AND OBTAINED RESULTS

Statement of advancement

The literature survey of thermodynamic data for hydrolysis reactions and carbonate complexation of Th, U, Np, Pu and Am has been completed. The compilation and assessment of individual data are in progress. In the mean time, the carbonate complexation of Am(III) and hydrolysis reactions of Am(III), Am(V) and Np(V) in different ionic strengths have been reinvestigated. Particular emphasis is given to the characterization of solid compounds used for their solubility measurements. The solubility product of a well crystallized PuO_2 is redetermined. Verification of the literature data by own experiment or by theoretical correlations is in progress.

Speciation of actinides in Gorleben groundwaters has been carried out by Laser-induced photoacoustic spectroscopy (LPAS). In the mean time, Laser-induced fluorescence spectroscopy (LFS) is set up for the speciation study of An(III) and An(VI). LFS provides sensitivities for Cm(III) and U(VI) over 1000 times better than to be attainable by LPAS.

Progress and results

1. Hydrolysis reactions of Am(III)

Using a radiometric pH titration, hydrolysis reactions of Am(III) in $NaClO_4$ and NaCl have been investigated. At first the solubility product of $Am(OH)_3$ (amorphous) is determined spectroscopically at pH = 6.0 measuring absorbance at 502.9 nm. Hydrolysis constants are then determined in the pH range of 6 $\sim$ 13. The results are given in Table 1. The solubility product of $Am(OH)_3$ (am) becomes increased by 0.7 in logarithmic unit, when the specific α-activity in the solution is changed from 0.1 Ci/L to > 1.2 Ci/L.

Table 1: Solubility products of $Am(OH)_3(s)$ and hydrolysis constants of Am(III) in $NaClO_4$ and NaCl solutions

log Ksp	log β_1	log β_2	log β_3	Medium
-25.7 ± 0.3	6.3 ± 0.3	12.2 ± 0.4	14.4 ± 0.5	0.1 M $NaClO_4$, 0.1 Ci/L
-25.0 ± 0.3	6.3 ± 0.2	12.2 ± 0.3	14.4 ± 0.2	0.1 M $NaClO_4$, 1.2-5 Ci/L
-25.1 ± 0.5	6.0 ± 0.4	12.2 ± 0.5	14.8 ± 0.5	0.1 M NaClO, 2-5 Ci/L
-25.0 ± 0.1	5.6 ± 0.3	11.6 ± 0.4	14.1 ± 0.5	0.6 M NaClO, 2-5 Ci/L

2. Verification of hydrolysis constants by speciation

Based on the hydrolysis constants given in Table 1, the Am(III) species that might be present in the solution are speciated by calculation as shown in fig. 1. Following this calculation, at pH = 7 ∿ 8, three species, i.e. Am^{3+}, $Am(OH)^{2+}$ and $Am(OH)_2^+$, are to be present. Whether or not such a calculated speciation is relevant to the real solution has been verified by laser-induced photoacoustic spectroscopy (LPAS). The spectra are shown in fig. 2. The first spectrum from the upper part represents the Am^{3+} ion with its absorption at 5.03 nm and the second

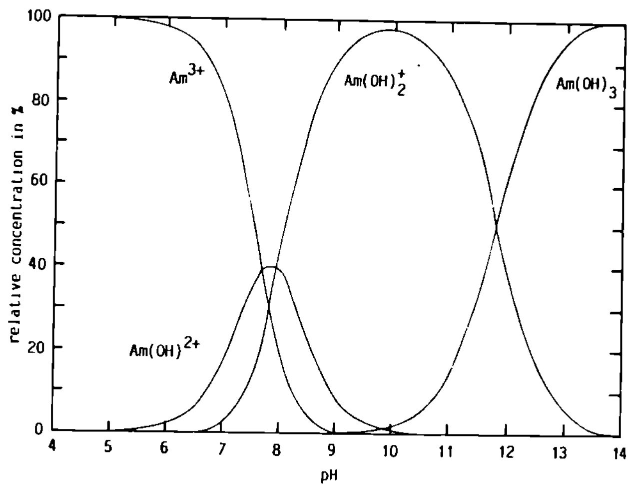


Fig. 1. Relative amounts of Am(III) species in 0.1 M $NaClO_4$ as a function of pH. Calculation is made by the solubility product and hydrolysis constants determined in 0.1 M $NaClO_4$ with the specific α-activity of 0.1 Ci/L (cf. Table 1).

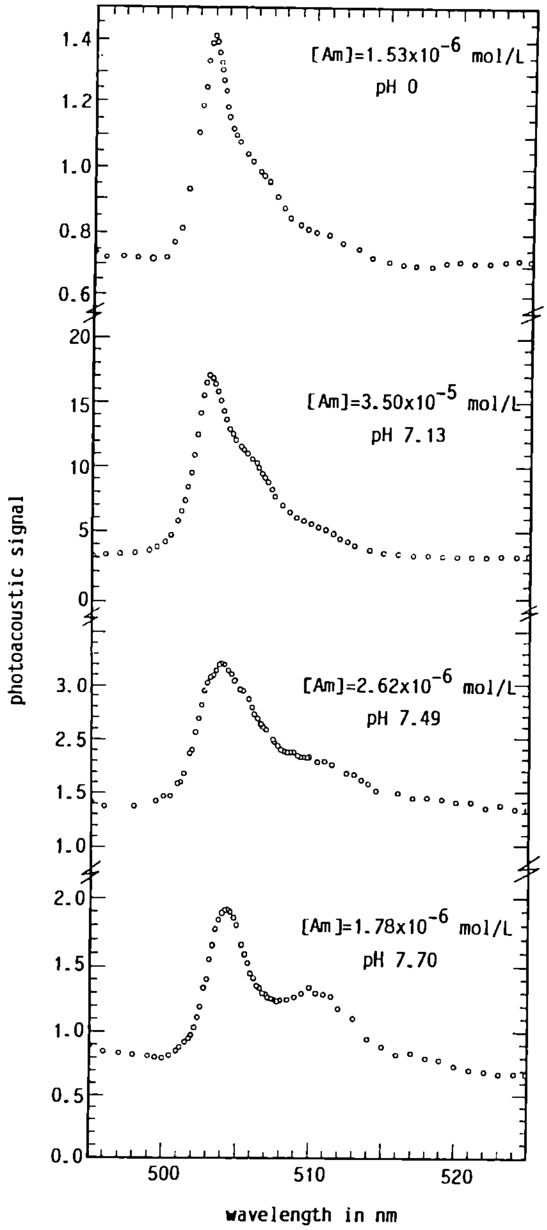


Fig. 2. Photoacoustic spectra (equivalent to absorption spectra) taken by the LPAS for the hydrolysis reactions of Am^{3+} in 0.1 M $NaClO_4$ with varying pH. Speciation spectra shown here verifies the theoretical speciation given in Fig. 1.

one, being a mixture of Am^{3+} and $Am(OH)^{2+}$, shows broadening of the absorption peak. The third spectrum reveals the appearance of a peak at 504.5 nm which is attributed to $Am(OH)^{2+}$ and at the same time the growth of a shoulder at 510 nm which is assigned to $Am(OH)_2^+$. The last spectrum shows distinctively two peaks at 504.5 nm and 510 nm indicating the presence of $Am(OH)^{2+}$ and $Am(OH)_2^+$ at pH = 7.7. The spectroscopic speciation has thus verified the accuracy of the thermodynamic data given in Table 1.

3 Speciation of Am(III) in groundwater

The Am^{3+} ion in 0.1 M $NaClO_4$ (pH = 6) is introduced in a Gorleben groundwater, which is kept under inert gas atmosphere (Ar + 1 % CO_2). The groundwater contains about 5 mg L^{-1} DOC (dissolved organic carbon), of which a substantial fraction is humic and fulvic acids. After 3 months of natural conditioning, the groundwater is filtered by a Nuclepore filter with pore size of 400 nm. The LPAS speciation of the filtrate is shown in fig. 3 (spectrum (a)). The absorption peak observed at 506.0 nm indicates either the carbonate or humate species, while the FWHM value of 4.8 nm suggests the Am carbonate. In consideration of the total carbonate concentration (approx. 3 x 10^{-4} M) and the humic/fulvic acid concentration (approx. 5 x 10^{-5} equiv. mol L^{-1}) and, taking account of their complexation constants of Am(III), both carbonate and humate (also fulvate) species of Am(III) can be present in this groundwater.

In order to verify the actual chemical state, the solution is transfered from the Ar + 1 % CO_2 atmosphere to Ar atmosphere and, at the same time, the pH is lowered to 4.0 in order to decrease the carbonate concentration in the solution. By this process, the Am carbonate species are decomposed to produce the Am^{3+} ion. Upon releasing the partial pressure of CO_2, the colloid generation takes place in the solution, which is then filtered at 400 nm pore size. The Am concentration is decreased about 16 % to 4.3 x 10^{-7}mol L^{-1}. The photoacoustic spectrum of the filtrate is shown in the spectrum (b), which illustrates the peak shift to 503.2 nm, suggesting the production of the Am^{3+} ion from decomposition of its carbonates. The high shoulder at 506.0 nm indicates the coexistence of another chemical state of Am(III). Since the carbonate concentration in the solution at pH = 4.0 becomes negligibly small ($<10^{-7}$ M), the shoulder peak is suspected to indicate the presence of Am humate species. To confirm the presence of Am humates, the solution is further filtered with the smaller pore size of 1.3 nm. The Am concentration in the filtrate is decreased about 47 % to 2.3 x 10^{-7} mol L^{-1} and the spectrum (c) shows the unambiguous identification of the Am^{3+} ion with the better pronounced peak at 503.2 nm. The relative height of the shoulder peak is decreased by filtration at 1.3 nm pore size but the peak does not disappear. This remaining shoulder peak may be ascribed to Am fulvates which are much smaller in size and remain more stable over a wider range of pH than Am-humates.

From the systematic follow-up of the spectral work with the aid of ultrafiltration, as well as the change in the partial pressure of CO_2 and pH, the original chemical state of Am(III) in the given groundwater is confirmed as being the $Am(CO_3)^+$ ion (or $Am(OH)CO_3$) mixed to some extent with both Am humates and fulvates.

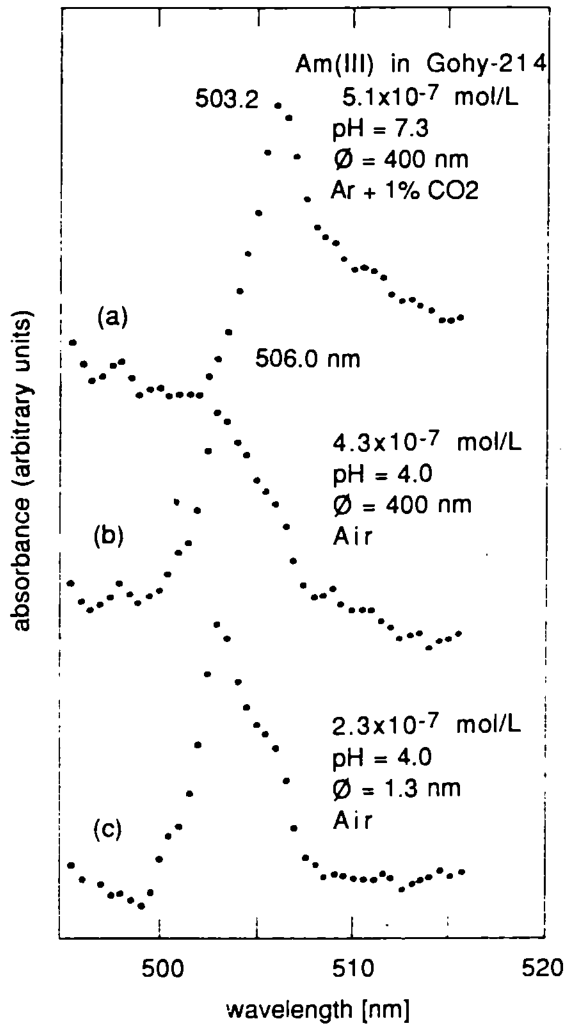


Fig. 3. Speciation of Am(III) in a Gorleben groundwater (Gohy-214); (a): under Ar + 1 % CO_2 atmosphere after filtration at 400 nm pore size; (b) under Ar atmosphere after filtration at 400 nm; (c) under normal atmosphere after filtration at 1.3 nm

List of Publications

[1] S. Stadler, J.I. Kim: "Hydrolysis Reactions" of Am(III) and Am(V)", Radiochimica Acta, 44/45, 39-44 (1988)

[2] R. Klenze, J.I. Kim: "A Direct Speciation of Transuranium Elements in Natural Aquatic Systems by Laser-Induced Photoacoustic Spectroscopy", Radiochimica Acta, 44/45, 77-85, (1988)

[3] R. Klenze, J.I. Kim: "Speciation of Transuranic Ions in Groundwater by Laser-Induced Photoacoustic Spectroscopy", Mat. Res. Soc. Proc. (1989) in print

COMPLEXATION OF RADIONUCLIDES WITH NATURALLY OCCURRING ORGANIC COMPOUNDS IN GROUNDWATER

Contractor : British Geological Survey, Keyworth, Nottingham
Contract No. : FI-1W-0203 (UK)
Duration of contract : from October 1988 to March 1991
Period covered : from October 1988 to December 1988
Project leader : G.M. Williams

A. OBJECTIVES AND SCOPE

The broad objective of the programme is to study the ability of natural organic compounds present in groundwater to form mobile complexes with radionuclides. The work involves development of techniques to extract natural organic material from groundwater with minimal alteration, the separation of those organic fractions that complex with cations, and the development of geochemical speciation models to describe or predict metal binding. The modelling approach will be tested by applying it to the study of the mechanisms which enhance the migration of nickel from a disposal lagoon into which a wide range of organics and heavy metals have been discharged. Liaison with Loughborough University of Technology has been established to develop appropriate techniques to determine radionuclide speciation directly and to separate organic fractions.

B. WORK PROGRAMME

The project is divided into the following research areas :

1. Assessment and development of techniques to extract and separate natural organic material from groundwater.

2. The separation, characterisation and quantification of those organic fractions which complex with radionuclides.

3. Development of a model to describe metal binding capacity based on the characterisation of natural organic compounds.

4. Investigation of the mobility of nickel which has apparently migrated over 100 m in groundwater organically contaminated with industrial wastes, in an attempt to apply and validate the modelling approach.

C. PROGRESS OF WORK AND RESULTS OBTAINED

STATE OF ADVANCE

The work has only recently commenced. A review is in hand of methods to extract organic material from groundwater. Attention is also being given to the separation of nickel complexes from groundwater sampled from the Villa Farm site, using gel chromatography. Equipment is being assembled to carry out on site ultra-filtration of groundwater and on site characterisation using anionic and cationic exchange resins, in conjunction with DEAE-cellulose for removal of "humic" material. A review /1/ has been completed of existing approaches for modelling interactions between natural organic compounds and cations which has refined the objectives for future model development.

PROGRESS AND RESULTS

Detailed results are not available at present.

D. REPORTS

/1/ Falck W E 1988. Modelling the interaction between natural organic matter and metal cations : a review.
British Geological Survey Fluid Processes Research Group
Report No. WE/88/49, 62 pp. (DRAFT).

Role of Colloids in the Transport of Radionuclides in Geological Formations

Contractors: UKAEA, Harwell, UK/CEA, Fontenay aux Roses, France.

Contract No: FIIW/0204
Duration of Contract: from July 1988 to December 1989
Working Period: July 1988 - December 1988

Project Leaders: J D F Ramsay, A Billon

A. OBJECTIVES AND SCOPE

Recent research has emphasised the potential importance of colloids in the migration of radionuclides from a waste repository into the geosphere /1,2,3/. Thus, if radionuclides are associated with colloids their fate may be different from that of simple ionic species in solution. In particular, depending on the size and charge of colloid species retention effects may occur due to filtration, or in contrast there may be enhanced migration due to differences in flow mechanisms and a reduction in the interaction with mineral surfaces.

Our objective here is to improve the understanding of the role of colloids in radioactive waste disposal. This is being achieved firstly by detailed characterisation of natural colloids derived from groundwaters, typical of those in a granite geology proposed for waste disposal, and secondly by investigating the interaction of natural and synthetic colloids (as model systems) with granite surfaces which have been characterised petrographically. This should provide an insight into retention mechanisms and would lead to subsequent investigations of the transport and hydrodynamic behaviour of colloids in these and other geological systems. Here expertise and techniques developed at both Harwell and Fontenay are being applied in a collaborative investigation.

B. WORK PROGRAMME

B.1 Preparation and characterisation of synthetic colloids.
B.1.1 Preparation of silica, iron oxide, alumina and humic materials.
B.1.2 Characterisation by light scattering, electrophoresis and electron microscopy.
B.2 Characterisation of natural colloids occuring in groundwaters.
B.2.1 Characterisation of host rock.
B.2.2 Characterisation of groundwater.
B.2.3 Characterisation of natural colloids by light scattering techniques, ultrafiltration and electron microscopy.
B.2.4 Isolation of organic colloids.
B.3 Studies of radio-colloid retention processes:
B.3.1 Association (incorporation, sorption) of actinides and rare earth elements with natural/synthetic colloids.
B.3.2 Interactions of colloids with well characterised mineral surfaces.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The characteristics (size, concentration) of natural colloids in groundwaters have been established by light scattering techniques (static light scattering and photon correlation spectroscopy). Investigations have focussed on samples from Grimsel and Fanay Augères within the Co Co interlaboratory exercise. These techniques have highlighted unforeseen artefacts which may arise during the collection, concentration and storage of natural waters containing colloids. State of the art equipment for photon correlation spectroscopy and electrophoretic light scattering has been installed at Fontenay. This has been used together with apparatus at Harwell in a detailed characterisation of a range of humic acids.

Model colloids (silica, ceria, iron oxide) have been prepared and characterised. The interaction of these colloids (ceria dispersions) and pseudocolloids (ceria-coated silica) on mineral surfaces (mica, untreated and prehydrated silica) has been studied. Preliminary investigations using an ion beam technique (RBS) to establish the different mechanisms involved in colloid retention are very promising.

Progress and results

1. Preparation and characterisation of synthetic colloids

Model colloids have been prepared for detailed investigations of sorption and retention mechanisms (see 3). These include silica, iron oxide (haematite) and ceria. These systems contain relatively monodispersed particles with diameters of $\sim$80 and 6nm respectively. Colloidal silica particles containing actinide and rare earth tracers (U(VI) and Ce(IV)) have also been prepared as 'pseudocolloid' analogues. These systems have been characterised by light scattering and electrophoresis.

2. Chracterisation of natural colloids occurring in groundwaters

2.1 Groundwaters

Light scattering techniques have been used to characterise natural colloids derived directly from granite groundwaters from the Grimsel site in Switzerland and from Fanay-Augères, Massif-Central. More detailed investigations have been conducted on humic acid colloids extracted from the water of the latter site (B.1.1).

Based on static light scattering intensities (which are $\sim 10^3$cps above background) it is tentatively suggested that particle concentrations are $\lesssim 10^{10}$ particles dm^{-3}. This value is broadly comparable to that which has been reported for Grimsel water by others using SEM and LPAS /4,5/. The Grimsel and Fanay groundwaters were filtered through 1.0µm and 0.1µm nucleopore polycarbonate membrane filters. The 1.0µm filter reduced scattering from the Fanay water to background levels whereas for the Grimsel water a 0.1µm filter was necessary to produce the same effect. From photon correlation spectroscopy (PCS) measurements values of particle translational diffusion coefficients in the range of $\sim 10^{-9} cm^2 s^{-1}$ have been obtained, corresponding to hydrodynamic diameters of 2 to 3µm. Although the polydispersity of samples has not been quantified, these values are significantly larger than particle sizes derived from SEM (<1µm).

This difference may be attributed to the presence of weak or transient particle aggregates in groundwaters, and hence indicates the importance of in situ measurements. Other artefacts which arise in groundwaters on storage and concentration by ultrafiltration (where colloids are retained on membrane surfaces) are also evident from light scattering.

2.2 Humic Acids

Both static light scattering and PCS measurements on different humic acid solutions are in accord and show evidence of aggregation of smaller units in solution. This is evident from values of radius of gyration (Rg ∿100nm) on samples from Aldrich, Fanay Augères and Gorleben. About ten humic acid samples were studied by photon correlation spectroscopy. They all contained a variable but always significant amount of particles in the 50-200nm hydrodynamic equivalent diameter range (Table 1). Such particles are absent in blanks and in "synthetic" humic acid samples; they were isolated by ultrafiltration and confirmed by scanning and transmission electron microscopy. The influence of pH and ionic strength on the size of particles from a number of HA samples was investigated, and found to be minimal.

The effect of increasing Ca(II) concentration on particle size was remarkable. No variation was apparent up to a critical concentration ([Ca] $\sim 5 \times 10^{-3}$ moldm^{-3}), at which point coagulation occurred yielding larger stable particles, or aggregates, with a size of ∿2µm as derived from PCS. This finding is significant because the transport behaviour of metal humates will depend on the physical properties of these larger particles.

Preliminary investigations of the electrophoretic properties of humic acid scatterers indicate negative surface charge. Increasing Ca(II) concentration reduced the absolute charge with no apparent discontinuities at the precipitation discontinuity (Fig 1).

3. Studies of radio-colloid retention processes

The mechanisms of colloid retention on mineral surfaces have been investigated using Rutherford back scattering (RBS) techniques /6,7/ using model colloids of ceria and silica coated with ceria as prepared in C1. Thus on contact with both mica and silica surfaces only colloidal particulates of pure ceria are sorbed. The lack of any retention with the ceria coated silica particles can be ascribed to the similarity between the negative charge of both colloid and mineral surface which results in electrostatic repulsion. The same effect is evident for both untreated and prehydrated amorphous silica. However, it is evident from the RBS spectra for Ce that with untreated silica the CeO_2 colloids are only fixed on the very surface (adsorption) whereas the depth extension is larger on prehydrated silica indicating possible penetration into a gel layer at the interface. Deconvolution to obtain quantitative depth profiles is underway and investigations are being extended to iron oxide and silica colloids with presorbed uranium.

TABLE 1 Average particle diameter (z-weighted) and scattered intensity of humic acid samples

Sample Average Diameter (M_Z) (nm) / Scattered Intensity (kcounts sec^{-1})

Sample	non centrifuged	centrifuged 5'	centrifuged 30'
Aldrich purified	123 / 492	122 / 495	110 / 415
Aldrich (Na salt)	129 / 533	141 /(711)	123 / 556
Lake Bradford	94 / 127	94 / 121	83 / 102
Podzol	246 / 769	191 / 651	156 / 474
Sediment	216 / 469	164 / 367	138 / 295
Rendzine	340 / 172	330 / 116	125 / 58
Fanay	808 / 697	433 / 347	203 / 194
Gorleben	123 / 19	132 / 16	72 / 11

	non filtered	filtered 0.45 µm
# Mol Water	250 /(370)	143 /(147)

Mol water has a humic acid content of about 200 mg/L, ionic strength 0.02 M (mainly due to $NaHCO_3$), and pH 8.5. It was collected, filtered and transported under rigorously anoxic conditions.
All other samples were prepared by dissolution of solid humic acids to give: [HA] = 100 mg/L, [$NaClO_4$] = 0.01 M, pH = 7.0.
Laser power was 0.5 W, except Mol water (0.15 W. Reported value is normalized to 0.5 W). Observation angle is 90°.
Electrolyte scatter (6.3 Kcounts/sec) was subtracted.

List of publications

/1/ RAMSAY, J.D.F., The role of colloids in the release of radionuclides from nuclear waste, AERE-R 11823 (1985).

/2/ RAMSAY, J.D.F. The role of colloids in the migration of radionuclides from nuclear waste, Proc. of Conf. "Migration 87", Munich, 14-18 September 1987, Radiochimica Acta, in press.

/3/ RAMSAY, J.D.F., AVERY, R.G. and RUSSELL, P.J. Physical characteristics and sorption behaviour of colloids generated from cementitious systems. Radiochimica Acta 44/45, 119 (1988).

/4/ DEGUELDRE, C.A., et al. In laboratory, on site and in situ sampling characterisation of Grimsel colloids - Phase I. EIR report TM-42-87-20 (1987).

/5/ KIM, J.I., Characterisation of colloids in groundwater. RCM 01687 (1987).

/6/ DRAN, J.C., PETIT, J.C., DELLA MEA,G., MENAGER, M.T., and PACCAGNELLA, A., Sorption of actinide analogues on granite materials studied by MeV ion beam techniques. Radiochimica Acta, in press.

/7/ MENAGER, M.T., PARNEIX, J.C., PETIT, J.C., and DRAN, J.C., Migration of uranium, thorium and REE in a fossil intragranitic geothermal system: implications for the mobility of actinides in radwaste disposal, Radiochimica Acta, in press.

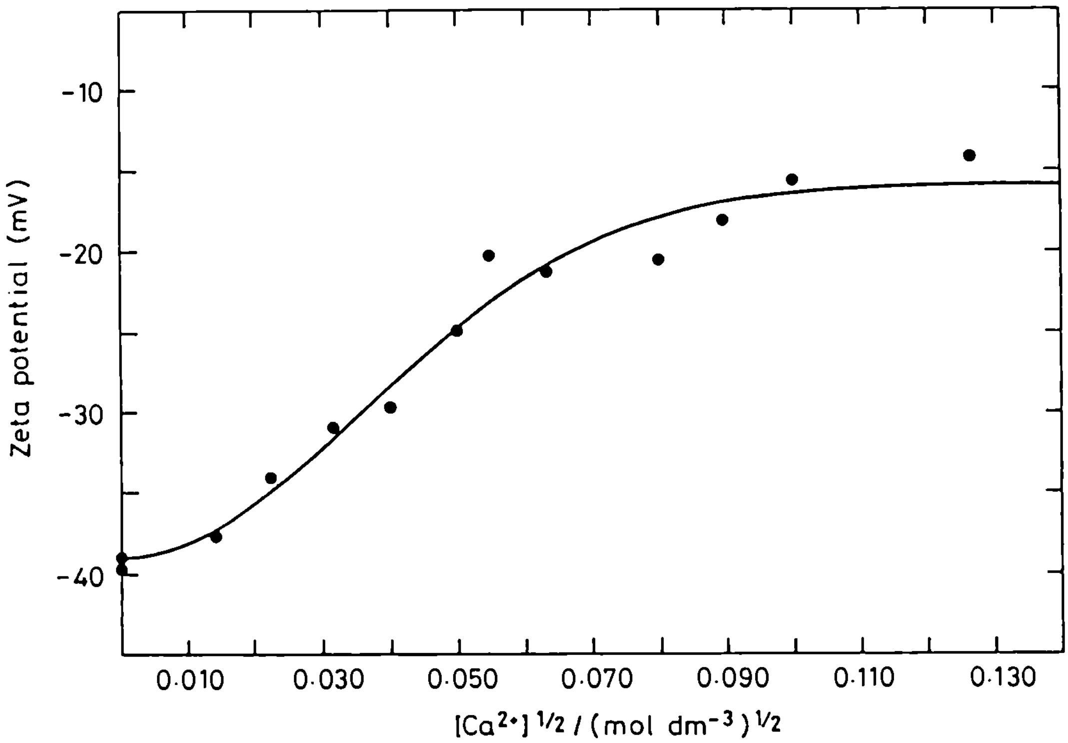


FIGURE 1. DEPENDENCE OF ZETA POTENTIAL OF PURIFIED ALDRICH HUMIC ACID SOLUTIONS (pH 7) ON Ca^{2+} CONCENTRATION.

ASSESSMENT OF EXPERIMENTAL RESEARCH TECHNIQUES FOR THE INVESTIGATION OF RADIONUCLIDE MIGRATION IN AQUIFERS

Contractor: GSF, Neuherberg, Federal Republic of Germany
Contract No.: FI1W/0210
Duration of contract: August 1988 - July 1990
Period covered: August 1988 - December 1988
Project leaders: P. Fritz, C. Wolfrum

A. OBJECTIVES AND SCOPE

Knowledge of the potential migration behaviour of radionuclides from repositories in deep geological formations is a prerequisite for the safety analyses of the entire disposal scheme. It requires detailed information about geochemical processes and reactions which may be important if a given radionuclide is released and migrates from the disposal site. Thus, much of the research which has to be undertaken concentrates on the geochemistry of selected elements. These investigations are usually performed as laboratory batch, column and diffusion tests. The experimental conditions can then be strictly controlled and varied with respect to a number of chemical and physico-chemical effects. The objectives of this work are to contribute to a better understanding of the transport behaviour of actinides as well as to present a critical review of the different laboratory techniques used in migration studies. The experimental work is focused on the radioisotopes of Tc, Eu, Pu, and investigates the essential influences on the transport behaviour and which are exerted by redox conditions, the formation of complexes with natural humic and fulvic acids as well as the formation and/or presence of colloids.
In the initial stage of the project samples from the Gorleben site, the planned German repository, are under investigation.

B. WORK PROGRAMME

1. The work was started with a literature study of sorption data obtained from various experimental techniques in comparison with own results and previous experience.

2.1 Sandy and clay sediments and corresponding groundwater samples from Gorleben with extensive mineralogical, chemical and physical characterisation were selected.

2.2 Sorption data were obtained by batch and diffusion procedures with special efforts to standardize experimental techniques (e.g. preconditioning, phase separation).

2.3 Kinetic and equilibrium experiments were started with Tc, Eu, Pu. In addition Sr (simple chemism) and Zr/Nb (complex/colloid forming properties) were investigated.

2.3.1 To assess the influence of different Eh conditions on the mobility of Tc, batch and diffusion tests are carried out using representative samples from Gorleben.

2.3.2 Effects of nuclide sorption on colloids and colloid formation for diffusion and sorption data are tested with tri- and tetravalent ions (Eu^{3+}, Zr^{4+}).

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Available practical experience from preceeding projects dealing with samples from Asse, Konrad and Gorleben sites permitted the immediate start of investigations using Sr, Zr, Nb. Preliminary tests with Eu and Tc were undertaken and experiments with Pu have been initiated at the beginning of 1989. Gorleben sediments and corresponding water samples (listed in Table I with mention of depth and lithology) are stored in a controlled atmosphere glove box (99% Ar with 1% CO_2) where as many as possible of the necessary manipulations will be carried out. In well-known standard batch and diffusion procedures sorption (Rs), desorption (Rd) respectively diffusion (D) coefficients are determined. These experiments are performed under ambient atmospheric conditions as well as under reduced oxygen levels in solution. Sorption/desorption behaviour is studied with respect to redox conditions, pH and contact time between solid and liquid phase at constant temperature, solution volume to solid mass ratio and initial radionuclide concentration. The central element of the project - the discussion about the dependance of sorption data on various experimental techniques (column, batch, diffusion) - should based on the program item 2 (laboratory studies) and therefore adjusted in the end of this report.

Progress and results

2.1, 2.2 SEDIMENTS, GROUNDWATERS AND EXPERIMENTAL PROCEDURES

For attainment of a realistic geochemical equilibrium system it is necessary to equilibrate solid and liquid phases before the real sorption experiments. About 1000 g of the sediment were put into 2000 cm^3 of corresponding groundwater, whereby the water samples were previously filtered using 0.45 μm cellulose acetate filters. The contact time was about three months and the samples were storaged in darkness to prevent photosynthetic activities. After this procedure the radionuclides of interest (for details see Table II) were added to these equilibrated water samples. Aerobic tests were prepared under laboratory atmosphere, anaerobic tests in glove boxes (O_2 content less than 0.1%).

2.3, 2.3.1 SORPTION EXPERIMENTS WITH REGARD TO KINETIC, EQUILIBRIUM AND THE EFFECT OF REDOX CONDITIONS

A large number of sorption data were produced in a preceeding project using material from the Asse for both oxic and anoxic atmosphere. Under oxidizing conditions Tc exists in solution as the anionic species TcO_4^-, in the 7+ valence state and shows weak sorption on geological materials (results: Rs = 0-2 cm^3/g, Rd = 0-5 cm^3/g). Although the reduced forms of Tc form a variety of slightly soluble or cationic species, which consequently exhibit much lower mobility, in our former investigations differences in the Rs/Rd values varied by at most a factor 10 if compared with data obtained in the presence of oxygen. Sorption/desorption behaviour of ^{99}Tc was studied in seven different sediment-groundwater-systems (sand, clay, marl) from the Gorleben site (Table I) with respect to redox conditions (Eh), pH and contact time at a constant temperature (21±2 °C), V/m=2.5 cm^3/g and initial ^{99}Tc concentration of 10^{-6} mol/l. After an experimental time of two months first results (Table III) show that within this time no remarkable sorption of Tc was observed (Rs = 0.1-1 cm^3/g, Rd = 0-1.6 cm^3/g). Parallel ^{99}Tc-batch-experiments with pyrites (FeS_2), we performed under identical conditions in contact with water of high salinity (GoHy 1092), yield a strong sorption (Rs = 840-1520 cm^3/g, pH = 4.5, electrical conductivity σ = 35000 $\mu S/cm$). For four Gohy systems diffusion experiments were performed under Ar-atmosphere. Diffusion behaviour was investigated from spiked groundwater through unspiked samples into unspiked groundwater. Diffusion coefficients were calculated from break-through curves (Ficks first law: D_s) and on the basis of Ficks second law (D_{ns}). In Table IV diffusion coefficients evaluated from both methods and hence following the sorption coefficients are compared.

2.3.2 SORPTION EXPERIMENTS WITH REGARD TO THE EFFECT OF COLLOIDS

Migration of colloid forming elements in natural systems is expected to be as fast as natural groundwater flow through aquifers not considering possible mechanical filtration effects. Thus, colloids are of special importance in a safety analyses. Here, experiments concentrated on Eu which constitutes as a model ion for the trivalent actinide ions. Experiments were undertaken to study colloid formation in groundwater under natural pH conditions by filtration (two examples in Figure 1) and in parallel, under the same experimental conditions, attempts were made to determine equilibrium values for sorption and desorption of ^{152}Eu (Rs = 57-2385 cm^3/g, Rd = 51-6210 cm^3/g). In a series of experiments the formation and behaviour of colloids are investigated by using radioactive Zr, Nb and Eu compounds which are added to natural groundwater samples in concentrations of 10^{-8} mol/l (^{95}Zr, ^{152}Eu) and 10^{-10} mol/l (^{95}Nb) (see Table II). In fixed time intervals small sample volumes were filtered through 0.45 and/or 0.22 μm. The decrease of the γ-activity in the filtered in contrast to the initial solutions indicates a time dependant formation of colloids. Calculations with the computer code "Mineql" will be undertaken to yield information about the behaviour of Zr, Nb and Eu in the natural GoHy water samples. However, substantial differences between calculated and experimental solubilities were noted in some cases and require further attention.

1. COMPARISON OF DIFFERENT EXPERIMENTAL METHODS

The central element of the project will be a discussion of the dependance of sorption data on various experimental techniques (column, batch, diffusion). As indicated above, chemical equilibration of sediments with corresponding waters requires substantial time and, therefore, not all experiments can be terminated within the present timeframe. Where this is the case it will, nevertheless, be possible to present preliminary results. In addition it must be noted that the complex geochemical behaviour of Pu makes it difficult to compare directly the results obtained through different experimental methods. In the case of Tc the behaviour was so far found to be that of ideal elements, because only weak sorption (no reduction) occurs and the diffusion coefficients are in the same range as for Sr. Eu may exist in two different species. As colloid with a usually negative charge it is expected to diffuse very fast because of negative charges on sediment surfaces, although it may show some filtration effects, and as cation the diffusion is influenced by sorption and ion exchange processes.

TABLE I: SAMPLE DESCRIPTION AND SUMMARY OF REALIZED EXPERIMENTS

GoHy sedi-ment	GoHy water	litho-logy	depth (m)	batch Tc	batch Eu	batch Pu	batch Sr	batch Zr	batch Nb	diffusion Tc	diffusion Eu	diffusion Pu	diffusion Sr	diffusion Zr	diffusion Nb
3	2131	clay, silt	-14/-14.3	x	x	x	x	x	x	x	x	-	x	x	x
5	1281	sand	-45	x	x	x	x	x	x	-	-	-	-	-	-
6	1281	clay	-49.3/-49.6	x	x	x	x	x	x	-	-	-	-	-	-
8(1)	1092	marl	-85.8	x	x	x	x	x	x	-	-	-	-	-	-
8(2)	1281	marl	-85.8	x	x	x	x	x	x	-	-	x	-	-	-
9(1)	1281	marl	-109.5	x	x	x	x	x	x	-	-	x	-	-	-
9(2)	1341	marl	-109.5	x	x	x	x	x	x	x	x	x	x	x	x
13	1092	sand	-187.5	x	x	x	x	x	x	x	x	-	x	x	x
17	641	sand	+3.9	x	x	x	x	x	x	x	x	x	x	x	x

marl: calcerous boulder clay

TABLE II: DETAILS OF RADIONUCLIDES USED IN BATCH AND DIFFUSION TESTS

radio-nuclide	half-life $t_{1/2}$	emis-sion	compound	specific activity	added conc. (mol/l)
^{85}Sr	60 d	γ	$SrCl_2$	100 GBq/g	10^{-8}
^{95}Zr	65 d	$\gamma(\beta)$	$Zr(Ox)_2$	90 GBq/g	10^{-8}
^{95}Nb	35 d	$\gamma(\beta)$	$Nb_2(OX)_5$	2 TBq/g	10^{-10}
^{99}Tc	$2.1 \cdot 10^5$a	β	NH_4TcO_4	630 MBq/g	10^{-6}
^{152}Eu	12.4 a	$\gamma(\beta)$	$EuCl_3$	140 GBq/g	10^{-8}
^{238}Pu	87.75 a	α	$PuO_2\ (NO_3)_2$	634 GBq/g	10^{-10}

TABLE III: SORPTION (Rs)- AND DESORPTION (Rd) COEFFICIENTS FOR ^{99}Tc IN GORLEBEN SAMPLES UNDER AMBIENT (Eh:+280 to +570 mV) and Ar-ATMOSPHERE (Eh:-180 to +245 mV)

GoHy samples	ambient atmosphere Rs/Rd (cm^3/g)	Ar-atmosphere Rs/Rd (cm^3/g)
4	< 0.1/0.4	0.2/0.5
5	0.1/0.2	0.1/1.2
6	< 0.1/ 0.1	0.5/1.6
8(1)	0.1/0.3	0.2/0.9
8(2)	0.1/0.4	0.7/2.7
9(1)	< 0.1/0.2	0.5/2.0
9(2)	< 0.1/0.3	0.05/0.3
13	< 0.1/0.3	0.5/2.4
17	< 0.1/0.4	0.1/0.5

TABLE IV: DIFFUSION (D_s/D_{ns}) AND SORPTION (Rs) COEFFICIENTS FOR ^{99}Tc DETERMINED UNDER Ar-ATMOSPHERE

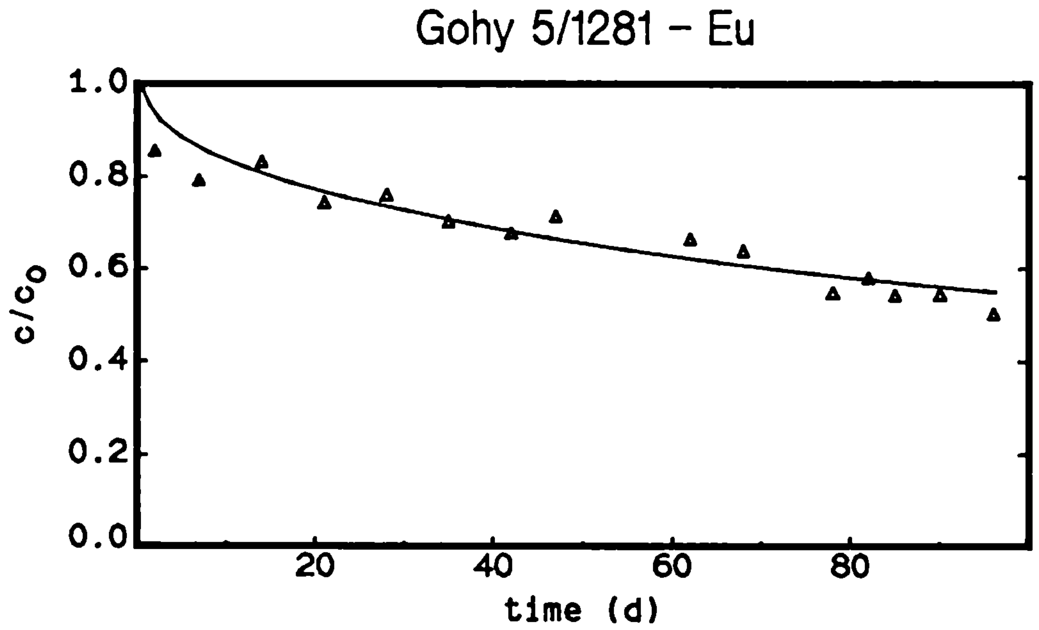
GoHy samples	D_s/D_{ns} (10^{-6} cm^2/s)	Rs (cm^3/g) (diffusion)	Rs (cm^3/g) (batch)
4	0.6/ -	0.3/ -	0.2
9(2)	1.0/1.0	0.02/0.02	0.05
13	1.2/1.5	0.6/0.8	0.5
17	1.1/1.6	0.3/0.5	0.1

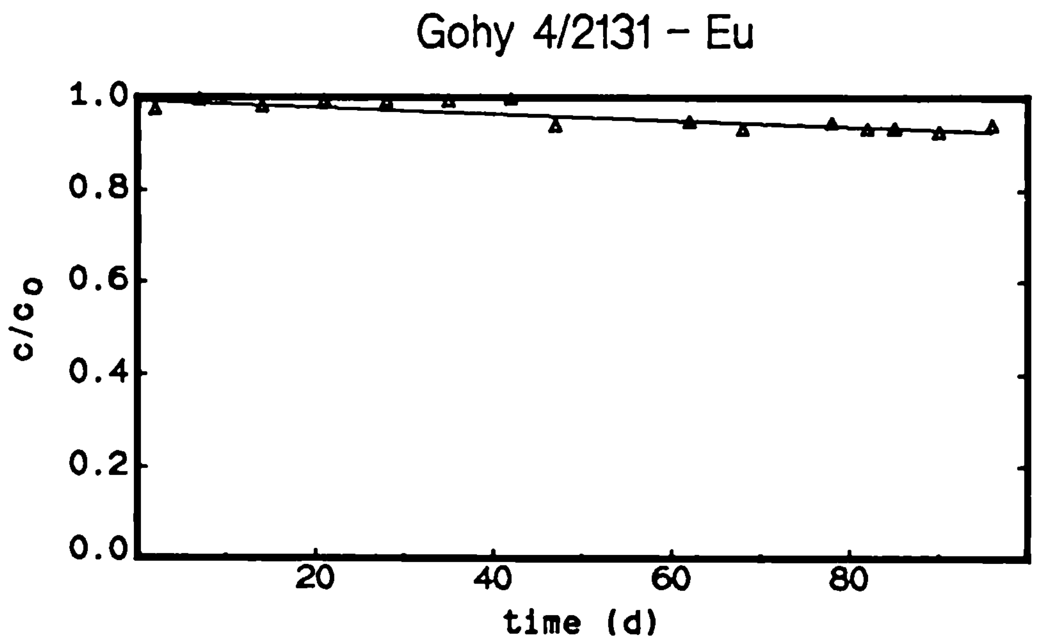
- not evaluable

FIGURE 1: STABILITY OF ^{152}Eu IN GOHY WATER 1281 (SEDIMENT 5) AND 2131 (SEDIMENT 4)

c_0: radionuclide concentration added

c : radionuclide concentration after filtration by 0.45 µm





CHARACTERIZATION OF ACTINIDES, COMPLEXES AND COLLOID MIGRATION IN GRANITE (EL BERROCAL)

Contractor: ENRESA, MADRID, Spain
Contract No.: FI1W230
Duration of contract: December 1988-November 1990
Period covered: December 1988-November 1990
Project leader: Julio Astudillo

A. OBJECTIVE AND SCOPE

The objective of the study is to characterize the migration processes in a fissured granitic formation by means of in situ and comparative laboratory experiments.

The studies will be conducted at El Berrocal uranium mine, located in a granitic batholith of the "Sistema Central" of Spain.

After an extensive characterization of the El Berrocal batholith, the natural migration processes of uranium from the intergranitic uraniferous veins and its desintegration products will be studied by sampling and analysing the natural colloids and the groundwaters. The objectives of this phase are to clarify the migration processes which have naturally occurred, as well as the system explanatory model, so that the sensibility of all different parameters involved in the radionuclides transport can be elucidated.

Granite and fissure material samples will be used in laboratory columns, under oxic and anoxic conditions, to investigate the migration of uranium in fissures and its diffusion through the rock matrix. The results will be compared with those of the in situ observations.

In parallel with the natural migration studies in the Berrocal, stability complexes and colloids analysis will be developed with natural uranium-humic acid complexes obtained from uraniferous lignites.

B. WORK PROGRAMME

1. Characterization of the Berrocal granitic Batholith: Full geological characterization; sampling from boreholes, galleries and mineralized fissures; physical parameter and natural radionuclide analysis and mineralogical and geochemical characterization.
2. Characterization of the liquid and colloidal environment: Sampling and analysis for ionic and colloidal phase (filtration, dyalisis, size distribution and in situ measurements of the physico-chemical parameters.
3. Stability analysis of actinide humic acid colloids and complexes: Isolation and characterization of uranium humic acids of other spanish sites, detailed characterization and stability analysis.
4. Laboratory migration experiments: Development of column migration apparatus, with preliminary experimentation, migration experiments in oxic and anoxic conditions in rock matrix and fissures in several physico-chemical conditions.
5. Data analysis using conventional models for chemical equilibrium and transport, to compare laboratory and in situ migration results.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The site preparation work: 4 Km. of access road to the site, cleaning of the existing gallery and excavation of 40 meters of new gallery into the fresh rock, away from the mineralized zone, have been completed. The tasks of the research work in progress are:

- Characterization of the batholith near by the mineralized vein.
- Set up of the analitic techniques for both the in situ and laboratory measurements with respect to the sampling and characterization of the organic and colloidal phases.
- Stability analysis of the humic-organic complexes, starting by the sampling of uraniferous lignite deposits with different degrees of metheoric alteration (oxidation), and the subsequent lixiviation of the organic fraction and humic acids.

Progress and results

1. Characterization of the Berrocal site

Geology: The Berrocal is a calc-alkaline two micas granite (mostly muscovite) stock. Microlyne crystals are also present. The grain size is medium to coarse.

The Berrocal granite intrudes the adamellitic granite of the region. It is cross-cut by an 80 meters wide by 20 Km. long aplitic dike, older than the Berrocal granite.

Some leucocratic outcrops can be observed in the topographic heights of the area.

Petrology and mineralogy: The mineralogical composition of the fresh granite is: Quartz (35%), Plagioclase (31%), Potash Feldspar (27%), Muscovite (4%), Biotite (3%). The altered granite, near the mineralization, shows a lower content in plagioclase and an increase in sericite (28%).

Geochemistry: The geochemical composition is shown in Table I. The Berrocal granite may be classified in an alkaline peraluminic granite or hipocalcic granite. The altered granite shows an increase in K_2O and H_2O^+ and a decrease in ONa_2 content. With regard to trace elements, the high content in Li confirms the highly evolved nature of this granite. The altered granite presents an important increase in Ba and Zn and a decrease in Li.

The geochemical background of uranium (17,6 $\pm$ 2,21 ppm, and 8,6 $\pm$ 0,8 ppm for thorium (U/Th $\simeq$ 1,80)

Near to the ore vein, the concentration of uranium ranges between 5 and 17 ppm. The uranium lixiviation in the minerals is: 5-42% in refractory minerals, 10-70% in clay minerals and 70-100% in iron minerals.

Tectonic: The remote sensing analysis of the zone, supported by field checking, shows a major system of fractures striking N110º, a less developed N-S trending system, and a third one with N50º to N70º orientation.

2. Characterization of the liquid and colloidal environment

The liquid phases characterization shows an uranium content of 100 ppb in the water in contact with the mineralization and 1 ppb in the water of the granite not affected by mineralization. The uranium in the water is present mostly as uranil carbonated complexes and also as uranium hydroxides.

3. <u>Stability analysis of actinide humic acid colloids and complexes</u>

The results of the characterization of the uraniferous lignites sampled and the analysis of the humic acids lixiviated from those lignites are shown in Table II.

4. <u>Laboratory migration experiment</u>

Permeability and porosity analysis have been done in samples of granite taken in the gallery. The results of porosity obtained with water, mercury and helium indicate two types of rock as related to the degree of alteration. A systematic sampling of granite cores, 20 cm long by 2 cm in diameter, has also been done to test the design of the column migration experiments.

TABLE I: BERROCAL GRANITE COMPOSITION

MAJOR COMPON.	FRESH GRANITE (%)	ALTERED GRANITE (%)	TRACE ELMTS.	FRESH GRANITE ppm	ALTERED GRANITE ppm
SiO_2	75,2	75,4	Ag	<5	<5
Al_2O_3	13,5	13,5	As	25	<25
Fe_2O_3	0,39	0,50	Ba	34	50
Fe O	0,69	0,52	Be	<5	<5
TiO_2	0,078	0,08	Ce	24	26,5
MnO	<0,03	<0,03	Co	<5	<5
MgO	0,16	0,38	Cr	17	15,85
CaO	0,48	0,40	Cu	<5	-
K_2O	4,30	4,78	La	5,8	7,62
Na_2O	3,3	1,82	Mo	5,2	5,04
P_2O_5	0,26	0,29	Ni	5,7	5,07
SO_{2t}	0,03	0,02	Sr	5,8	8,58
H_2O^+	0,90	1,37	V	5,5	5,78
H_2O^-	0,27	0,52	W	<25	<25
TOTAL	99,75	99,74	Y	6,5	7,7
			Zn	89	135
			Li	230	127
			Zr	63	64
			Rb	460	466
			Th	8,6	8,9
			U	17,6	18,4

TABLE II: BERROCAL LIGNITES COMPOSITION

COMPONENTS	TYPE I	TYPE II	HUMIC ACIDS TYPE I
H_2O (%)	13,1	12,6	
Ashes (%)	48,1	45,0	11,24
S ($SO_4^=$) (%)	<0,1	0,198	
Volatile Components (%)	39,0	38,8	
Fe^{2+} (%)	0,87	1,06	
C total (%)	26,6	27,8	45,96
C organic (%)	25,5	26,8	
$CO_3^=$ (%)	5,5	5,5	
$S^=$ (%)	<0,1	<0,1	
Cl^- (%)	0,03	<0,01	
U (ppm)	1180	134	566,7
pH	5,5	6,9	

IMPROVEMENT OF COLLOID SAMPLING TECHNIQUES IN GROUNDWATER AND ACTINIDE CHARACTERISATION OF THE GROUNDWATER SYSTEM AT GORLEBEN (FRG) AND EL BERROCAL (E)

Contractor: Harwell Laboratory, UKAEA, UK
Contract No.: FI1W/0234/UK
Duration of contract: October 1988-March 1990
Period covered: October 1988-December 1988
Project leader: M. Ivanovich

A. OBJECTIVES AND SCOPE

The apparent radionuclide solubilities in groundwater may be enhanced by their association with colloids, so that a realistic model for the retention of actinides by a radioactive waste repository requires the knowledge of the radionuclide loading of the geocolloids as well as the extent of their transport through the geosphere. Ultrafiltration techniques have been developed to collect groundwater samples in the field, and their component phases, particulate, colloid and aqueous solution phases. The research described hereafter is concerned with uranium studies of natural colloids designed to quantify the effects of actinide/colloid association on the past and future migration of radionuclides at Gorleben (FRG) and El Berrocal (Spain). The aims are as follows:

To sample natural colloids from several boreholes at the above sites in collaboration with Technical University, Munich and ENRESA/CIEMAT staff.
To characterize particulate, colloid and solute phases in terms of their physical, chemical and actinide composition.
To use artificial colloids labelled by artificial radionuclides to study in greater detail the uptake characteristics of the ultrafiltering system used.

B. WORK PROGRAMME

1. Sampling and characterisation of Gorleben groundwater in terms of actinide inventory and physical, chemical characterisation of geocolloids, with intercomparison exercise (Harwell/TUM).

2. Study of colloid uptake on tangential flow ultrafilters using model colloids.

3. Characterisation of El Berrocal groundwater in terms of actinide inventory and physical, chemical characterisation of geocolloids.

4. Active participation in CoCo club both in terms of defining future actions and carrying out future benchmark exercises.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

A field trip to the Gorleben site, FRG was carried out in the period 12-15 December 1988 in order to collect groundwater samples. Two boreholes were selected for sampling on the basis that they represented extremes in water chemistry, and had suitably high flowrates. Borehole 2227 had a high organic content (~100 mg/L DOC) while Borehole 1231 had a high chloride content (7900 mg/L). Samples of unfiltered groundwater from a depth of ~130 m were collected in 50 L platinised aluminium barrels and samples of aqueous colloid concentrate were obtained on site using tangential flow ultrafiltration rigs (Amicon DL10LA and CH2PA) under anaerobic conditions (N_2 + 1% CO_2 atmosphere). Samples were returned to Harwell and Munich for analyses.

Progress and results

1. Sampling and characterisation of Gorleben groundwater

A summary of samples collected is given in Table I together with an indication of the analyses to be carried out. The analyses are currently in progress at Harwell and Munich (TUM) and relevant measurements will be subjected to an intercomparison exercise by the two teams in the early spring 1989.

TABLE I: SUMMARY OF SAMPLES COLLECTED FROM BOREHOLES 2227 AND 1231 IN DECEMBER 1988 FROM THE GORLEBEN SITE WITH DETAILS OF ANALYSES TO BE CARRIED OUT AT HARWELL

SAMPLE DESCRIPTION	QUANTITY (litres)	ANALYSES
Unfiltered source	100	Laboratory study
Unfiltered source	20	USD, ICP, TOC, SEM, MOB, PCS, LIPAS
Prefiltered (1 μm filter)	20	USD
Colloid concentrate	20	USD, ICP, TOC, SEM, MOB, PCS, LIPAS
Colloid concentrate 1 μm–0.1 μm	6	USD, SEM, ICP, TOC
Colloid concentrate 0.1 μm–5.4 nm	4	USD, SEM, ICP, TOC
Colloid concentrate 5.4 μm–1.5 nm	6	USD, SEM, ICP, TOC
Ultrafiltrate	20	USD, ICP, TOC, SEM, MOB, PCS, LIPAS
Unfiltered	5	^{222}Rn and ^{226}Ra content
Prefiltered	5	^{222}Rn and ^{226}Ra content

USD = Uranium series disequilibrium measurement (U/Th isotopic ratios)
ICP = Inductively coupled plasma optical emission mass spectrometry
TOC = Total organic carbon
SEM = Scanning electron microscopy
MOB = Colloid charge/mobility configuration by electrophoresis
PCS = Photo correlation spectroscopy
LIPAS = Laser induced photoacoustic atomic spectroscopy

4.3.B. In situ migration experiments and development of measuring techniques

IN SITU DETERMINATION OF THE EFFECTS OF ORGANICS ON THE MOBILITY OF RADIONUCLIDES IN CONTROLLED CONDITIONS OF GROUNDWATER FLOW

Contractor:	British Geological Survey, Keyworth, Nottingham
Contract No:	FI-1W-0064(UK)
Duration of Contract:	from July 1986 to June 1989
Period covered	from January to December 1988
Project Leader:	G.M.Williams

A. OBJECTIVES AND SCOPE

The broad objective is to verify by means of *in situ* field tracer tests, predictions of the mobility of radionuclides in a shallow glacial sand aquifer, having taken into account the potential effects of organics (natural and introduced) on radionuclide speciation and mobility. The results will be used to improve the prediction of radionuclide migration in the geosphere and the overall safety assessment of waste disposal facilities. Liaison has been established with Loughborough University (LUT) for direct speciation measurements, the University of Wales College of Cardiff UWCC (formerly Institute of Science and Technology (UWIST) for radionuclide speciation modelling, and Delft Geomechanics for solute transport modelling.

B. WORK PROGRAMME

(1) Aquifer characterisation and instrumentation - Involves the determination of aquifer hydraulic properties, its geochemistry, mineralogy and groundwater composition, particularly the nature and amounts of natural organics (humic and fulvic acids) and colloids.

(2) Characterisation of complexes and colloids - interlaboratory comparison within the CEC, to characterise, and determine stability constants with selected radionuclides, for commercially available humic acid and natural organics

(3) Laboratory sorption studies - includes various sorption experiments to determine the effects of natural organics on radionuclide sorption, kinetic measurements, and direct speciation determinations of radionuclide complexation in groundwater after equilibration with the sediment.

(4) Modelling - Speciation models will be used to predict the speciation of radionuclides in the sorption experiments and help to determine the important mobile species in the field test.

(5) Field tracer experiments - initial tracer tests will compare conservative tracers (^{131}I, Cl, and ^{3}H) Subsequent tracer tests will involve reactive radionuclide species with organic solutes.

C PROGRESS OF WORK AND RESULTS OBTAINED

STATE OF ADVANCE

Phase 2 of the work involving field tracer tests has continued with a second experiment using ^{131}I and ^{3}H. Natural organic material has been extracted from Drigg groundwater but < 1 g has been isolated. Work is progressing to extract a much larger sample for distribution to the COCO participants for characterisation.

PROGRESS AND RESULTS

(1) Aquifer characterisation and instrumentation (B1)

The ^{131}I and ^{3}H test was performed satisfactorily using an *in situ* g detector attached to a winch controlled by a IBM-PC. Comparison between multilevel samplers (which sample a small volume of aquifer) and the *in situ* g detectors (which are very sensitive and "see" a large volume) indicate the effect of the sampling device on the form of the breakthrough curve (BTC). The *in situ* g detector imparts a wide dispersion on to the BTC and collimation is necessary to reduce the size of the detection field. Two additional wich systems are now being developed along with borehole collimation.

(2) Complexes and colloids (COCO excercise, B2)

A large volume of water (325 l) removed from the array has yielded 60 mg humic acid and 800 mg of fulvic acid (FA). This is equivalent to a recovery of about 60% of the TOC in the water. Ultra filtration of the Drigg FA and GOHY HA shows that 80% of the FA and 67% HA is retained by a 10,000 Dalton membrane contradicting the idea that FA is generally smaller molecular weight than HA. 97% of Drigg FA and 99.5% of GOHY HA were rejected by a 500 MW membrane. In both cases the ratios of measured uv absorbance at 300 and 254nm were maintained before and after filtration, suggesting that the filtrate and retentate were materials of similar structure. A further anomaly was provided by work with oven dried Drigg fulvic acid - on redissolution 52% of this material passes through a 10,000 Dalton filter. Work is progressing to evaluate other methods for extracting natural organics using XAD and other resins, and ultra filtration extracts are being characterised by HPLC with UV absorbance monitored by a photodiode array detector.

AERE Harwell have sampled Drigg groundwater for colloids using dia-ultrafiltration and a report on thsi work was presented at the MRS conference in Berlin Oct 88 (Longworth et al.)

(3) Laboratory sorption studies (B3)

Cobalt migration has been studied in small-scale laboratory columns designed to provide a controlled intermediate test between laboratory batch and field tracer experiments. In particular the columns provide data on the stability of the cobalt- FA complex in order to demonstrate the feasibility of utilising cobalt in a field tracer test, as well as to validate R_d values estimated from batch sorption experiments. The Co was pre-equilibrated with the groundwater concentrated to x 10 by rotary evaporation. Only 6% of the input cobalt concentration was recovered compared with 76% of the iodine. At the end of this experiment the column was sectioned and the core segments analysed; the cobalt concentration in the sediment was found to decrease almost exponentially from the input. Assuming that only uncomplexed cobalt, as Co^{2+}, is sorbed by the sediment, and that the rate at which the cobalt-groundwater complex dissociates controls the rate of production of uncomplexed cobalt, the sorbed cobalt concentration gradient along the column should be a simple function of the dissociation constant for the groundwater complex and the solute transport velocity. Initial estimates for the rates of complex dissociation, are very fast, and greater than those obtained from individual batch kinetic experiments. It is thought that the difference in rates between batch and column experiments may be a reflection of the differing solid:solution ratios. If the rates of complex dissociation estimated from the BGS column experiments are correct and the sorption coefficient for Co^{2+} is of the order of 200ml/g, it would take about 8000 hrs (i.e. about one year) before cobalt was detected at the nearest sampling point in the field array although a very low peak of Co-FA complex (4% of initial concentration) would be expected after 10 hrs. This rate of transport would make a fully-manned field experiment imprtactical. As a result, column experiments are now in progress with a Co/EDTA complex. Initial results show that the free EDTA elutes with a slight retardation compared with ^3H but that the Co-EDTA complex is retarded by a factor of about 8. EDTA also complexes with other cations (Ca, Mg, Fe) and the mobility of these complexes is also similar to Co-EDTA.

Work with Loughborough University has progressed using Sephadex gels to develop non-invasive techniques to separate bound from free radionuclides in Drigg groundwater. Chemical analysis of uv-absorbing fractions collected during Sephadex separation has been completed using ICP-OES and IC. Samples of 10x concentrated groundwater eluted from Sephadex G10, corresponding with the uv-absorbing peaks at 210nm, have been analysed for major inorganic species. The Sephadex gel excludes larger ions and complexes so that the larger ions are eluted prior to the smaller. In order of elution the uv absorbing peaks were found to correspond with (i) sulphate at 130mg/l, cf. 25mg/l in the input 10x concentrated groundwater, together with minor Ca, Sr, Mg, Ba and Mn; (ii) chloride at 140 mg/l (32 mg/l input) and sodium at 40 mg/l (25 mg/l input), together with major amounts of Ca, Sr, Mg, Ba and Mn, probably as chloro-complexes; (iii) silicon at 8 mg/l (2mg/l input); (iv) sodium at 60 mg/l (25 mg/l input) and (v) potassium at 6 mg/l (1 mg/l input). The second sodium peak at (iv) probably corresponds to the free ion. Iron concentrations were below the analytical limit of detection (ca. 0.01 mg/l) in all of the samples and TOC/TIC determinations are awaited. The major cobalt peak resolved by Sephadex G10 lies at the uv peak (ii). Further work is planned using G15. Further consideration had also been given to the species separated by G15 which resolves the cobalt (gamma) spectrum into two main peaks, rather than into a peak and shoulder as seen with Sephadex G10. Studies have also been undertaken into the effect of temperature and pH on Co sorption. The rapid loss of Co complexing ability in Drigg groundwater at pH < 6 suggests that the binding may be due to a weak electrostatic surface interactions rather than true chemical complexation.

4. Modelling (B4)

Geochemical modelling of Drigg groundwater has been undertaken jointly by BGS/ UWCC. The results show (a) most of the non-transition elements will be present as free ions; (b) most of the trace elements are predicted to be present as $CO_3^=$ complexes; (c) $CaCO_3$ will not be oversaturated in the pH range 5.5-8.0 although $Fe(OH)_3$ precipitation is predicted at high pH. The switchover point from Fe^{2+} as the dominant solution species occurs between pH 7 and 7.5, above Eh +170mV, when $Fe(OH)_3^\circ$ becomes the major solution species, and precipitation of amorphous ferric hydroxide becomes possible; (d) the only other element which may change speciation over the pH and redox range of 5.5 to 8.0 and -150 to +250mV respectively, is sulphur, although sulphate is most likely to dominate the solution phase. Modelling cobalt speciation in Drigg groundwaters using preliminary pH (from 7.0-8.5) and Eh (from -150 to +200 mV) data from pumping tests in the array suggested that under natural conditions (pH 6.8-7.2) about 98% of the total cobalt would be present as Co^{2+}, with about 2% as $CoSO_4^\circ$. From the standard potential of the Co(III)/Co(II) couple it can be predicted that below +180 mV less than 1% of the cobalt would be present as Co(III) species, if no organics were present. This predicted speciation would be virtually unaltered by the addition of 10^{-10}M EDTA at a cobalt concentration of 10^{-9}-10^{-10}M. A review of modelling cation interactions with humic compounds has been produced by BGS.

Solute migration in the tracer test has been analysed using a one-dimensional simulation of an exponential decay source, formulated by Van Genuchten and Alves. Type curves had been generated for various Peclet numbers, and R* values, where R* is a function of tracer source decay rate, retardation velocity, distance and dispersion length. The breakthrough data at various sampling points have been visually fitted to the type curves to give characteristic solute velocities and dispersion lengths. These showed that the longitudinal dispersion values of D_L ranging from 5 x 10^{-2} to 1 x 10^{-3} m. None of the breakthrough curves, even in the main flow path, showed peak heights comparable with those predicted by the one-dimensional model, suggesting that tracer 'loss' is occurring due to lateral dispersion. This shows that a one-dimensional model does not describe the observed solute migration behaviour adequately, and that account must be taken of dispersion transverse to the direction of flow.

5. Tracer tests (B5)

Following a tracer test with non active chloride a second tracer test has been performed using ^{131}I and ^{3}H under exactly the same hydraulic conditions. Results show that ^{131}I moves approximately at the same rate as chloride although the detection of the latter was associated with much more scatter. ^{3}H however was slightly delayed relative to the ^{131}I.

It is proposed that the next tracer test will be carried out using Co-EDTA complex since sorption work with Eu, Np, and U show that these elements are still strongly sorbed under most conditions studied, and suitable short lived isotopes are not available. Other organics derived from cellulose breakdown are also being considered in addition to acetate and EDTA, to determine wheteher they will significantly affect radionuclide mobility and may be considered in a future field tests although this would have to be the subject of a new proposal to CEC/DOE.

D <u>REPORTS</u>

A large number of reports are in the state of advanced preparation.

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Title : DEVELOPMENT AND APPLICATION OF A RETENTION PROPERTIES MEASUREMENT SYSTEM IN A GEOLOGICAL ENVIRONMENT USING RADIOACTIVE TRACERS IN THE DRILL-HOLE (self-contained probe FORALAB)

Contractor : CEA/IPSN CEN CADARACHE
F 13108 St. Paul-lez-Durance

Contract No. : FI1W/065

Working period : November 1986 - November 1989

Project Leader : J. PORCHERON

A. OBJECTIVES AND SCOPE

Radionuclides from a subterraneous waste storage place have to force their way through, and interact with, several barriers prior to reaching the geological medium itself.

They are diluted by the suberraneous water which, by modifying their chemical structure, settles them into a final balance with the medium.

The purpose of this study is to determine the delay term of the radionuclides during their migration through the deep geological environment.

It became evident that it was preposterous to attempt in-laboratory duplication of the prevailing parametric conditions of the natural medium, whether physical, chemical or biological.

To avoid the uncertainties connected to laboratory experiments, the probe "FORALAB", whose performances had already been ascertained during the preceding contract (WAS 377-83-7), was developed to permit studying the radionuclide sorption-desorption phenomena in a geological environment in a condition of equilibrium with undisturbed subterraneous waters.

The probe need not simulate the environment as it is plunged into it.

The contract scope is the *"in-situ"* qualification of the probe, using a dual tracing system, i.e. Tritium and Eu on the one hand, Pu and Np on the other hand.

The probe will then be operated in 3 geological sites, of some interest for the Community, i.e. AURIAT (granite), MOL (clay) and GORLEBEN (salt), in order to assess the containment properties of these environments,

The probe is essentially composed of a pump, a syringe, a 20 mm dia., 200 mm long test column and 40 sampling pots. Its double insulation is a safety against drill-hole pollution.

B. WORK PROGRAM

B1. Probe Qualification

The probe will be checked for performance in the hole drilled in the granitic site of AURIAT.

The column, filled with Fontainebleau sand, will be traced by means of Europium and Tritium.

The drill-hole water will be circulated in the column for one day before the tracer injection.

B2. Tests on the Reference Sites

The tests will be performed in the deep holes at AURIAT, MOL and GORLEBEN.

The columns will be filled with a mixture of Fontainebleau sand and 1 to 3 % clay from the site.

The radionuclides used will be am, Pu and Np.

Each column will be Tritium-calibrated before each individual test.

B3. Finally the results from each individual test will be mathematically processed to yield the delay terms and the adsorption isotherms of each pollutant used.

C. WORK PROGRESS AND RESULTS OBTAINED

C1. Work progress

The development of the probe will be continued in situ in order to resolve the many problems associated with the behavior of certain constituents which enabled the probe to operate for several hours but not for sixty. The following modifications were carried out: an automatic degassing system was fitted to the pump, the solenoid valves were changed, a radiator was fitted and the instrumentation and control was modified in order to make the probe independent of telluric interference.

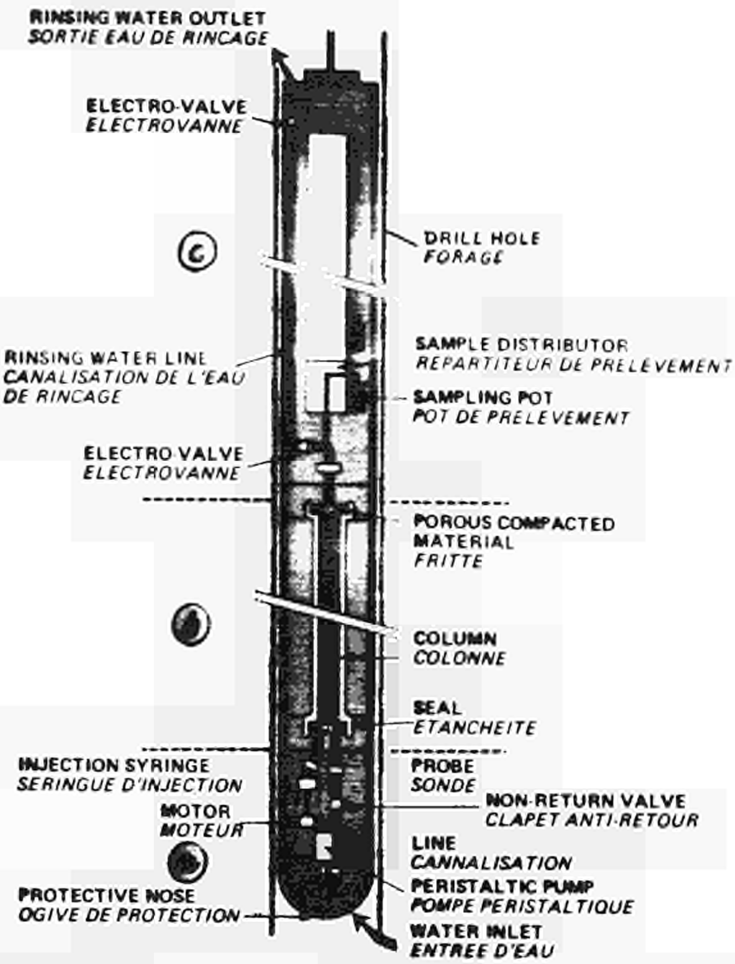
The probe is now operational.

Injection tests into a column of Fontainbleau sand showed that the Europium Oxide remained in the head of the column.

The characteristics of the columns filled with unweathered, crushed granite were established in the laboratory.

C2. Drawings

The unweathered, crushed granite tests will be carried out in 1989.



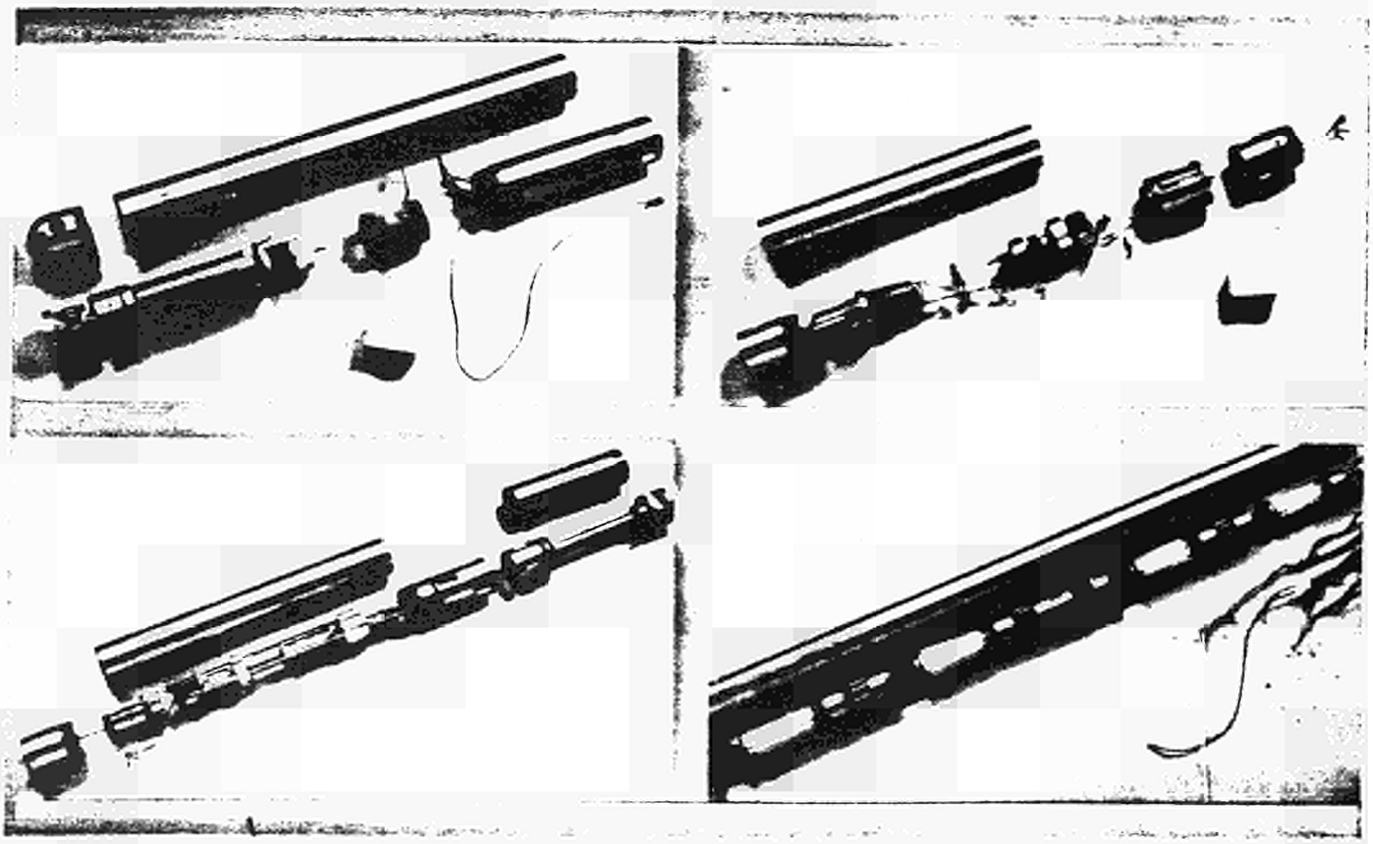


Schéma de principe et vue du système FORALAB

FIELD VERIFICATION OF ADVANCED TRANSPORT MODELS FOR RADIONUCLIDES IN HETEROGENEOUS SOILS

Contractor : Delft Geotechnics, Netherlands
Contract No. : FI1W/0083-NL
Duration of contract : from march 1987 to december 1989
Period covered : march 1987 to januari 1989
Project leaders : Dr. Ir. M. Loxham, drs. W. Visser

A. OBJECTIVES AND SCOPE

The retardation of contaminants by adsorption on soil components such as clay and iron oxides is usually the most important mechanism that has to be taken into account when making the safety assessment of a toxic landfill, be it of radioactive components or other chemicals. The mobilisation of the retardation capacity in any given soil profile is not only a function of the mass of adsorption sites but also of the availability in the flow situation considered. In other words, the effective retardation capacity is a function of the structure or heterogeneity of the soil on the length scales relevant to the problem. Earlier studies by the present researchers, supported in part by the EEC (phases I and II) have demonstrated the key importance of these phenomena both theoretically and at the bench scales.

The objective of this study is to verify the concepts developed in the earlier programs at a field scale using the data generated by the communities investigation conducted by the British Geological Survey (BGS) at Drigg in the United Kingdom. Whilst the study is directly related to the problems associated with the geosphere safety assessment technologies for the shallow burial of low level radioactive waste, the problem definitions, solution methodologies and problem solving experience are directly applicable to all toxic waste and contaminant migration studies.

B. WORK PROGRAM

I. Data collation and critical review.
II. Predictive modelling of the field experiment
II.1 Calculations at level 1 (Homogeneous Approximation)
II.2 Calculations at level 2 (Stratified Approximation)
II.3 Calculations at level 3 (Fully determinate case)
III Evaluation of the Modelling Exercise
III.1 Inter-model level comparison
III.2 Comparison with the results obtained from the Drigg experiments

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Owing to some initial startup problems at the test site at Drigg, field data from the BGS only became available to Delft Geotechnics in the course of 1988. At a request from Delft Geotechnics an extension of the duration period of the contract with 1 year (till 31-12-1989) has therefore been granted by the Commission. At this moment data on geological stratification, site layout, near- and local field permeability and porosity distribution are available. Tracer-test data with chloride are also available. After critical review the data in our possession have been used for first and second level calculations for a conservative tracer. Comparision of the results of the first calculation with the chloride tracer test results indicate that the scale of the experiment at Drigg is not optimal for meeting the original objectives of the study. Presently a meeting is being set up between BGS and Delft Geotechnics in order to discuss the possibility of optimising the tracer array with respect to reaching the original goals of the project. Possibly some of the objectives will have to be altered, of course in close consultation with the Commission.

progress and results

I. DATA REVIEW

Available data can be grouped into 4 types:

- data on hydrogeologic setting and system dimensions.
- data on soil properties.
- data on flow conditions during tracer test
- data on tracer injection and breakthrough

The tracer array is situated in a thin sand aquifer wedged in between glacial till deposits. The system can therefore be considered as confined. Measured groundwater heads under undisturbed conditions show there's normally virtually no groundwaterflow. The dimensions of the system are given in Figure 1.

Main soil properties of interest with respect to groundwater flow and transport of non-adsorbing (conservative) tracers are permeability and porosity distribution. Besides content of clay, iron oxide and organic matter are key parameters if adsorbing tracers are considered. Permeability is known on a near field scale (5 - 10 m from the array) at three points. This gives some idea about lateral distribution of mean aquifer permeability. Furthermore the vertical distribution of the permeabilities is known at two points about 2 m from the actual tracer array. Averaging local permeabilities at these two points gives a value of mean aquifer permeability which is well within the range of measured avarage aquifer permeabilities at the near field scale. Local and near field permeability distributions are reflected in Tabel I. The vertical porosity distribution is measured in the tracer array. There's only a relatively small variation. Mean aquifer porosity is 31.6 % (n=8, s=2.73 %). At the moment there's no information on content of clay, organic matter and iron oxides.

During the tracer test pumped volumes and groundwater heads were monitored. Using these data the Thiem-Dupuit's well formula was used to optimize with respect to the mean transmissivity of the aquifer in the sphere of influence of the test. This gave a mean permeability of $7.9*10^{-6}$ m/sec, which is well within the range of the other measurements (Table I). The calculated heads gave a good fit with the measured heads. It showed that between tracer injection and furthest multi level sampler (m.l.s.) a constant gradient can be assumed ($\Delta h/\Delta x=0.27$).

Tracer injection was monitored by cloride concentration measurements in the tracer injection well. Breakthrough of chloride was measured at 3 multi level samplers (for position see Figure 1). At each multi level sampler groundwater samples were taken at 12 levels at a certain time interval and analysed on cloride. Average breakthrough was not monitored. The radioactive tracers will be monitored directly by way of gamma-probes.

II. PREDICTIVE MODELLING OF THE FIELD EXPERIMENT

II.1 Homogeneous approximation

Using a best parameter estimate the breakthrough of chloride was simulated analyticaly /1/ as well as numericaly /2/ (Tabel II). The results are shown in Figure 2. The agreement between numerical and analytical results is good.

II.2 Stratified approximation

To check whether the top and bottom clay layers are of any concern for the tracer migration behaviour an analytical solution /3/ was used which assumes only convection in the sand and only diffusion in the clay. Dispersion in the sand is not considered. Due to the higher rate of convection in the sand compared with diffusion in the clay the results showed no influence of the clay on the calculated breakthrough curve. For the given flow regime influence of the clay will only become significant with tracers which show a very strong adsorption behaviour on clay. Checking the use of fissure models /4/ as a meaningful approach in risk assessment of a toxic waste dump or shallow burial site using data from the current tracer test setup will therefore be impossible. In a second approach only the sand aquifer was considered. The system was modelled as a series of horizontal strata with different permeabilities. Because there is no information on the vertical permeability distribution in the line of the tracer array itself the information had to be obtained by way of interpolation between two points at some distance from the array. Calculated chloride breakthrough was generated at the position of the different minifilters.

III. EVALUATION OF THE MODELLING EXERCISE

III.1 Inter-model level comparison

Using the stratified approach also an avarage breakthrough curve was generated. This is compared with the homogeneous approximation in Figure 3. The effect of preferent migration through the two most permeable layers is evident, which indicates the shortcomings of the homogeneous approximation in this case. At this stage the database is not sufficient for attempting a fully determined approach.

III.2 Comparison with the results obtained from the Drigg experiments

Unfortunately the avarage breakthrough was not monitored during the experiment so it is not possible to compare the homogeneous calculations with field measurements. For port 10 the calcultated breakthrough of chloride in the stratified approach is compared with the measured breakthrough (Figure 4). Considering the uncertainties in measured permeabilities the fit is not bad. Port 10 is located in one of the two most permeable layers which dominate the system. For the second permeable layer (port 5) no breakthrough data are available. The fit for the less permeable layers is not as good. It seems that stratified modelling of the tracer experiment requires more detailed information on local permeability distribution then is available at this moment.

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TABEL I: AVERAGE AQUIFER PERMEABILITIES

	log K (m/sec)	minimum	maximum
near field (D202+D203+204)	-5.323	-5.569	-5.119
local field (D205+D207)	-5.435	-6.569	-4.745
Thiem-Dupuit	-5.101		

TABEL II: BEST PARAMETER ESTIMATE

Hydraulic conductivity	$k = 7.919 \cdot 10^{-6}$ (m/sec)
Mean aquifer porosity	$n = .32$
Hydraulic gradient	$i = 0.27$ (m/m)
Longitudinal dipersivity	$\alpha L = 0.0155$ (m)
Diffusion coefficient	$D_s = 5.5 \cdot 10^{-11}$ (m^2/sec)

FIGURE 1

DRIGG SITE TRACER ARRAY LAYOUT

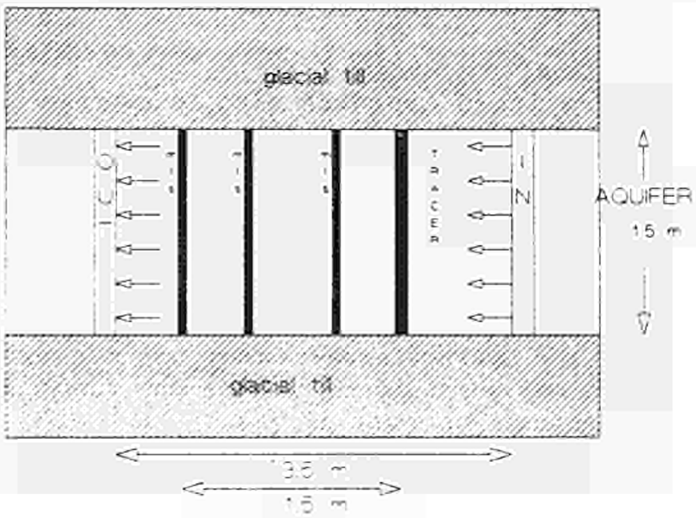


FIGURE 2

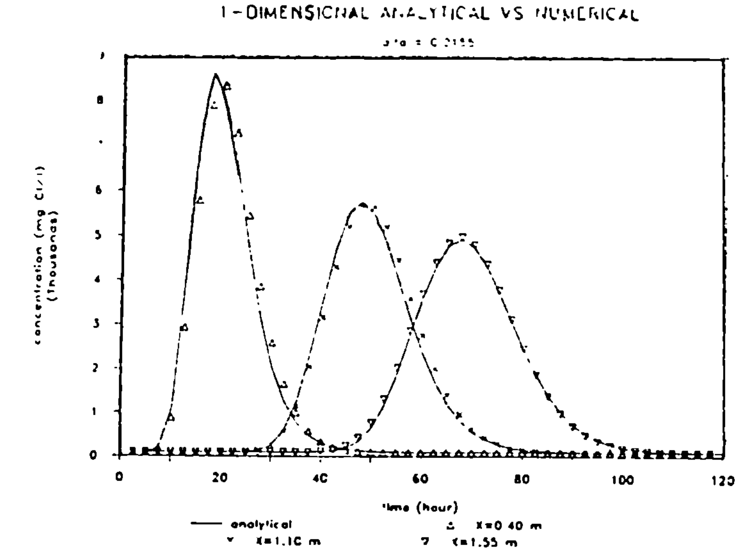


FIGURE 3

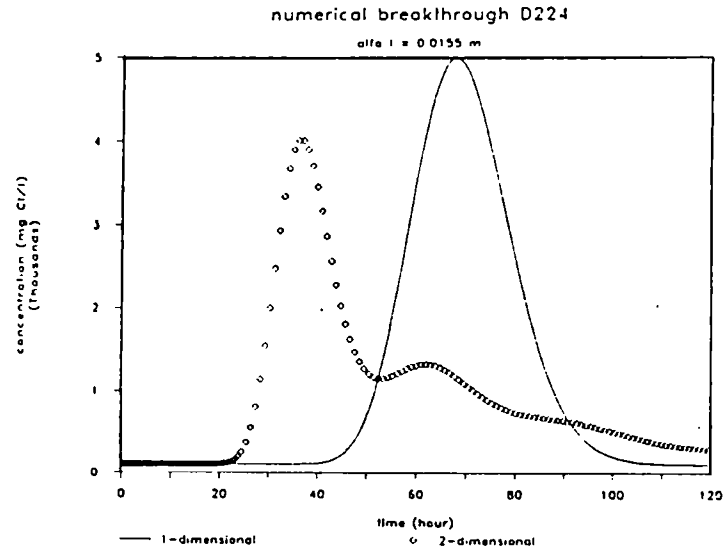
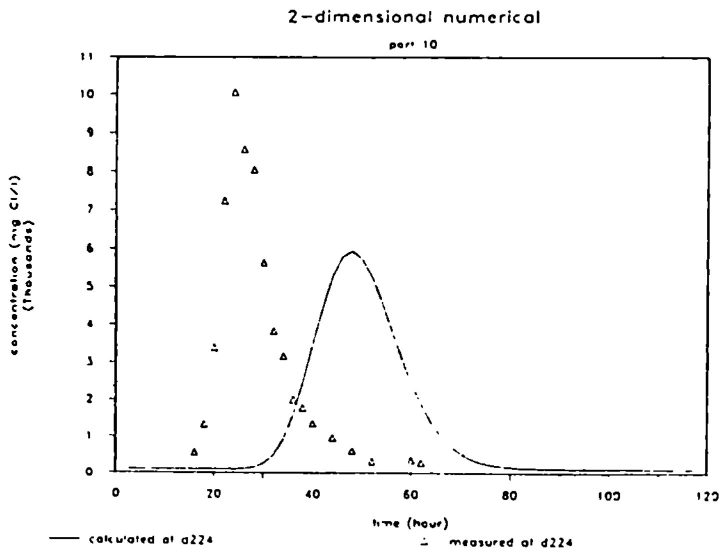


FIGURE 4



Title : *IN-SITU* STUDY OF RADIONUCLIDE DIFFUSION IN CLAYS BY MEANS OF THE AUTOLAB PROBE

Contractor : CEA/IPSN CEN CADARACHE
F 13108 St. Paul-lez-Durance

Contract No. : FI1W/0144

Working period : January 1988 - July 1990

Project Leader : J. PORCHERON

A. OBJECTIVES AND SCOPE

The storage of nuclear wastes in a deep geological formation calls for that a dependable knowledge of the site safety is gained. Significant data on the radionucléide diffusion throughout argillaceous materials should be acquired by *in-situ* observations and measurements in a drill-hole, provided that the conditions of the receptor environment are safeguarded.

In the field of the *in-situ* measurements, two families of instruments[1] presently exist or are under development :

a) Equipment making it possible a direct measurement of the environment characteristics, such as pH, Eh, Ca^{++} content, temperature, radio-activity, resistivity. Those probes make it possible to obtain a logging of the previously-mentioned parameters in a drill-hole. A drill-hole chomatography probe, for the indirect measurement of chemical elements present as traces in the drill-hole, is presently being developed.

b) The second class of probes is related to the experimentation in a drill-hole to obtain the velocity of radionuclide migration in the fourth barrier. The velocity of the water which is the vector of the radioactive pollution may vary in wide proportions.
First, there is the domain of the convection-disperson which is that of the FORALAB probe presently used, second, the domain of the diffusion where the very slow progress of the phenomena requires that the *in-situ* experiment complies with special specifications.

c) Representativeness of the *in-situ* experiments

The representativeness of the *in-situ* tests is related to the drill-hole water characteristics because, as the hole is being drilled, water is used which is entirely foreign to the environment. Moreover, the hole may also fill itself with water from a higher-lying water table.

From the chemical point of view, however, as chemistry is the criterion for the radionuclide condition, an equilibrium is gradually gained between the foreign water and the geological environment with which it is in contact. There are two phases in the drill-hole, viz. the solid environment which is "in excess" and the water ; the equilibrium is reached by dissolution, the solid environment acting as a buffer.

1 CEA/IPSN - Cadarache Centre.

B. WORK PROGRAM

B1. AUTOLAB is a probe designed for measuring the delay term of the radionuclide *in-situ* in the drill-hole clays.

B2. Specificity of the AUTOLAB probe

The probe is designed for diffusion measurements, which entails particular characteristics for grappling the slowliness of the phenomena.

The specification data required from the probe are as follows :

- operability in a drill-hole without time limitation (in practice, 6 months),
- complete autonomy, no supervision, no power feed,
- good exchange between the drill-hole water and the inside of the probe under static conditions (no pump operating),
- no pollution of the environment.

B3. Description of the probe (Fig. 1)

The probe is composed of a reaction chamber, of 5 l. capacity (65 mm dia., 1500 mm high), capable of containing 10 samples (approximately 10 mm dia., 50 mm high).

The tracer is contained in a glass bulb which will be ruptured by a weight (messenger) sliding along the carrier cable.

The reaction chamber may be put in communication with the drill-hole by windows (36 cm^2 surface) which may be actuated using the drill-hole pressure. It suffices for this to lower the probe by a few meters, which acts upon a hydraulic valve held closed by a spring set at a pressure corresponding to the drill-hole depth ; the valve, when opening, permits the action of the hole water on a piston opening or closing the windows.

B4. Experiment

The probe will be used for measuring the *in-situ* migration of radionuclides in clays.

a) Equilibrium stage

In a first stage the argillaceous material samples (in 10 mm dia., 50 mm high tubes) are left to come into an equilibrium with the drill-hole water. The water diffuses into the material ; a diffusive front of equilibrium is formed and will slowly progress for several months.

b) Experimental stage

When the foregoing stage is completed, the windows get closed, the tracer (double tritium-lanthanide or actinide marking) is released from the glass bulb using the messenger (this also puts an electric cell-driven agitator into action).

The tracers are diffused into the clay. A second diffusion front is formed, offset with respect to the first one and propagating in a zone already settled to a chemical balance with the drill-hole water.

B5. Tests planned

- Qualification of the probe on the AURIAT site.

Samples are selected with various sand contents and various natures of argillaceous materials.

The tracers will be Tritium and Europium.

The experiments will take place:

. in the granitic site of PARTENAY,
. in the saline site of GO RLEBEN,
. in the argillaceous site of MOL.

Use will be made of Tritium, of an actinide or, if unavailable, of a lanthanide.

The number'and composition of the samples will be determined for each particular case, filling materials may be used, as necessary.

C. WORK PROGRESS AND RESULT OBTAINED

C1. The probe was successfully tested at Auriat (Limousin, France) in December 1988. All the sequences were used. Given the simplicity of the probe, we can reasonably expect that the final design will be perfected with few if any problems.

C2. Drawings

An experiment in an argillaceous site will be carried out in March 1989 on a drilling kindly made available to us by CCR ISPRA (Italy).

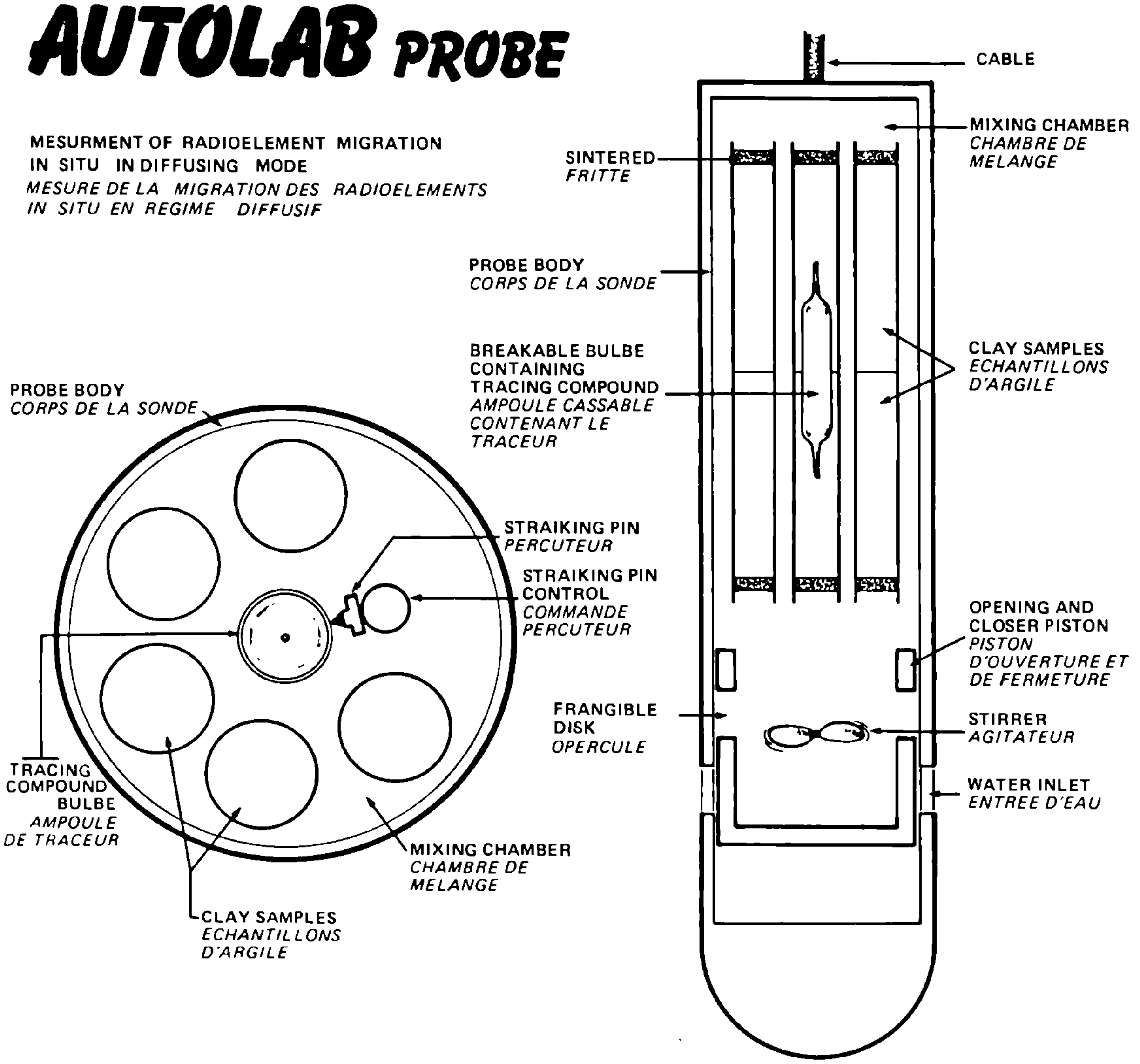

AUTOLAB PROBE
MESURMENT OF RADIOELEMENT MIGRATION
IN SITU IN DIFFUSING MODE
MESURE DE LA MIGRATION DES RADIOELEMENTS
IN SITU EN REGIME DIFFUSIF
PROBE BODY
CORPS DE LA SONDE
STRAIKING PIN
PERCUTEUR
STRAIKING PIN
CONTROL
COMMANDE
PERCUTEUR
TRACING
COMPOUND
BULBE
AMPOULE
DE TRACEUR
MIXING CHAMBER
CHAMBRE DE
MELANGE
CLAY SAMPLES
ECHANTILLONS
D'ARGILE
CABLE
MIXING CHAMBER
CHAMBRE DE
MELANGE
SINTERED
FRITTE
PROBE BODY
CORPS DE LA SONDE
BREAKABLE BULBE
CONTAINING
TRACING COMPOUND
AMPOULE CASSABLE
CONTENANT LE
TRACEUR
CLAY SAMPLES
ECHANTILLONS
D'ARGILE
OPENING AND
CLOSER PISTON
PISTON
D'OUVERTURE ET
DE FERMETURE
FRANGIBLE
DISK
OPERCULE
STIRRER
AGITATEUR
WATER INLET
ENTREE D'EAU

Figure 1

In-situ determination of the macropermeability of a clay formation in view of assessing leakage and mass transfer in a deep argillaceous formation

Contractor : SCK/CEN, Mol, Belgium
Contractor No : FI1W/0145-B
Contract period : July 1987 - December 1989
Period covered : January 1988 - December 1988
Project leader : A. Bonne

A. OBJECTIVE AND SCOPE

The objective of the experiment is to determine "in-situ" the permeability of a clay formation at the mesoscale, the overall permeability of an argillaceous host formation at that scale being an extremely important parameter for the long-term and short-term performances of a geological disposal concept. The experiment is of particular interest because it will be performed into the Boom clay formation. The permeability of this formation is determined on the regional scale as well as on the hand specimen scale, and it is expected that, from this macropermeability test, the relevance of possible heterogeneities and discontinuities will be detected.

B. WORK PROGRAMME

2.1. Design of the experimental set-up and development of research model
2.2. Emplacement of the experiment
2.3. Operation of the experiment
2.4. Model validation
2.5. Final interpretation and restoration of the experimental location

C. PROGRESS OF WORK STATE AND OBTAINED RESULTS

State of advancement

Due to reallocation of research resources within the broader activities of SCK/CEN, the research power to be all located for this particular experiment was temporarily suspended. A reexamination of the proposed dimensions of the experiment and an assessment of the profit to eventually combine the proposed macropermeability experiment with other tests or experiments (e.g. the planned gallery heating test) is becoming evident and are to be foreseen. For the period covered and since the initiation of this particular experiment only efforts devoted to this experiment concern the dimensioning and the development of a specific research model for it.

Progress and results

A concept that has been evaluated up to now consist of a large borehole lined with a metal tube, having a length of 10 m and is to be emplaced into the clay near to the N-end of the HADES laboratory along a horizontal direction. In order to avoid the zone where the clay might be strongly influenced by a freeze/thaw cycle of the construction procedure, the screened length should start 5 m behind the gallery lining, extending over 5 m in the clay formation. The total screened surface is about 5 m^2.

To be able to estimate an optimal emplacement of the pressure gauges surrounding the experiment, the expected distribution of the potentiometric heads in the surrounding of such a drain has been simulated by the METIS-code. In this simulation of course, the actual pressure field, that is strongly determined by the small vertical exploratory shaft, has been taken into account.

Several alternative simulations have been evaluated by varying the permeability of the clay or the dimensions of the drain.

It was calculated that, regardless of the configuration (i.e. dimensions of the drain, applied boundary conditions) or the admitted permeability, the pressure sensors should be installed at distances of at least 0.05 m, with intervals of about 0.1 m. Previous experiences however indicate that such a small spacing is quite unrealistic and very difficult to be realised because of difficulty to control deviations in the drillings.

On the other hand, permeability has a decisive influence on both the time needed to obtain a steady state regime and on the total discharge rate expected (see table below).

TABLE I. INFLUENCE OF CLAY PERMEABILITY ON DISCHARGE RATE AND DURATION OF TRANSIENT REGIME

a) K (m/s)	transient regime (days)	Q (l/day)
10^{-10}	10	19.3
10^{-11} (b)	120	4.9
$4x10^{-12}$	120	0.8

a) $S = 10^{-5}$

b) lowest permeability within 5 m around gallery lining

LABORATORY AND FIELD TESTS FOR RADIONUCLIDE MIGRATION AND HIGH FLOW PATHS IN CLAY

Contractor : UKAEA, Harwell Laboratory, UK
Contract No : FI1W/0154
Duration of Contract : 1 January 1988 - 31 March 1989
Period Covered : January 1988 - January 1989
Project Leader : Mr P J Bourke

A OBJECTIVES AND SCOPE

Two field programmes are contained within this task. Their objectives are to investigate mass transfer in clay-rich geological materials.

The principal investigation is at Culham Laboratory, where it is proposed that water flow within the Kimmeridge Clay is measured. The objective is to determine if silt-rich or carbonate-rich horizons within the Kimmeridge Clay act to provide fast transport paths for water flow through this unit.

A subsidiary investigation is to be undertaken at SCK/CEN Mol, Belgium, where an in-situ measurement of solute transport by diffusion will be made.

B WORK PROGRAMME

Culham

B1 Geological characterisation of Kimmeridge Clay unit, using cores recovered from boreholes.
B2 Development of suitable techniques for measuring water flow in the Kimmeridge Clay.
B3 Measurement of hydraulic conductivity at different horizons in the Kimmeridge Clay, in order to investigate potential fast flow horizons.
B4 Comparison of in-situ and laboratory measurements of hydraulic conductivity.

Mol

B1 Design and manufacture of suitable apparatus to install in Port B, Underground Research Laboratory.
B2 Design and manufacture of automated, computer controlled collimated radiation detection apparatus.
B3 Installation of apparatus in Port B, Underground Research Laboratory.
B4 Laboratory-scale trial tests to provide data for comparison with in-situ test.
B5 In-situ test to measure diffusivity of Boom Clay using I-131 tracer.

C PROGRESS OF WORK AND OBTAINED RESULTS

1 Culham

Planning permission for field work at Culham was granted in June by the South Oxfordshire District Council. No progress has been made with the work. Field investigations will begin at the Culham site early in 1989.

2 Mol

The Underground Research Laboratory at Mol provides access to the Boom Clay at a depth of approximately 220m below ground level. As part of the characterisation programme undertaken at Mol, the diffusivity of the Boom Clay has been measured in numerous laboratory tests. However, no in-situ measurements of diffusivity have yet been made and it is, therefore, intended to make such a measurement in order to provide a comparison between laboratory and in-situ techniques.

The in-situ test will involve the injection of a small volume of a non-sorbed gamma-emitting tracer (I-131) into the clay. The diffusion of the ^{131}I away from the point of injection will be monitored using a collimated gamma spectrometer. The geometry of the experiment is indicated in Figure 1a. As the radionuclide diffuses away from the injection point, a small component enters the collimated beam of the spectrometer. The effective diffusion coefficient of the clay, D_{eff} ($D_{eff} = D_i/\alpha$) can be determined by monitoring the count rate as a function of time. The count rate passes through a maximum (Figure 1b). From a knowledge of the migration distance from the source, and the time at which the maximum count rate is measured, it is possible to determine D_{eff}.

During construction of the URL at Mol, the clay surrounding the gallery was frozen. Therefore, in order to reach undisturbed clay, it is necessary to gain access to the clay face at distances greater than 4m from the gallery walls.

In 1987-8 SCK/CEN have been performing a number of heater/corrosion experiments in the clay surrounding the gallery. Retrieval of the heater at the end of each experiment was required in order to monitor corrosion rates of metallic components placed in contact with the clay. The resulting excavations produced 5-6m long horizontal tunnels with diameters of approximately 0.8m. These tunnels were stabilised by the insertion of steel lining tubes designed to withstand the stresses exerted by the clay at this burial depth.

A stainless steel end plate for such a tunnel was designed and manufactured at Harwell (Figure 2). The end plate incorporates a hole through which the injection of I-131 can be made, and a cross-like region in which the 4cm thick steel has been reduced to a thickness of 1-2mm. This cross produces a region where attenuation of radiation is minimal and through which measurement of ^{131}I migration can be made using a gamma ray spectrometer mounted on a two-axis stage.

In August 1988 a heater/corrosion experiment was removed from Port B in the URL. Under supervision of staff from Harwell, the end plate was successfully positioned against the clay face and steel lining tubes were installed to prevent the tunnel from closing. The installation of all components required for the in-situ migration experiment is now complete.

To maximise the number of data points which will be obtained from this experiment over a 30 hour duration, both the movement of the collimator on the two-axis stage and the operation of the gamma ray spectrometer are controlled using a fully automated computerised system. Two laboratory experiments have been performed to test the apparatus and to measure migration of I-131 in a block of Boom Clay taken from the URL.

In the first experiment, 150MBq of I-131 (as NaI in 5µ litres of solution) was injected into the clay at a depth of 11cm. However, due to attenuation of the photopeak and large Compton scatter, it was not possible to monitor migration from the point of injection.

In a subsequent experiment, 150MBq of I-131 was injected at a shallower depth in the clay block - approximately 3cm. Figure 3 shows the diffusion of I-131 away from the point of injection as a function of time. The injection point on the traverse is at the position -4mm.

The variation of count rate with time at position X = -8mm is shown in Figure 4. An effective diffusion coefficient of 3.5×10^{-10} m^2 s^{-1} is calculated. Assuming a porosity of 0.2, this value for D_{eff} is equivalent to an intrinsic diffusion coefficient of 7×10^{-11} m^2 s^{-1}. The effective diffusion coefficients calculated from this experiment are in good agreement with laboratory data published by SCK/CEN, Mol, where values of between 1.4×10^{-10} m^2 s^{-1} - 4×10^{-10} m^2 s^{-1} are quoted.

Fig. 1a

Collimated radiation detection experiment

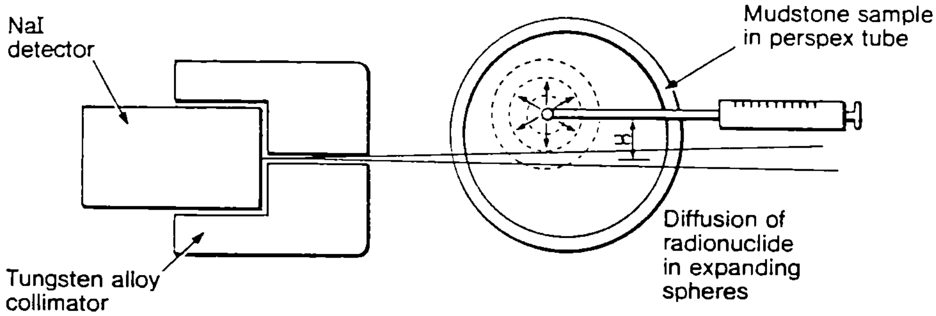
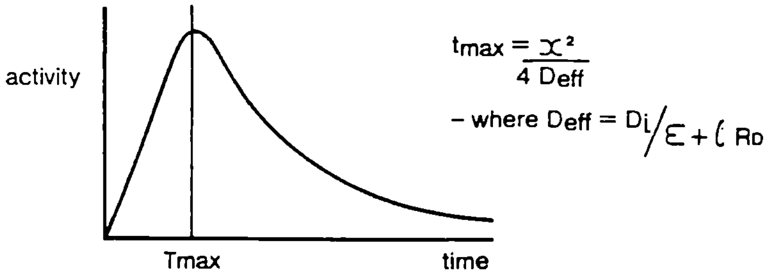


Fig. 1b

A small volume of highly concentrated gamma-active radionuclide solution is injected into the clay. As the radionuclide diffuses away from the injection point, a small component enters the collimated beam. A plot of activity detected by the gamma-ray spectrometer versus time yields a value for D_{eff}.



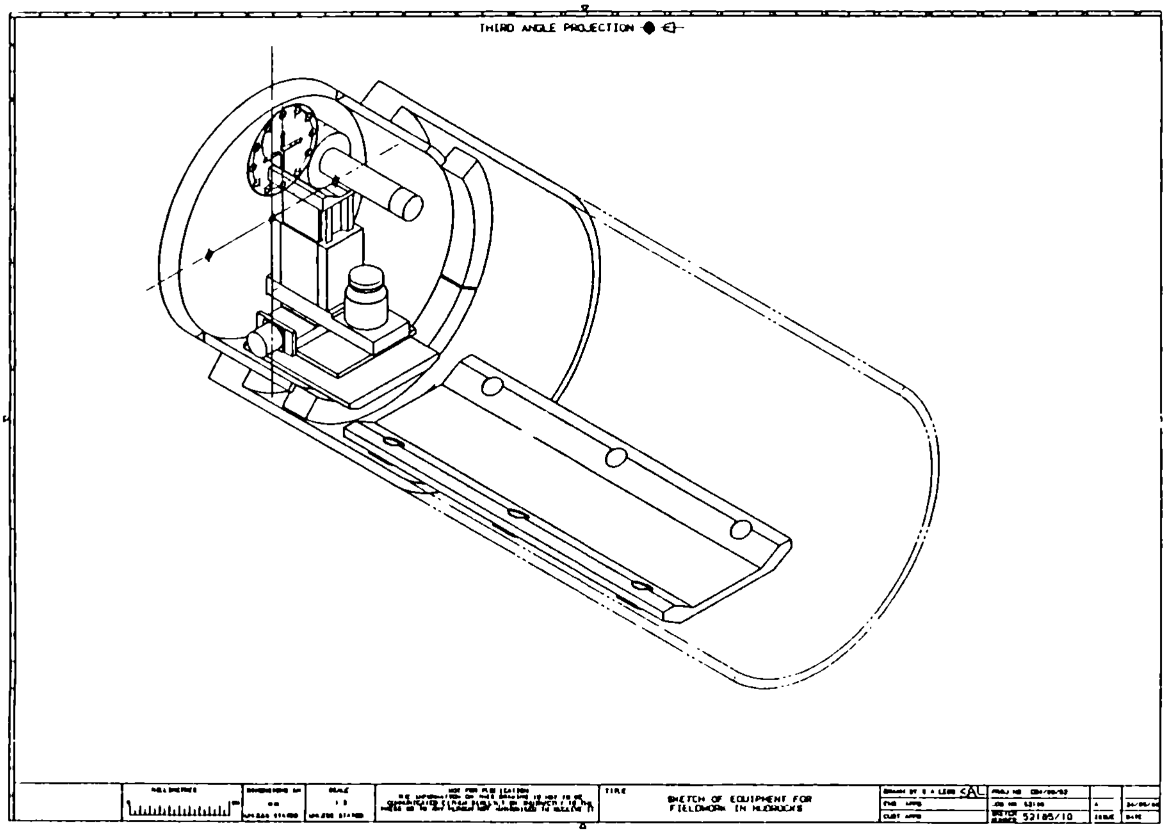


Figure 2: End plate, designed and manufactured at Harwell. This end plate is now installed in Port B, URL, Mol. The gamma-ray spectrometer mounted on the two-axis stage is shown, together with trolley for transporting apparatus along the tunnel.

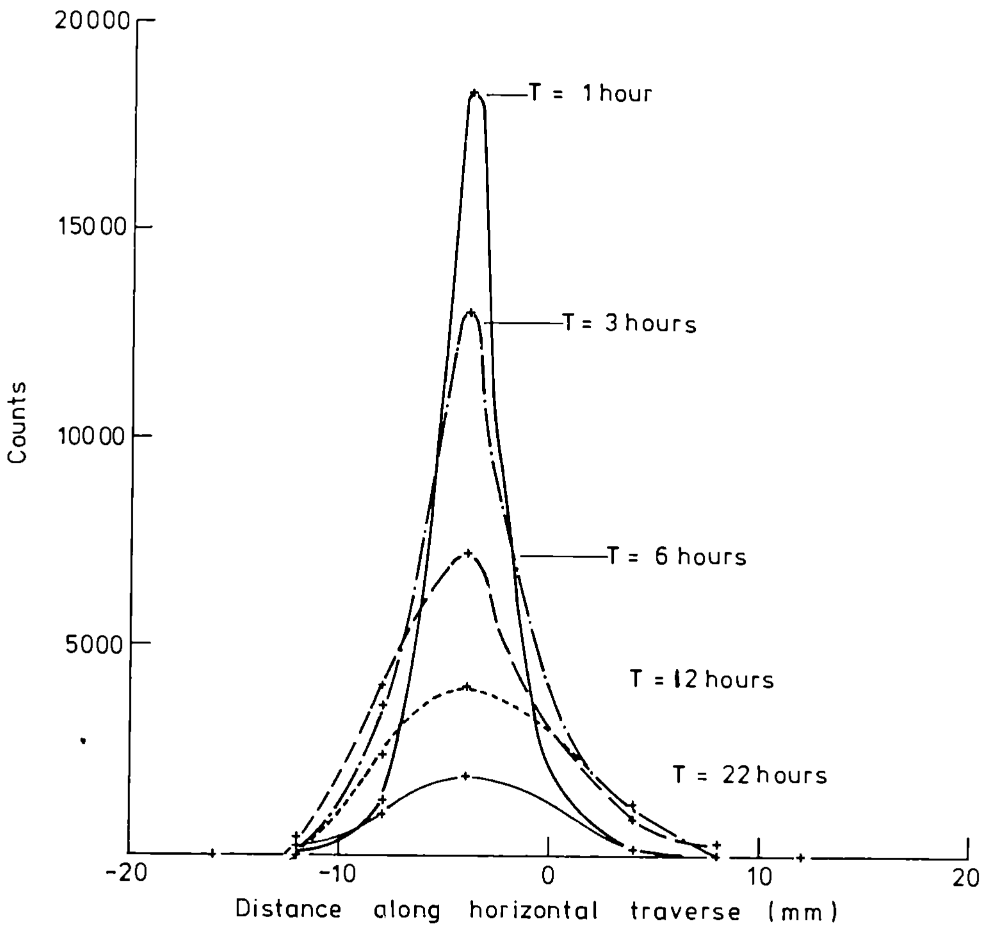


FIGURE 3. MIGRATION OF I-131 IN A BLOCK SAMPLE OF BOOM CLAY

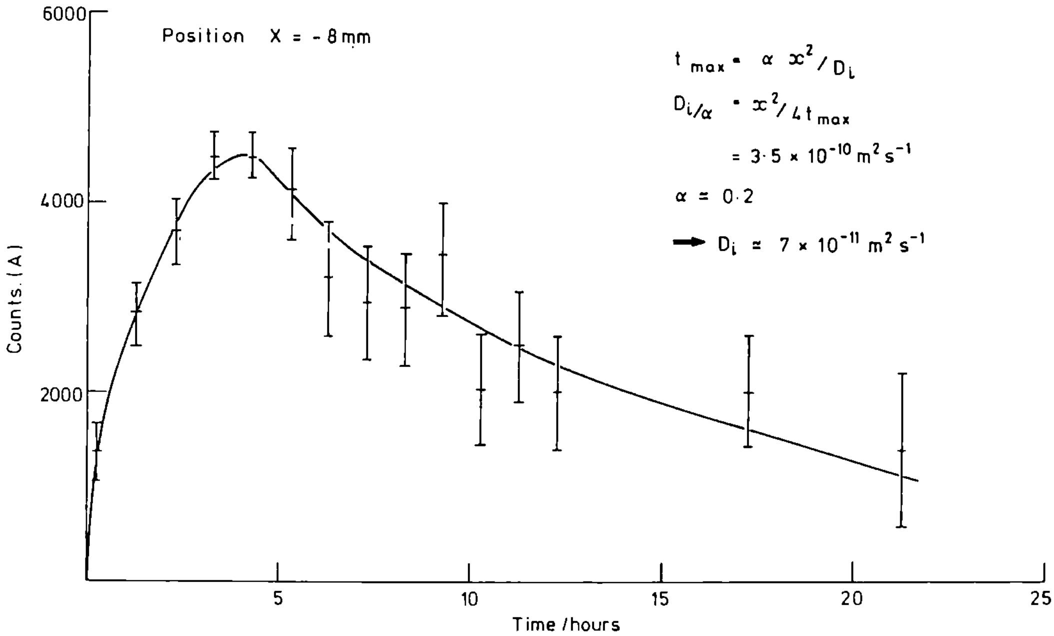


FIGURE 4. COUNTRATE AT A DISTANCE 4·5 mm FROM INJECTION POINT AS A FUNCTION OF TIME

4.3.C. Natural analogues

FIELD INVESTIGATION WITH REGARD TO THE IMPERMEABILITY OF CLAY FORMATIONS

Contractor : ENEA, CRE Casaccia, Rome (Italy)
Contract No : FI1W/00 63-I
Duration of contract : January 1987 - December 1989
Period covered : January 1988 - December 1988
Project leader : C. Polizzano

A. OBJECTIVES AND SCOPE

The main aim of this work is to assess surficial methods for detecting the secondary permeability in clay and shale in sedimentary basins by means of direct in situ observations and detection of some noble gases in soil-gas as tracers of fractures, faults etc.

The chosen gases are helium 4 and radon-222. Both these gases are used from several years for locating ore, oil and geothermal fluids as well as earthquake precursors.

In this report only helium will be discussed. The helium in the atmosphere is a mixture of helium of different origin : radiogenic helium mainly from the metamorphic basement, primordial helium from the mantle. The rate at which helium escapes from the crust is lower than the rate of its production so that the crust itself may represent an accumulation zone for this element. Helium reaches the surface using fractures and faults as preferential routes for escaping. The choice of this element for detecting fractures relay on :
- it is a chemically fully inert element;
- its atomic radius is very small, comparable with that one of hydrogen.

These characteristics make helium one of the most mobile element and therefore it can behave as an excellent tracer of geological discontinuities as well as of the primary permeability of the rocks. This report presents the results of the survey done at Vasto and Val di Paglia, as a continuation of preceeding researches carried out in Val d'Era.

B. WORK PROGRAMME

1 - preliminary survey of helium in soil-gas at a regional scale in sedimentary basins characterized by distensive tectonics and, for comparison, by compressive ones;
2 - in situ observations and information from operators about the hydrological meaning of fractures in clay.

PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

In the first annual report the scientific principles of the research were reported together with the results of an helium survey conducted in Val d'Era (Tuscany).

The results obtained in Val d'Era (annual report 1987) showed interesting relationships between helium leakage and structure features of the local graben. In fact, helium positive anomalies (helium values in soil-gas above the atmospheric content) were found in areas were geological evidences of a recent tectonic activity exist. No significant relationship was found between helium leakage and the lithology of the outcropping formations (mainly flysch with a thickness of several hundreds meters).

PROGRESS AND RESULTS

1 - For a better understanding of the results obtained in Val d'Era, other helium soil-gas surveys were carried out in 1988 in Valle del Paglia, a graben South-East of Val d'Era, and in Vasto basin. The latter is a sedimentary basin, on the Adriatic coast, in an area characterized by compressive tectonic stress.

During the two surveys about 1600 soil-gas samples were collected and analyzed in the surveyed areas. Other samples (800) now in course of elaboration were collected in the southern part of Valle del Paglia. The surveys covered a total area of more than 1500 square kilometers, with a sampling density ranging from 1 to 2 point per square kilometer.

In figures 1 and 2 some statistic data are reported. Figures 3 and 4 show the frequency distribution of the obtained results.

As it is possible to see the results show a significant difference with those observed in Val d'Era. In fact, the helium positive anomalies are much higher than those found in Val d'Era. Especially in Vasto basin most of the samples show an helium enrichment in soil-gas greater than 10% of the reference standard.

According to the Val d'Era results, in both areas the helium leakage does not seem related with the lithology of the outcropping formations (clay and sand of Pliocene age) nor with their thickness. The highest helium positive anomalies in Vasto were found in a zone where the pliocenic clay formation reaches the maximum thickness (1400 m).

Finally the figures 5 and 6 show respectively the maps of the helium distribution in soil-gas in Vasto basin and in Valle del Paglia.

As it is possible to observe in both areas the major anomalies are aligned according to NS and EW directions. The mentioned directions correspond to two of the major faults systems in Italy. In Vasto the youngest faults are indeed aligned NS and EW. The data, so far collected, strongly support the hypotheses of a possible use of the helium as tracer of occurrence and relative age of tectonic features even in clayey formations.

In the 1989 program, detailed surveys for helium and radon in soil-gas will be carried out. These surveys will be conducted according profiles crossing the areas of helium positive anomalies with a linear sampling density of 5-10 sample per km.

2 - Aside from helium distribution, direct in situ observations have been pursued for a better understanding of the hydrological meaning of fractures and faults in clay.

The research carried out, following in the steps of previous studies was aimed at widening the panorama concerning the behaviour of clay formations interested by discontinuities (either tectonic or sedimentary). The new information appears to confirm data obtained in the past years; in fact, excluding particular situations depending on local factors, tectonic dislocations do not represent preferential pathways to groundwater migration at depth. On the contrary, the effects of sedimentary discontinuities on groundwater flow depend on a number of factors and on local conditions (which can vary inside the formmation itself) so that they are most difficult to define.

A second part of the study was represented by a nation-wide search of sites suitable for the realization of experimental programmes and in situ measurements; the search has been aimed at founding a situation in which a true fault was recognized with certainty in a clay formation. The site search has been aimed at three kinds of clay exposures : valleys, quarries, and tunnels. It seems, however, that the best sites to perform field tests are found in quarries.

X2: Δ ello-4

Mean:	Std. Dev:	Std. Error:	Variance:	Coef. Var.:	Count:
800.133	1114.148	38.813	1241326.228	130.245	824
Minimum:	**Maximum:**	**Range:**	**Sum:**	**Sum Squared:**	**# Missing:**
-1300	11030	12330	659310	1549147600	0

Fig 1 - The table shows some results of the helium survey carried out in Vasto basin. In this figure, as well as in the following ones, the helium contents in soil-gas are expressed as the difference between the content (in ppb v/v) in the sample and the atmospheric level (5240 ppb) taken as the reference standard.

X1: ello-4

Mean:	Std. Dev:	Std. Error:	Variance:	Coef. Var.:	Count:
432.401	393.602	13.033	154922.328	91.027	912
Minimum:	**Maximum:**	**Range:**	**Sum:**	**Sum Squared:**	**# Missing:**
-460	3600	4060	394360	311651700	0

Fig 2 - Table shows some results of the survey conducted in Valle del Paglia.

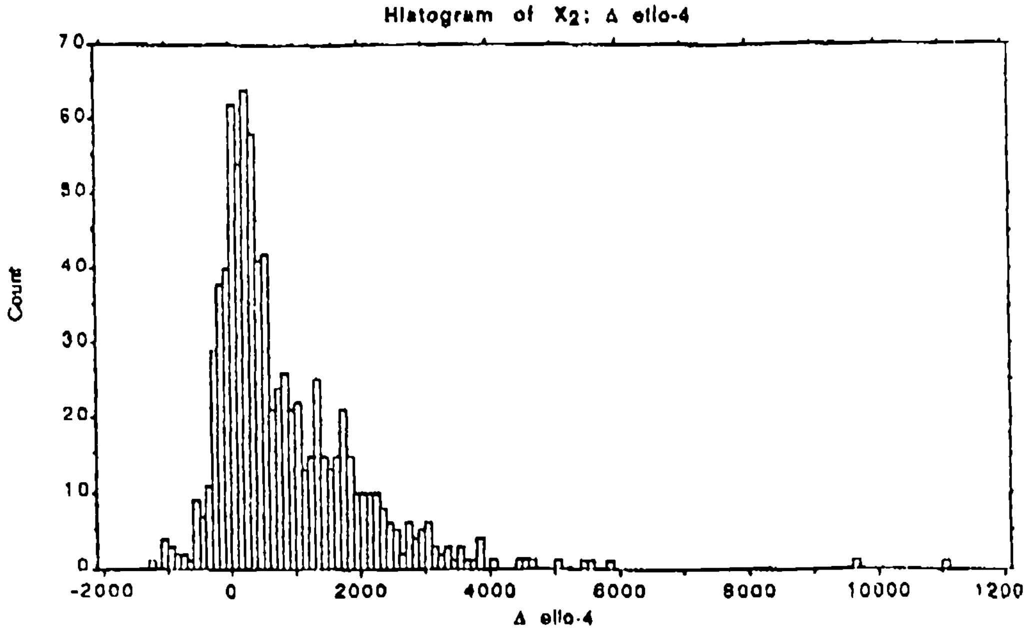


Fig 3 - Frequency distribution of helium contents (ppb v/v) in soil-gas in Vasto basin.

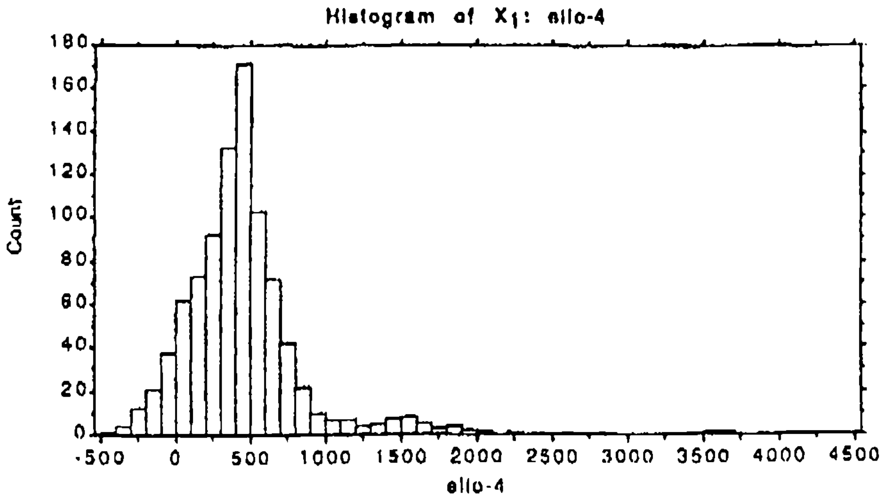


Fig 4 - Frequency distribution of helium contents (ppb v/v) in soil-gas in Valle del Paglia.

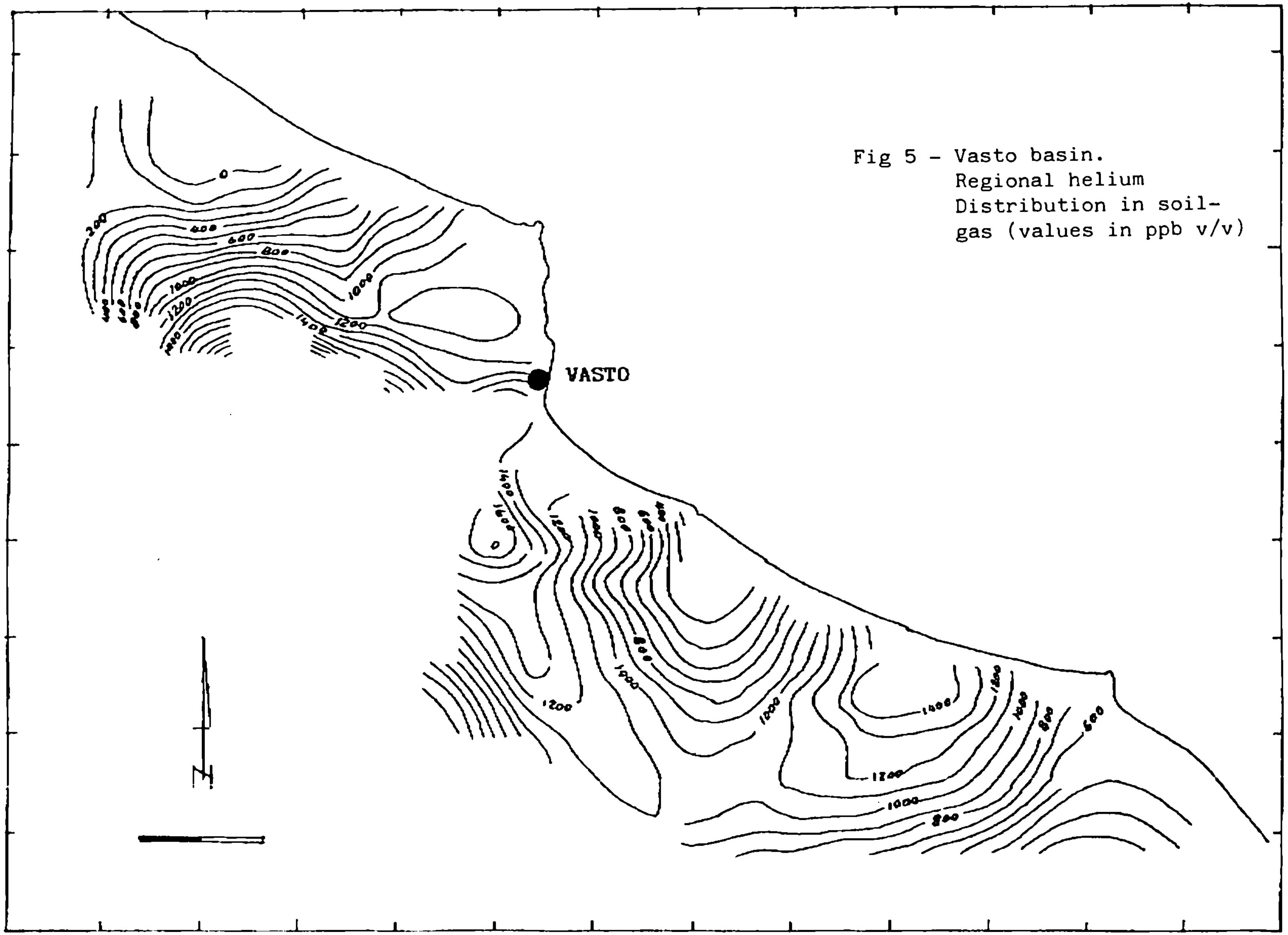


Fig 5 – Vasto basin.
Regional helium
Distribution in soil-
gas (values in ppb v/v)

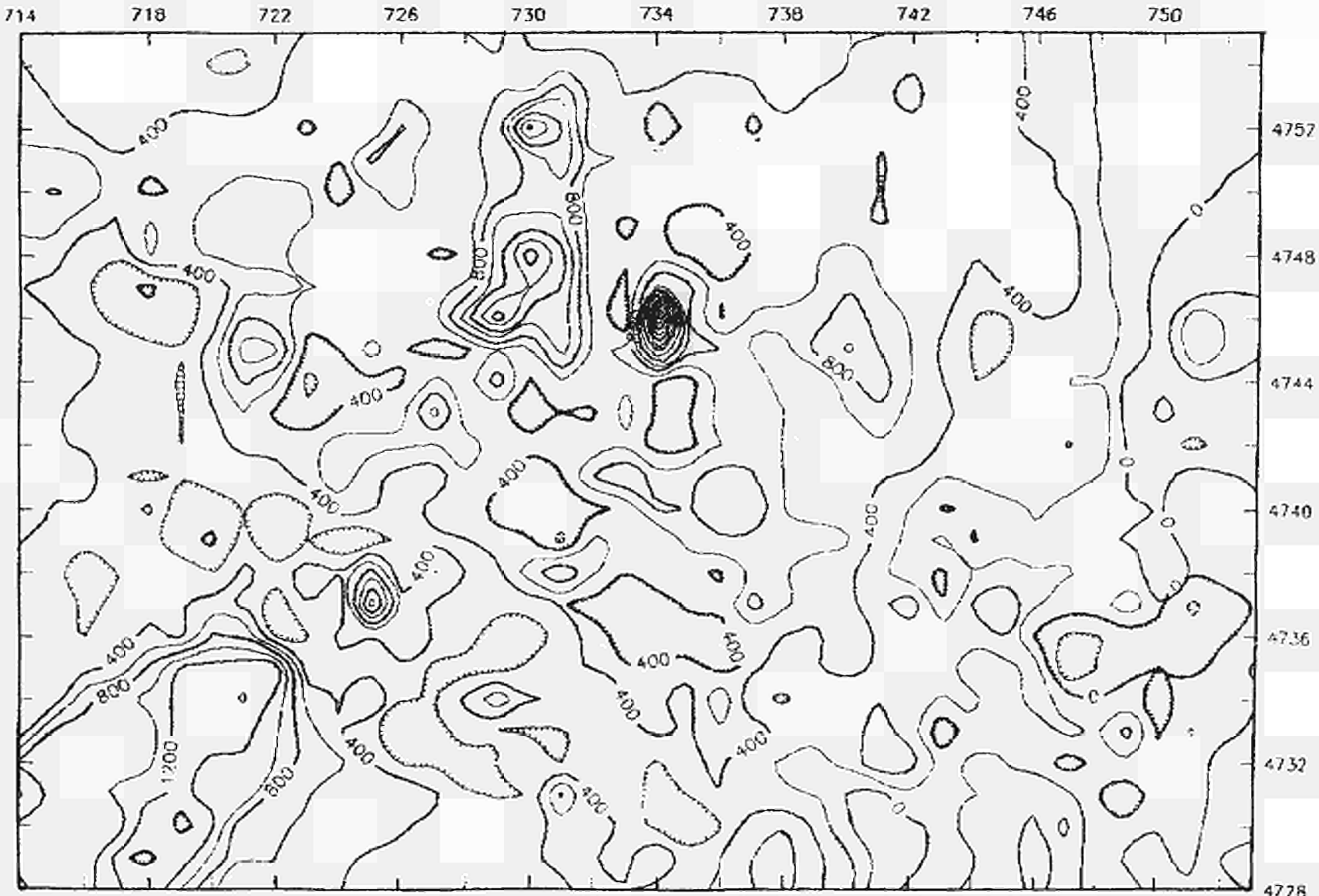


Fig. 6 : Valle del Paglia - Helium distribution in soil gas (values in ppb, v/v)

MODELLING OF RADIONUCLIDE MIGRATION IN THE GEOSPHERE : NATURAL ANALOGUE STUDIES

Contractor : Centre d'Etude Nucléaires de Fontenay-aux-Roses, CEA/IPSN/DAS/SAED - FRANCE

Contract n° : FI1W/0070

Duration of contract : January 1987 - June 1989

Period covered : January 1988 - December 1988

Project leader : P. Escalier des Orres

A. OBJECTIVES AND SCOPE

The study of the migration processes of elements in the geosphere through natural analogues is one of the best way to validate the calculation tools developped to predict radionuclides transfers on long time periods.

It has for objective to quantify these phenomena with slow kinetics, to show the thoroughness of the mechanisms taken into account in the models, or to permit their modification, if necessary.

The present study is based on the interpretation of the data obtained by the British Geological Survey on different "natural analogues" sites in the U.K.

B. WORK PROGRAMME

1. Modelisation of the elements transfers in the argillaceous sediments from Loch Lomond.

2. Pre-modelisation of natural analogues sites on the first sets of data gathered by the BGS.

3. Interpretation through models of the complete data sets : transfer in cristalline rocks (matrix diffusion), transfer in clay quaternary sediments, for example.

C - PROGRESS OF WORK AND OBTAINED RESULTS

1 - State of advancement

During the year 1988, substantial progress has been made in the understanding of the geochemical processes and the transport processes of radionuclides on the Needle's Eye site.

Our British partners have extended the network of piezometric observation and sampling thus making available a large number of water samples from various depths. In the light of the chemical analyses of these samples taken in August and November 1987 and in February 1988, we have been able to work on their interpretation and draw conclusions concerning both the geochemical processes and the hydrogeological behaviour of the site.

Using the results of these studies, which will be explored in more detail as we obtain new data from further analyses, and with the help of the incomplete but valuable information on hydrogeological parameters (permeabilities, porosities) collected in the field by the B.G.S. team, we can now plan a serious modelling of the phenomena involved in the movements of fluid in the ground. This in turn will clear the way for the coupled geochemical-transfer modelling.

The tools used for the computations are the following codes : CHIMERE (geochemical speciation), METIS (transport in porous media) and STELE (coupled model), all developped at the Centre d'Informatique Géologique of the Paris School of Mines.

2 - Progress and results

The study concerns 40 water samples (23 taken in November, 17 in February) on which complete analyses were made for major ions and certain non-ferrous metals and uranium content. Each one of them was introduced into the CHIMERE model which is able to calculate not only the respective quantities of the dissolved aqueous species but also the saturation indices of the minerals. This makes it possible to draw conclusions about the active geochemical equilibria (those involving the minerals that precipitate or dissolve) which concern the major ions as well as the uranium. The locations were the samples were taken can be seen in figure 1.

With the help of this simple, static analysis one can distinguish on the site two distinct geochemical sectors, roughly separated by the line of the E-W fault (ordinate line 14 on figure 1). This differentiation is particulary evident in the saturation indices of calcite which is clearly deficient in the hornfels zone to the North and balanced in the sedimentary cover of the limestones to the South (figure 2). The contact at the lower end with carboniferous limestones even contribute to the appearance of oversaturation with a clear tendency to a state of equilibrium from the bottom toward the top of the piezometers in question (figure 3). The equilibrating mineral phase of the silica appears to be a pole close to chalcedony (figure 4). Finally, an increasing mineralization is evident toward the South particularly at depth, which clearly indicates a contamination by seawater.

We have also studied the behaviour of uranium at the site. This behaviour is, a priori, a complex one, bearing in mind on the one hand, the lack of knowledge of the source term and on the other, the complexity of the lithology of the recent alluvial deposits, the organic content of which is extremely variable. We have been able to contribute some elements toward a solution to the problems in the area.

First of all, there is no obvious direct link between the organic content of the waters and their uranium content. However, the conditions of oxydo-reduction and pH imposed locally by the decomposition of the plant material are qualitatively correlated to the degree of water contamination by uranium.

The source term of the uranium can, in all likehood, be found in the mineralizations of the cliff, since the sedimentary series are, above all, contamined by the runoff from the cliff and by upwelling at the foot of the sedimentary series in the extreme North, which is a justification for the artesian behaviour in this part of the site. This flow from the bottom upwards is confirmed by observing the saturation indices of a mineral created by the alteration of uraninite, i.e. liebigite (figure 5). This does not have any bearing on the presence or absence of this mineral on the site but it does indicate that the equilibrium of the waters is indeed imposed by a mineral close to uraninite.

The bottom of the recent sedimentary series, systematically rich in uranium, justifies the assumption of a level with a rapid N-S circulation ("beach level").

The role of the major E-W fault which separates the limestone and the hornfels is more difficult to elucidate. It can, however, be said that it certainly plays a diluting role which suggests that there is some important flow of water in this area. This flow is confirmed by the presence, downstream from the fracture, of hight concentrations of non-ferrous metals (Pb, Zn, Ba etc.) which can only originate in the fault. Moreover, since these samples have a low uranium content, it can be concluded that the contribution made by the fault to the uranium production in the sediments is small, either because it is not hydraulically connected to a source term or because the uranium precipitates at the outlet of the fault under influence of a contrast of physico-chemical conditions.

3 - Conclusions

The major advances made in this work during 1988 concern the understanding of the hydrogeological behaviour of the Neddle's Eye site. In particular, we have been able to confirm :

- the position of the major E-W fault and its hydraulic importance,
- the existence of a lower lithological stratum with rapid N-S circulation,
- the existence of a zone with vertical upward flow in the northern part of the site.

The interesting feature is that these results have been made available only with the help of geochemical investigations which proves how powerful a tool this is.

Concerning the behaviour of the uranium on the site, the study clearly emphasizes the role of the mineralizations of the cliff and minimizes that of the major E-W fault. Furthermore, the equilibrating pole of the uranium appears to be a mineral originating in the uraninite alteration.

Coupled modelling requires complete knowledge of the active geochemical system and this study has made it possible to partly determine its characteristics, in particular where silica and carbonates are concerned.

4 - Major publications

JAMET Ph., SOUBEYRAN R., DOUBLET R. (1988). Modélisation de la migration des radionucléides dans la géosphère. Etude d'"analogues naturels", Rapport C.C.E, LHM/RD/88/75, 34p.

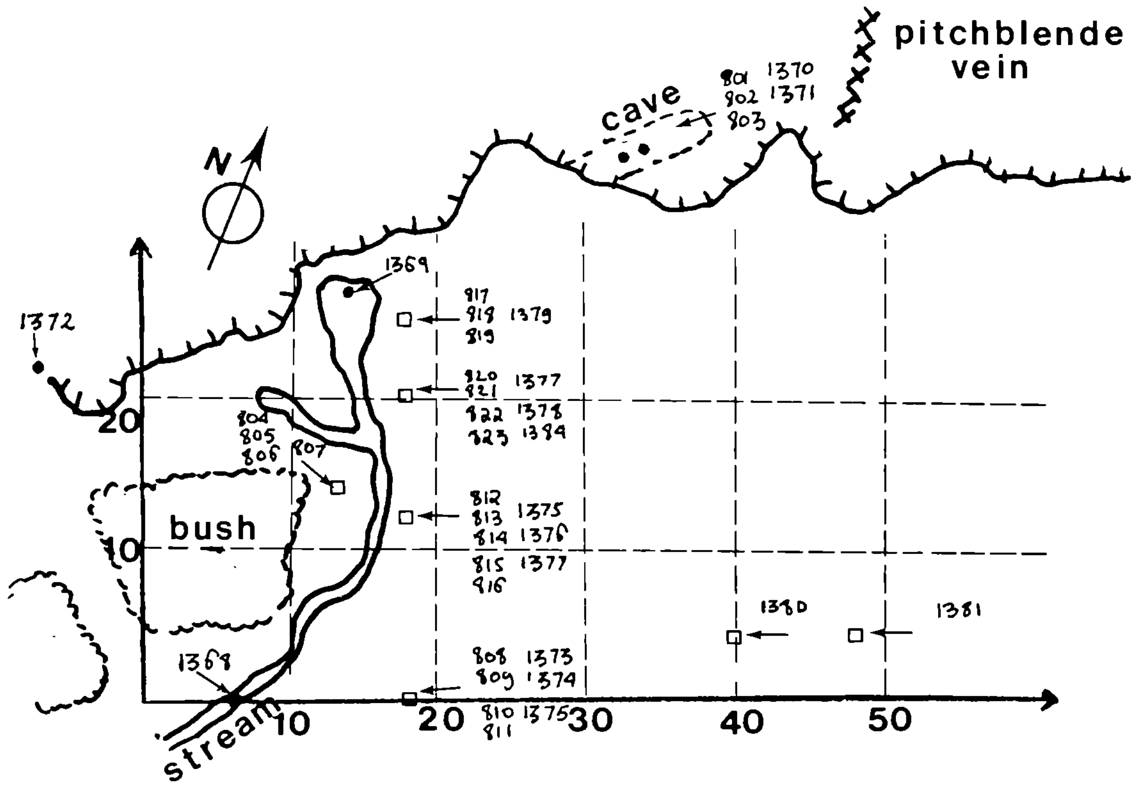


Figure 1 : Sampling network - Needles' Eye
□ = piezometers
• = occasionnal sampling

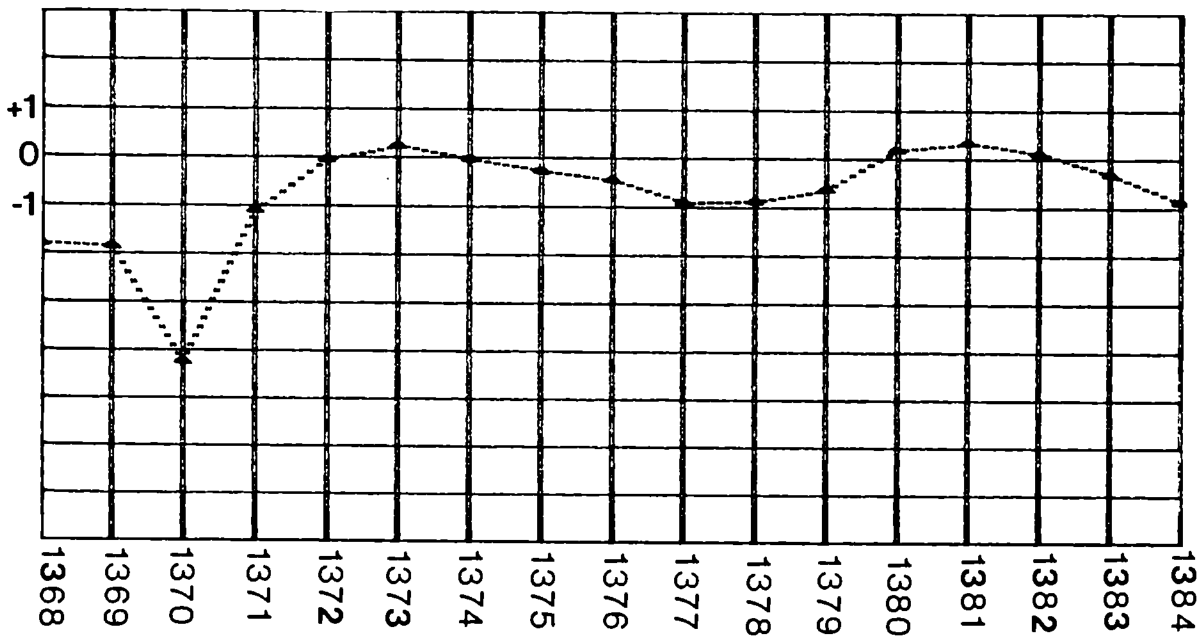


Figure 2 : Saturation indices of calcite
(february samples)

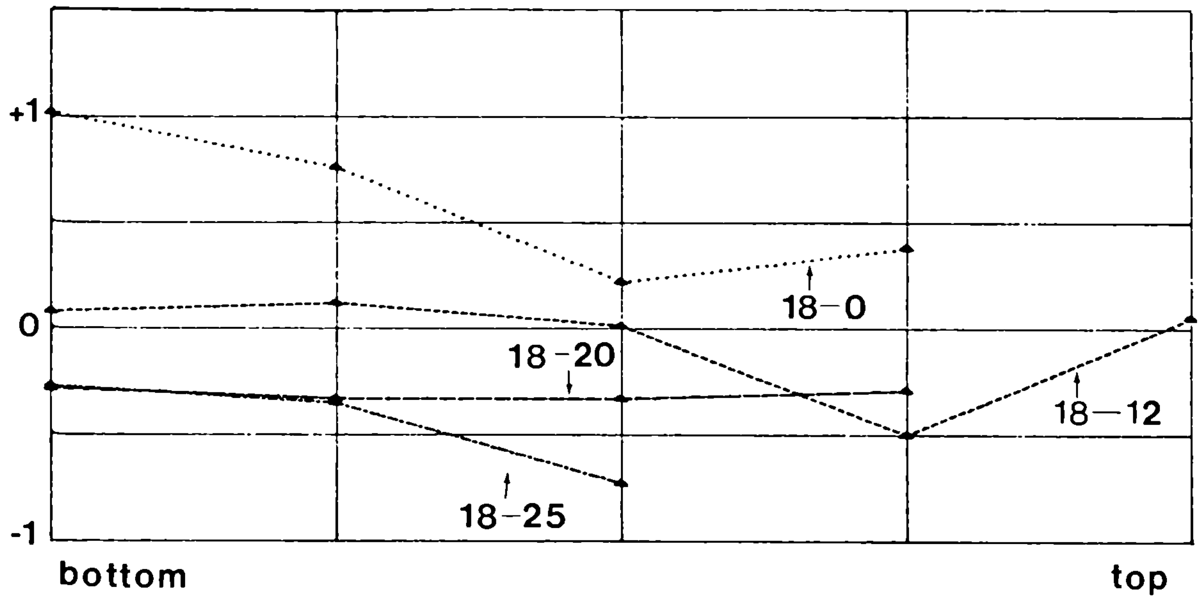


Figure 3 : 18-Y Piezometers : saturation indices of calcite (808 to 823 samples)

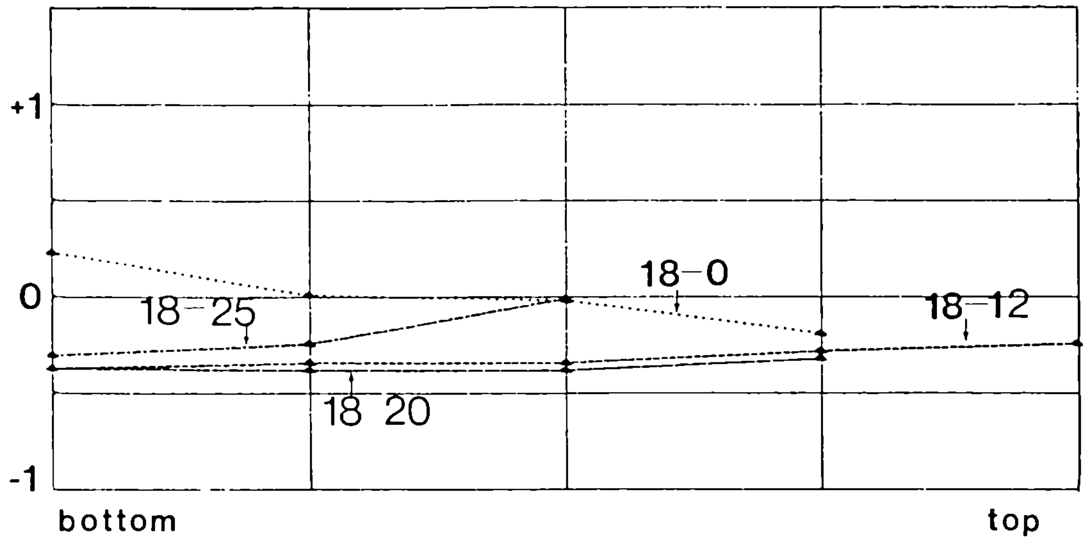


Figure 4 : 18-Y Piezometers : saturation indices of Chalcedony

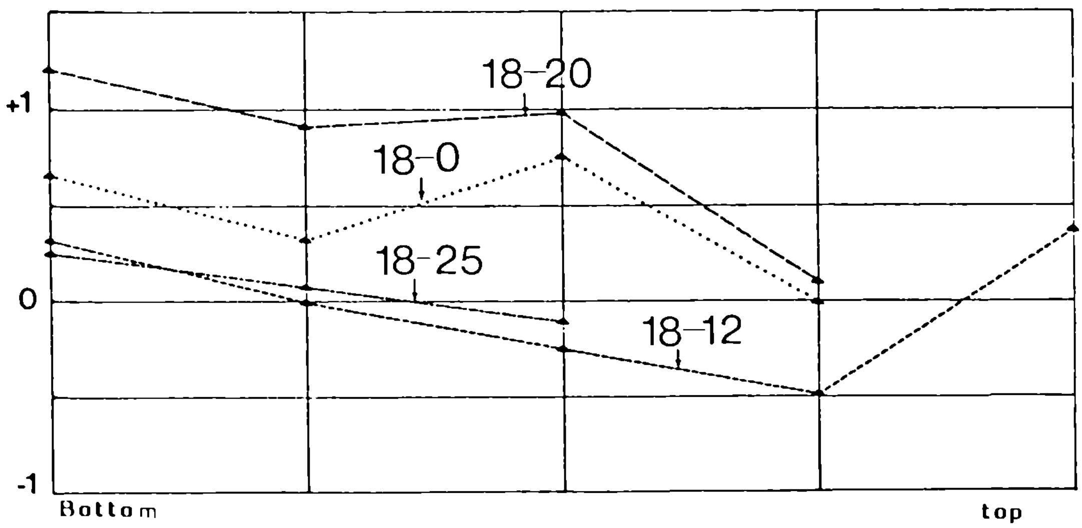


Figure 5 : 18-Y Piezometers : saturation indices of Liebigite Ca_2, UO_2, $(CO_3)_3$

STUDY OF MIGRATION PROCESSES IN CLAY FORMATIONS OCCURRING IN NATURE

Contractor : ENEA, CRE Casaccia, Rome (Italy)
Contract No : FI1W/0071
Duration of contract : January 1987 - December 1989
Period covered : January 1988 - December 1988
Project leader : A. Brondi

A. OBJECTIVES AND SCOPE

The permeability state of clay may be affected by tectonic events. Fractures and faults may indeed give rise to a secondary permeability within argillaceous rocks. Ochraceous bands aside from the fracture planes evidence circulation of meteoric water within fractures systems in clay. The penetration depth of these water in clay should reveal the thickness of clay affected by secondary permeability due to tectonics. The present work is a contribute aimed at ascertaining the real influence of tectonics on clay in particular formations in the Siena basin in central western Italy. The tectonics of the basin, a typical graben, is well defined.

Complementary researches in other situations, i.e., in Pasquasia mine, where clay are highly fractured, offer the opportunity of investigating a clay system isolated from surficial water.

B. WORK PROGRAMME

Three main lines have been envisaged :
1 - Selection of the most appropriate situations referred to the general tectonic frame;
2 - Studies on the variations of the geochemical system of clay because of the penetration of the meteoric water;
3 - Investigation on the extent and causes of the secondary permeability of clay and their importance with regard to the geochemical stability of clay formation.

C. PROGRESS AND OBTAINED RESULTS

State of advancement

Among the many possibilities offered by the Italian territory the Siena basin, located in central western Italy, has been considered as highly significative for the envisaged research.

The Siena basin is made by a deep tectonic distensive trench, parallel to the Appennines chain, filled by a thick series of pliocenic clay and overlying sand. Because of the orogenic uplift still in course, the pliocenic series undergoes to accelerated erosion. Clay unloaded by the overlying masses, removed by erosional activity, expands in volume without a contemporaneous mass increase. This causes latent fractures to be opened and penetrated by surficial waters. A major effect of surficial water circulation is the oxidation of the fracture walls accompanied by a centimetric-decimetric alteration of the clay body. The clay oxidation along fractures may develop to a depth of some tens meters. These values may indicate the maximum penetration depth of surficial oxydizing waters within clay mass.

No morphological structure generally evidences the occurrence of fracture and fault plains intersecting clays. This fact practically hinders to recognize the possible relationships among the regional tectonic trend and the tectonic structures interesting clay.

PROGRESS AND RESULTS

1 - In order to face the mentioned problem the first step of investigation in the Siena basin has been directed to draw a map of the morpho-tectonic linear elements. The work has been conducted by means of photointerpretation. The obtained results are reproduced in figure 1. The trends of the linear elements conform to the general tectonic trend of this part of the Appennines chain.

2 - The observation data on clay tectonics are very scarce and ambigous. Only direct field mapping (figure 2) allows to recognize the tectonic structures. From field observations fractures in clays often result to be cemented by gypsum filling (figure 3). This fact testifies that circulation of water within fractures in clay may even represent a transitory condition possibly ending with the re-establishment of the initial situation of very low permeability. The oro-morphological evolution is undoubtedly the main controlling factor of the near surface secondary clay permeability.

3 - The circulation of fluids within clay out of the influence of surficial water is worthy of study.

Diagenesis and tectonic effects may determine the re-arrangement of clay particles as well as creation and desappearance of local micropermeability. Investigations conducted on clay samples from Pasquasia mine in Sicily give a contribution to this matter.

Analyses by normal and electron microscope give evidence of the microscale variations of physical and geochemical equilibria within clay.

Microfractures crossing clay are filled by gypsum aggregates (figure 4). Gypsum crystals grow starting from the fracture wall without crossing it. Indeed the fracture surface appears to act as a barrier to the solution diffusion, confirming the practical impermeability of the clay bulk. Pyritic aggregates and isolated crystals occurring in the mass (figure 5) testifies the reducing conditions prevailing in clay.

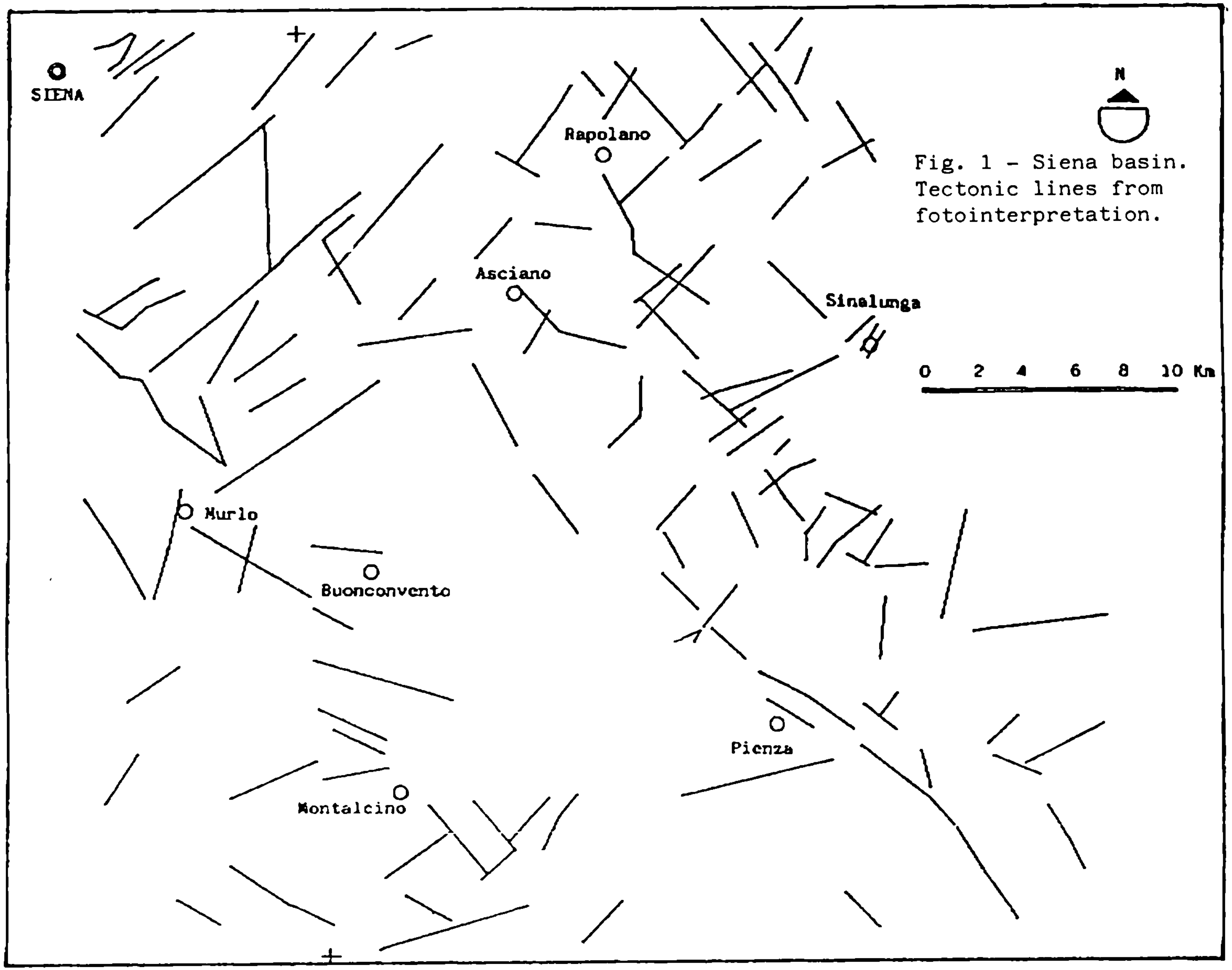


Fig. 1 - Siena basin. Tectonic lines from fotointerpretation.

Fig. 2 - Fractures in clay are clearly evident in quarry walls.

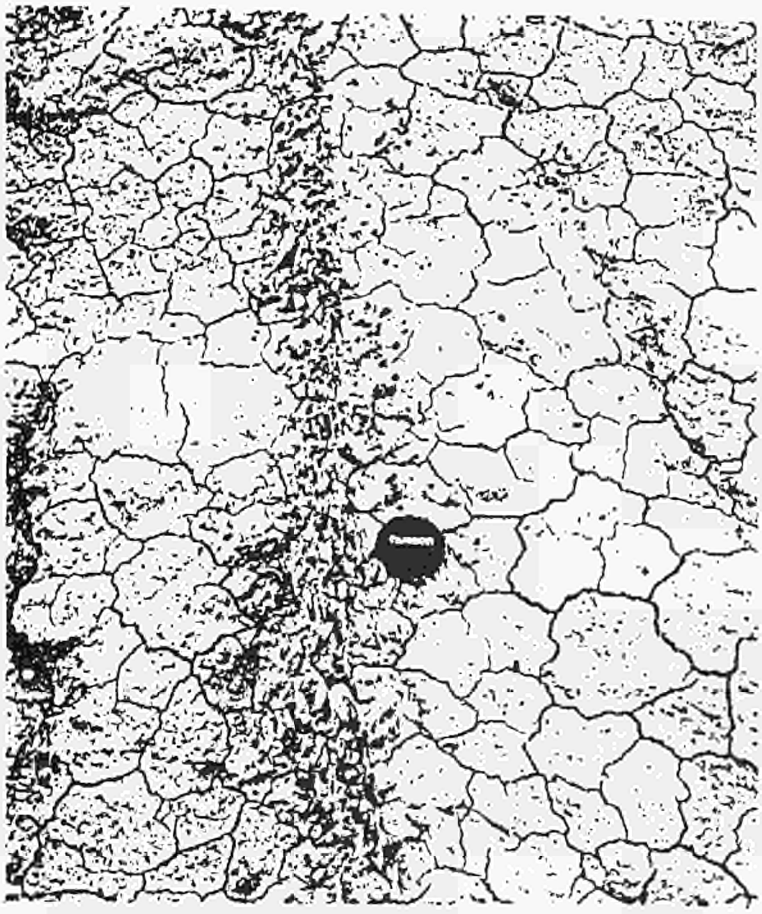


Fig. 3 - Gypsum filling fractures in clay intersects the natural ground surface.

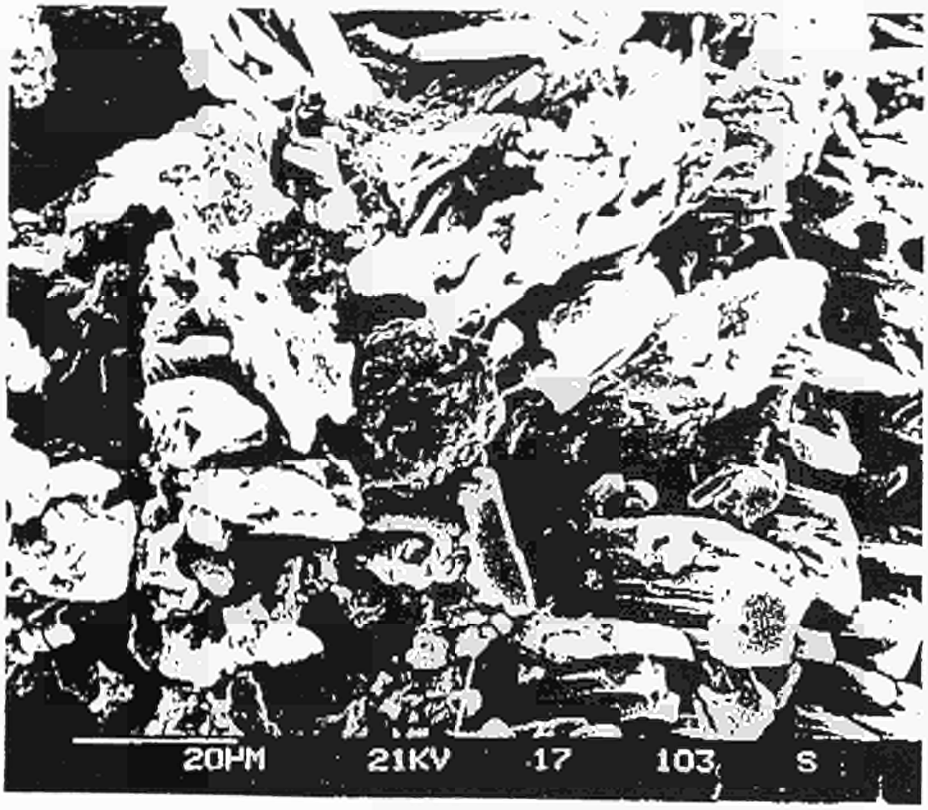


Fig. 4 - Gypsum crystals fill and cement microfractures in clay.

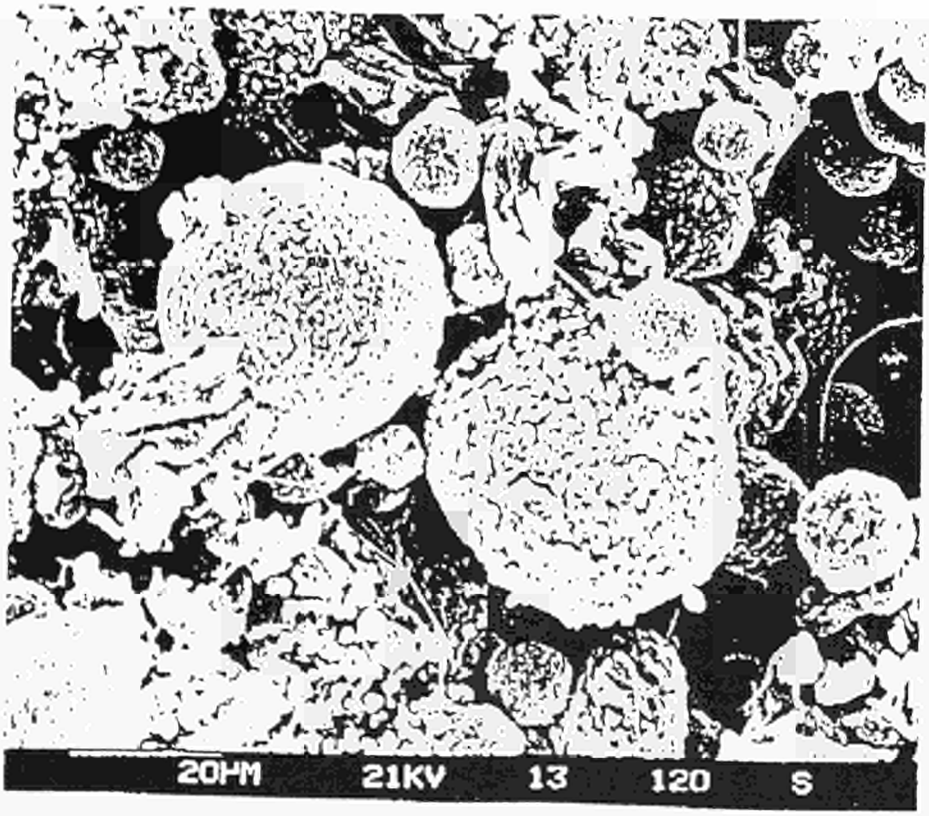


Fig. 5 - Pyritic aggregates indicates the prevailing reduction condition in clay.

Natural analogues of radionuclide migration in granitic rocks through the study of palaeo-hydrothermal alteration

Contractor : BRGM - Orléans, France
Contract n° : FI 1W/0072/F
Working period : August 1986 to August 1988
Project leaders: P. Peaudecerf et A.M. CHAPUIS (CEA)

A. OBJECTIVES AND SCOPE

Mineralized zones in granitic rocks which have been influenced by hot water (hydrothermal activity) over long geologic periods may be considered as useful natural analogues of the conditions occurring around a heat-producing repository in granite. Study of these zones gives valuable information about the migration and retention of elements analogous to the radionuclides present in the repository.

Development of research on one or more sites of former hydrothermal activity in a granite environment will enable an approach to the problems of migration and retention of elements such as the rare earths, uranium and thorium which are themselves analogous to the radionuclides in their geochemical behaviour. The time scale will be that of the geologic environment of 0.1 to 1 million years. The volumes considered will take into account the far field (pervasive alteration) and near field (vein alteration) phenomena. The final aims of this programme are :

1. To define the physicochemical conditions of the alteration examined and the mineralogical carriers of the elements analogous to the radionuclides.
2. To test on existing programmes (EQ6, PATH) or on those being (CEQCSY) developed, the data supplied by the direct approach (validation of thermodynamic parameters, assessment of the reaction kinetics on natural examples).

B. WORK PROGRAMME

B.1. Petrographic and mineralogical investigation of the various events which have affected the system.

B.2. Analysis of the analogue element carriers in two parts :

- B.2.1. Separation of the primary and secondary phases developed by the alteration and quantitative analysis of the analogue elements in the separated phase and in rock samples ;
- B.2.2. Location of analogue elements by nuclear methods. From these data the mass balance and the distribution and mobility of the analogue elements during the palaeo-alteration will be established.

B.3. An indirect approach by using mass transfer codes on major trace and analogue elements to mobilize the geochemical behaviour of the alteration systems.

C. PROGRESS OF WORK AND RESULTS OBTAINED

State of advancement

Following the signing of contract FI 1W 0072/F in October, it was decided to proceed to selection of an additional site for the study of palaeo-hydrothermal alteration enabling the continuous tow- or three-dimensional analysis of a system. This selection was made after a period of exploration during which thirty or so sites were visited in Brittany and the Massif Central.

Once the selection of the Fombillou site in the Massif Central was made, an inter-disciplinary field programme, involving mineralogists and geochemists was carried out. Work progress is as follows :

B.1. is completed.

B.2. is completed.

B.3. is partially completed. The report was delivered by the end of 1988.

PROGRESS AND RESULTS

B.1. Petrographic and mineralogical investigation of various events which have affected the system.

The main results obtained show that the hydrothermal system of Fombillou is polyphase and underwent at least three major hydrothermal events as well as supergene alteration especially on the wall rocks of the main hydrothermal system.

The first hydrothermal cycle which affected the site took place during leucogranite emplacement about 300 to 280 m.y.ago at temperature ranging from 400 to 300°C. It developed in the whole rock with a pervasive alteration of biotite generating association of chlorite-phengite-orthose. This cycle occurs with Sn, W and B anomalies in the walls of the main vein.

The second and third cycles have only affected the walls with respectively the development of phyllic alteration with occurrence of illite for the second cycle and argillic alteration with the formation of Ro-type randomly interstratified clay for the third cycle. The temperatures measured by fluid inclusion studies give for these two types of alterations temperatures about 300 to 200°C for the second cycle and about 100°C for the third. The last stage developed a sulfide mineralisation in the vein and an As, Pb and Cu anomaly in the host rock, and the second stage with a weak U anomaly.

Following the above work, computations based on Gresens' equations and on isocon diagram, were performed to evaluate changes in volume and in composition. Stoichiometric computations of mineral reactions during alteration were made. They enabled a comparison between data obtained from evaluations made on whole rock and evaluations of mineralogical reactions described in the hydrothermal system.

Study of the thermal signature of the main vein by the study of sealing fission tracks in apatite has provided a curve of relative age around the vein. This gave theoretical image of the geothermal gradient induced by hydrothermal solutions but discrepancies exist between these results and thermal modelling.

An age of 80 m.y. obtained for the vein system at temperature above 100°C and a plateau age of 240 m.y. corresponding to the last cooling phase of granite at temperature below 130°C was obtained for the most distal sample collected 15 m from the vein.

B.2. Analysis of analogue element carriers in two parts :

B.2.1. Separation of the primary and secondary phases developed by alteration and quantitative analysis of analogue elements (REE, Hf, U, Th) in the separated phases and rock samples.

The results obtained on rock samples display a REE (low and high) mobilization during hydrothermal events around the main vein. Results on separated phases (essentially phyllosilicates) show the capability of newly formed mineral to trap the REE released. The role of phosphate minerals is also considered because of P and REE positive correlation on near-vein environment and monazite presence in hydrothermalised rocks.

B.2.2. Location of analogue elements by nuclear methods:

The results obtained through mapping of uranium the study of induced fission tracks show a good correlation between the development of alteration phases and the distribution of uranium, the values of which increase in altered rock in the selvages of the vein and decrease on near-vein environment. But the support of these concentrations, mainly iron oxyhydroxides, suggests late reworking of this element in supergene environment controlled by adsorption on oxyhydroxide and pH of vadose waters. Leaching sulfur mineralization and hydrothermally altered rocks.

Study of the disequilibriums in the family of uranium 238 in the main vein environment provides coherent information on the question raised by mapping of the distribution of total uranium.

The must plausible evolutionary model for the Fombillou site is as follows :

- during the emplacement of mineralisation, several million years ago o first disequilibrium occurred in the system releasing uranium in some crystalline phases or pre-existing mineralisation. Then in the course of time, a new $^{234}U/^{239}U$ equilibrium was achieved in alteration phases which probably had not the same characteristics as the crystalline phase of granite and no nuclear recoil generating $^{234}U/^{238}U$ disequilibrium (oxyhydroxides);
- a recent or present-day alteration phase has reworked or is reworking the "free" uranium from the host rock towards the wall rocks of the vein accounting for $^{234}U/^{230}Th$ disequilibriums.

Geochemical studies performed with total thorium and thorium isotopes display a near immobility for this element in hydrothermal and supergene environment.

B.3. Indirect approach by using mass transfer codes

A preliminary thermodynamic simulation of mineral transformations and transfers of matter during hydrothermal alteration (phyllic stage) was performed using the calculation code CEQCSY (Chemical EQuilibrium in Complex SYstem). This simulation is based on the values of the main physical and chemical parameters deducted from analysis of the natural system.

On the hasis of the results obtained from Fombillou, an appraisal was made of the response of the granitic environment which has been disturbed by a hydrothermal system produced by heat emitted by storage of high-level radio-active waste as well a its potential capacities of retention in case of possible leakage.

List of publications

/1/ Sureau, J.F., Méloux, J., Jébrak, M., Lemière, B., Griffault, L., and Cantinolle P. (1986) Devenir à long terme des stockages de déchets radioactifs en formation géologique : analogie avec l'altération des gisements minéraux. Rapport C.C.E.

/2/ Griffault, L., Jébrak, M., Lemière, B., Piantone, P., and Sureau, J.F., (1987). Hydrothermal alteration systems as analogues of nuclear waste repositories in granitic rocks. An example : The Langenberg hydrothermal system. CCE symposium proceedings volume 1.

/3/ Griffault, L. (1987). Bilan des transferts de matière (Majeurs et Terres Rares) au cours des altérations hydrothermales des granites. Exemple du granite du Ballon d'Alsace (Vosges méridionale). Thèse Université Poitiers.

/4/ Lemière, B., Griffault, L., and Jébrak, M. (1987). Lithogeochimie and hydrothermal alteration of granite : principle and applications in the Vosges area, France. Revista Brasilera de Geosciências, 17 (4), pp. 614-616.

NATURAL ANALOGUE STUDIES OF RADIONUCLIDE MIGRATION

Contractor: British Geological Survey/NERC, Keyworth, Nottingham, UK
Contract No: FI1W/0073
Duration of contract: October 1986 to September 1989
Period covered: 1988
Project leader: PJ Hooker

A. OBJECTIVES AND SCOPE

It is important to be able to validate and support models of long-term predictions of radionuclide migration in the geosphere. The main aim of this research is to examine natural geochemical discontinuities and gradients as analogues of radionuclide transport in sediments. The mechanisms of processes of mobilisation, advection, diffusion and retardation for natural decay series elements and iodine and bromine will be addressed. This will entail some development of the techniques for measuring small concentrations and the speciations of these elements in both the solid and pore water phases. Analytical determinations by alpha spectrometry and neutron activation analysis will be carried out by SURRC, East Kilbride, and UKAEA Harwell under sub-contracts. Support in modelling will come from co-operation with the Ecole des Mines de Paris, Fontainebleau, and from a sub-contract with WS Atkins Engineering Sciences.

B. WORK PROGRAMME

B.1 Phase 1986-1987.

B.1.1 Site investigations.

B.1.1.1 Collection of fresh Loch Lomond sediments; analysis of I and Br depth profiles; preliminary modelling for effective diffusion coefficients.

B.1.1.2 Pilot investigation of I, Br, U and Th gradients across marl/clay boundaries in a well characterised sediment core from Lundin Castle, Fife, eastern Scotland.

B.1.2 A desk study of surface diffusion as a solute transport process for major cations through clays, with implications for trace radionuclide migration.

B.2 Phase 1988-1989.

B.2.1 Site investigations will be concentrated on measuring the speciation and mechanisms of distribution of I, Br, U and Th in sediments from Loch Lomond, Needle's Eye, near Dalbeattie in SW Scotland, and from Broubster in Caithness, N Scotland. (The investigation of Lundin Castle sediments was dropped in favour of the more fruitful work on the Needle's Eye and Broubster analogue sites.)

B.2.2 Modelling of the results and data from the field investigations for migration through sediments. (The application of the surface diffusion desk study has been dropped in favour of modelling the Needle's Eye and Broubster sites.)

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The surface diffusion desk study has been completed with the submission of a draft report [1]. The technical annex was changed to investigate the Needle's Eye analogue site near Dalbeattie, SW Scotland and the Broubster site in Caithness, N Scotland rather than pursue the Lundin Castle sediment study or extend the surface diffusion aspect. SURRC investigations of the Loch Lomond sediments have continued with the finalisation of a low volume, low blank method of determining halogen concentrations and chemical forms in interstitial water samples from the loch sediments. New cores were collected in September and are being treated and analysed; results for interpretation and modelling will be available in 1989. Work describing the geochemistries and hydrogeologies of the Needle's Eye and Broubster sites has continued with a view to understanding better the controls on the movements of U and Th in the systems.

Sediment samples from Needle's Eye are continuing to be examined by XRF, alpha and gamma spectrometry, autoradiography, fission track registration and SEM methods to define U distributions. At the same time, the Ecole des Mines de Paris, Fontainebleau (EMP) has carried out a hydrogeochemical modelling exercise with PHREEQE and CHIMERE and the CHEMVAL thermodynamic database to interpret the groundwater composition results from BGS. The importance of carbonato-U complexes and carbonate phases is highlighted in the EMP report. A Needle's Eye workshop held in October at BGS resulted in the aim of applying the STELE transport code to the modelling of U migration in the sediments.

The chemistries of the Broubster groundwaters have been modelled by WS Atkins to demonstrate the competitive complexation of U with humate and carbonate ligands as a function of pH; there is good agreement with experimental data. The potential usefulness of the CHEMTARD chemical transport code was demonstrated for the site.

PROGRESS AND RESULTS

B.2.1 Field investigations

Loch Lomond

The Loch Lomond sediment study is concerned with the pore water concentration and speciation profiles of I and Br. A method based on HPLC separation followed by neutron activation analysis has been developed at SURRC for measuring 0.1ml pore water samples. Collection of new cores took place on 6th September. Cores were extracted by a specialist team from the FBA Windermere Laboratory using a 6m Mackereth corer. The collection of a full 6m length core proved impossible due to the presence of a gravel band in the sequence; two cores of length approximately 5m were recovered. These cores were sectioned under oxygen-free conditions using glove bags and were returned to SURRC for analysis, with all subsequent operations being carried out under nitrogen. Halogen speciation determinations are currently being undertaken on extracted pore water samples.

Needle's Eye. One field trip was arranged to the site during December. This was to collect further water samples for chemical analysis and to collect sediment samples along a line from the cliff to the creek. All of these samples are currently being processed for elemental concentrations. Work has continued at SURRC on the analysis of natural decay series radionuclides in solid phase samples from four pits dug in 1987, and in water samples collected from the cliff and from auger holes made in the sediment deposits. The results so far available for one of these pits (NE Pit 2) lying directly above a pitchblende vein at the top of the cliff show a decreasing trend in U concentration with increasing distance from the vein. However, it is not possible to define the mechanism giving rise to this distribution until thorium measurements on these samples are completed.

The natural decay series analyses of water samples and pit section samples in conjunction with BGS hydrogeological and gamma spectrometry results [2], can be used to formulate a geochemical model for uranium migration in the cliff and sedimentary deposits over the past 2000 years or so, Figure 1. The essential features are:

a) There is uranium dissolution from the cliff area (in particular the pitchblende veins) and then transport of U(VI) in solution in oxygen-saturated surface run-off water. Characteristically, surface drainage samples show $^{234}U/^{238}U$ activity ratios (AR) of less than unity.
b) Interaction of the U-rich surface drainage water with highly anoxic conditions in the organic-rich soils close to the base of the cliff results in very efficient retention of U.
c) Artesian groundwater from fractured and faulted and mineralised bedrock beneath the sediments acts as a second input. Upward flow is modified to sub-horizontal flow by the laminated character of the deposits. This source has an enhanced U concentration relative to the interstitial water concentrations of the overlying sediments and a probable AR value of more than unity.
d) Scavenging by iron oxyhydroxides in the silts at about a metre depth removes a significant quantity of the U from this artesian input [2].
e) Spring and stream water samples draining the study area to the sea are depleted in U relative to the input source waters with concentrations typically less than 15 ppb.
Further descriptions of these points are in hand.

Broubster. Field work took place at the site during the week 26 June to 2 July, with the objectives of a) assessing the hydrogeology of the site, b) collecting groundwaters for speciation measurements with the portable speciation kit [3], and for U/Th series determinations at UKAEA on colloid fractions, c) collecting further samples of the source-term mineralisation to look at the uraniferous hydrocarbon phase, d) collecting more samples of the peat (sink-term) for C-14 dating and organic characterisation work, and e) collecting peat samples for carrying out U radiotracer diffusion tests in the BGS Radiochemistry Laboratory. Results show that for the depth interval of 22-32cm in the peat, the radiocarbon age is 4630 +/- 50 years BP. The laboratory K_d value for U sorption onto the peat is about 10^4 ml/g. Squeezed pore waters from samples of peat and clay demonstrate large U, Th and REE fractionations. Speciation analyses confirm the association of most of the U with organics in the peat waters. Determinations are continuing.

B.2.2 Modelling the field investigations

Needle's Eye. EMP has carried out some preliminary hydrogeochemical modelling [4] with the groundwater composition data obtained by BGS. The solubility of uranium in the system appears to be controlled by liebigite, $Ca_2UO_2(CO_3)_3.10H_2O$, whilst the major element chemistries of the waters imply precipitation of $NaHCO_3$. As $NaHCO_3$ is never found in such environments, caution is indicated when interpreting modelling outcomes. More accurate descriptions are required of the redox conditions of the sediments and of the nature of the uraniferous mineralisation for better interpretations of the groundwater results. These considerations were discussed at a joint meeting of BGS, SURRC, EMP and DOE at Keyworth on 4 th October. It was decided to consider the application of the coupled transport code STELE to the movement of U in the system; the extra data requirements were outlined.

Broubster. The unsaturated nature of the ground leading from the mineralisation down slope to the peat bog limits the task of coupled mass transport modelling to the saturated peat area. The speciation modelling of the geochemistry of the groundwaters has been successfully carried out [5]; a draft report by WS Atkins has been submitted [6]. Figure 2 is a cartoon of the salient features. Experimental measurements of U and Th using the speciation kit and ICPMS analysis support the speciation modelling results; a soluble humate complex of U(VI) is more stable at the pH of the peat environment than a carbonato-complex and is sorbed onto the peat giving rise to a chemical concentration of U as high as 0.1 wt%. Thorium behaves differently, being closely allied to amorphous iron hydroxides and their filtration or precipitation reactions in the peat. Further modelling will await the arrival of more comprehensive results from the analyses of the samples collected at the end of June.

Future modelling applications require better definitions of the background regional contributions of U and Th to the sediments and peat, better descriptions of the source-terms, and a study of the mechanisms of U retardation in organic-rich sediments. This last aspect involves the development of improved ways of measuring redox conditions and organic-metal interactions.

List of publications

[1] Cooke, A. (in press). A desk study of surface diffusion and mass transport in clays. BGS Technical Report WE/88/34.

[2] Roberts, P.D., Ball, T.K., Hooker, P.J. and Milodowski, A.E. (in press). A uranium geochemical study at the natural analogue site of Needle's Eye, SW Scotland. Proc. of the Twelfth Int. MRS Symp. on the Scientific Basis for Nuclear Waste Management, October 10-13, 1988, Berlin.

[3] Breward, N. and Peachey, D. (in press). Development of portable equipment to study physical and chemical phases in natural waters. BGS Technical Report WE/88/25 (DOE ref. no. DOE/RW/88.102).

[4] Doublet, R., Jamet, Ph. and Soubeyran, R. 1988. Modelling transfer in natural analogues. Ecole des Mines de Paris, C.I.G. CEC progress report to 30.6.88, LHM/RD/88/59.

[5] Read, D. and Hooker, P.J. (in press). The speciation of uranium and thorium at the Broubster natural analogue site, Caithness, Scotland. Proc. of the Twelfth Int. MRS Symp. on the Scientific Basis for Nuclear Waste Management, October 10-13, 1988, Berlin.

[6] Read, D. (in press). Geochemical modelling of the Broubster natural analogue site, Caithness, Scotland. BGS Technical Report WE/88/43.

NA Chapman presented an overview and review of progress to January 1988 of natural analogue studies in the CEC MIRAGE-2 Project (1985-9), and PJ Hooker gave a précis of the U.K. natural analogue programme at the CEC Natural Analogue Working Group's Third Meeting in Snowbird near Salt Lake City (USA), June 15-17, 1988. CEC report EUR 11725 EN.

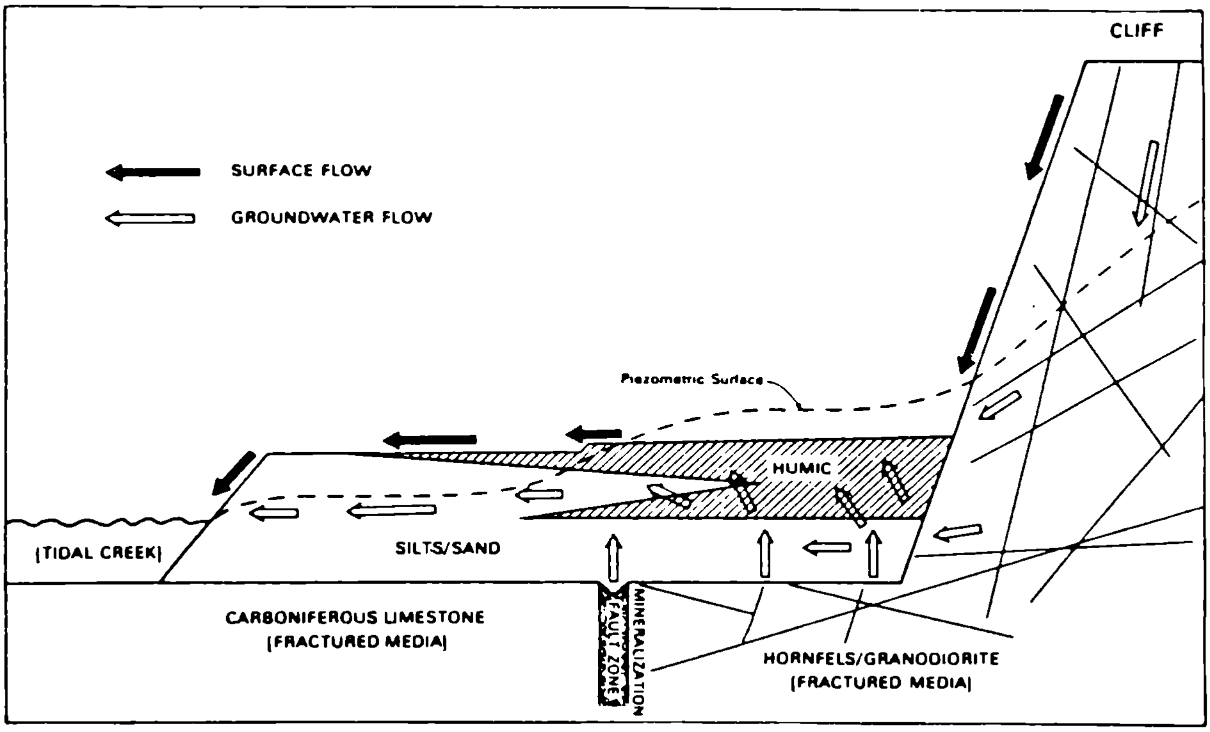


Figure 1. Schematic section of the Needle's Eye site.

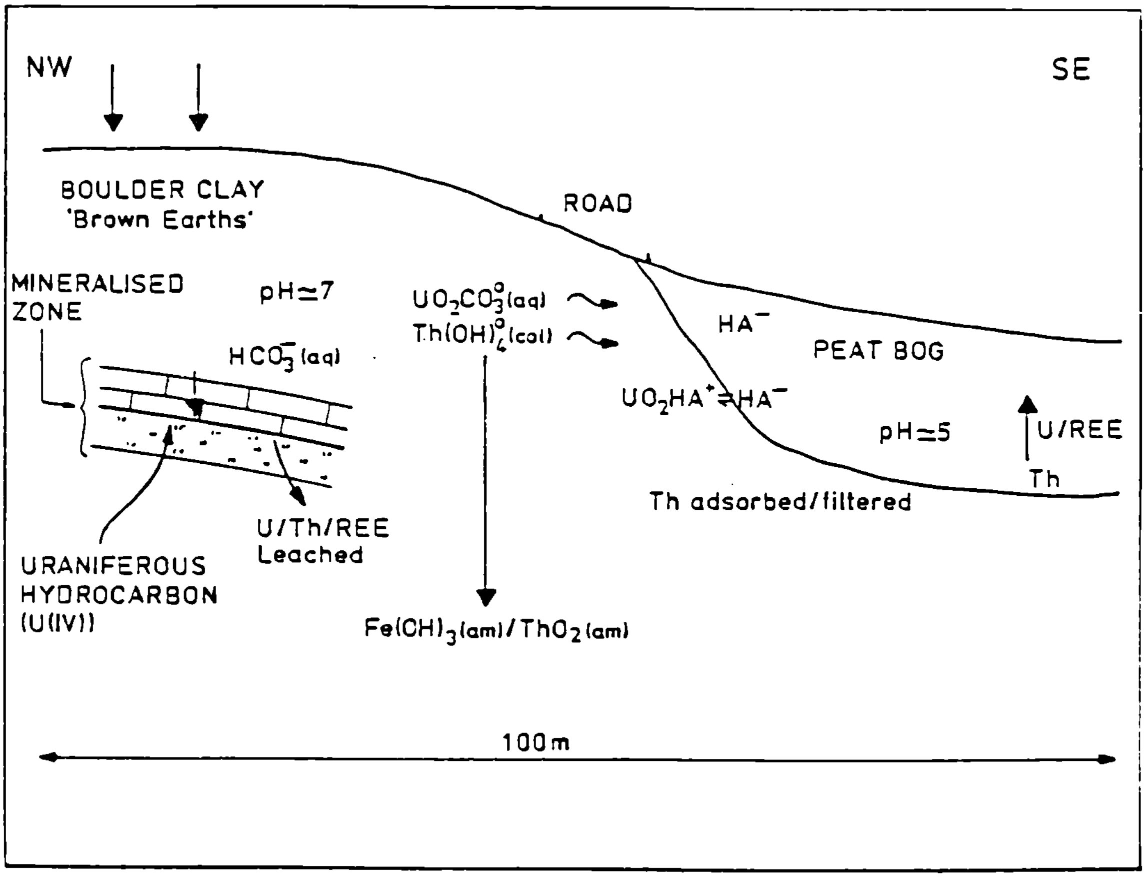


Figure 2. Conceptual model of U and Th transport.

MICROSTRUCTURE AND STATE OF RADIOACTIVE EQUILIBRIUM OF CRYSTALLINE ROCK IN RELATION TO THE RETARDATION OF RADIONUCLIDES BY DIFFUSION INTO THE ROCK MATRIX

Contractor: Univ. of Oviedo (E) and Univ. of Exeter (UK)
Contract N°: FI1W/0143
Duration of contract: 01.10.87 - 30.09.90
Period covered: 01.01.88 - 31.12.88
Project leader: M. Montoto - E. Durrance

A. OBJECTIVES AND SCOPE

In most radionuclide transport models it is assumed that radionuclides migrating in solution along fractures will diffuse into water held in pores within the rock matrix adjacent to fractures, and will thus be retarded relative to the flow of water. The possibility that there exists adjacent to fractures a zone to which diffusion might be restricted, or that the pores adjacent to fractures might be clogged by the products of alteration and/or mineralisation, has not generally been included in radionuclide retardation models.

The objective of this study is to assess the effectiveness of diffusion as a mechanism for the retardation of radionuclides, by examining the microstructure of crystalline rock adjacent to and remote from fractures, and by determining the extent of disequilibrium in the uranium-238 decay series in rock adjacent to fractures. The effect on porosity and potential diffusion of the fractures themselves is also being assessed, with particular regard to stress-relief effects and to pore-clogging by the products of alteration, mineralisation and precipitation. It is hoped that the results of the study will help to resolve a major area of uncertainty in existing radionuclide retardation models.

B. WORK PROGRAMME

(1) Measurement of the porosity and permeability of intact rock adjacent to and remote from water-conducting fractures; determination of the permeability of fracture walls.
(2) Examination by optical, electron and acoustic microscopy of the microstructure of rock adjacent to fractures and the availability of pores for diffusion, using digital image processing techniques.
(3) Examination of the mineralogy of the rock and of fracture and microfracture linings and infillings.
(4) Investigation of the penetration of modern waters into the rock adjacent to fractures by measuring the state of radioactive disequilibrium in the uranium-238 decay series using alpha spectometry.
(5) Determination of the state of oxidation of iron in rock adjacent to fractures using Mossbauer spectroscopy.
(6) Assessment of the effect of in situ stress conditions on the potential for diffusion.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Work to date can conveniently be described under the following headings:

(1) Evaluation of various systems for the determination of intact rock permeability and their application to this project.

(2) Microstructural analysis: digital image processing:
  - (a) Studies aimed at perfecting methods for the preparation of samples for petrographic characterisation by microscopic techniques.
  - (b) Studies of the suitability of acoustic microscopy for the petrographic characterisation of rocks.
  - (c) An appraisal of new parameters for the quantitative petrographic characterisation of rocks.
  - (d) Development and application of digital image processing techniques for the determination of textural properties.

(4) Uranium series disequilibrium studies:
  - (a) Preparation of alpha spectrometry system.
  - (b) Preparation of computer programs for uranium series calculations.

Samples studied to date included Gondomar granodiorite and El Escorial granite from Spain, and Stripa granite from Sweden.

Progress and results

1. Evaluation of various systems for the determination of intact rock permeability and their applicability to this project.

Steady state and non-steady state methods have been assessed for the determination of intact rock permeabilities down to nanodarcy levels, thus encompassing the full permeability range of granites likely to be encountered. Given that the rocks of interest are granitic and of low permeability (10^{-17} - 10^{-21} m^2), the non-steady state (transient or pulse decay) method is most suitable and an instrument of appropriate design has been sought. To this end, discussions have taken place with a number of manufacturers of permeameters in the UK, and the possibility of using existing equipment at Imperial College, London, has also been investigated. The possibility of incorporating a non-steady state permeameter into the project is now being considered.

2. Microstructural analysis: digital image processing.

(a) Studies aimed at perfecting methods for the preparation of samples for petrographic characterisation by microscopic techniques.

Methods for preparing samples for petrographic examination by microscopic techniques have been studied, particularly the impregnation of samples with fluorescent resins for fluorescence optical microscopy. Two aspects have been of particular interest: improvement in the use of existing resins, and the assessment of new resins.

An existing impregnation technique has been redesigned in order to improve the impregnation method. The use of new resins, the viscosity and surface tension of which might allow improved impregnation, has also been investigated. Tests on granites of very low porosity (c. 0.5%) has confirmed the suitability of the selected resins.

(b) Studies of the suitability of acoustic microscopy for the petrographic characterisation of rocks

The applicability of acoustic microscopy to the petrographic characterisation of rocks has been assessed. Scanning acoustic microscopy (SAM) has been used in this study. Early work has been carried out during a period at the University of Oxford on the "Oxford SAM", a 'prototype' developed by the Department of Metallurgy and Science of Materials, and on a commercial Ernst Leitz SAM (ELSAM). This represents one of the first attempts to apply SAM to the study of rocks.

The images obtained by SAM provide considerable information on rock samples. Textural characteristics, such as grain size and shape, are clearly visible. In the granite of El Escorial, for example, the difference in elastic properties of the minerals present allow textural features to be clearly recognised. El Escorial granite has been studied using a number of different techniques: polarising microscopy, fluorescence light microscopy, scanning electron microscopy and scanning acoustic microscopy. The most complete information on cracks is provided by SAM, more cracks being visible than in images obtained by other methods at the same magnification. These 'new cracks' are extremely fine and can only be seen at very high magnification under scanning electron microscopy. Information about sub-surface geometry of the cracks can also be obtained. Acoustic microscopy is also very useful in revealing the subgrain structure of deformed grains and the intragranular microstructure of the rock in samples of El Escorial granite.

(c) An appraisal of new parameters for the quantitative petrographic characterisation of rocks

The applicability and usefulness of the textural coefficient (TC) of Howarth and Rowlands (1987) /1/ to the quantitative petrographic characterisation of Gondomar granodiorite has been assessed in the hope that textural changes close to fractures might be identified. The TC is a non-dimensional quantitative measure of rock texture that describes the form, orientation and degree of interpenetration of the grain mass and the grain packing density. The determination of TC requires the measurement, in thin section, of the length, width, perimeter, angular orientation and area of each grain, and the percentage of grains and matrix. These measurements enable the form of each grain to be described in terms of the aspect ratio and the form factor. The parameters describing the form of the grain mass in terms of TC can then be calculated.

(d) Development and application of digital image processing techniques for the determination of textural properties.

A digital image processing system, developed in Oviedo, is being employed for the measurement of the microstructural properties of selected granites. The software for the system is written in EDL and runs under an EDX operating system. The IMAGO program pack, also developed in Oviedo specifically for the analysis of petrographic microscopy images, has been modified for the study of microstructural features.

The system has been used for the study of samples of Gondomar granodiorite. Grain characteristics have been measured on images obtained from thin sections by polarising optical microscopy. Each thin section was divided into zones using a grid system. To enable the characterisation of individual grains, two or more images were obtained from each zone with different angles between the nichol prisms. Images were digitised to a size of 256x256 pixels, distinguished in 256 levels of grey.

The textural coefficent obtained by this technique (2.14) compares well with that obtained manually (2.18), confirming the suitability of the technique for use in these studies. The values obtained show considerable variation, not only in different directions within the sample but also in the same direction.

4. Uranium series disequilibrium studies

The penetration of modern waters into the rock adjacent to fractures is being investigated by measuring the state of radioactive disequilibrium in the uranium-238 decay series. Alpha spectrometry is being employed following separation of uranium and thorium by ion exchange techniques. An existing method employed in work on the granites of south west England is being modified, with particular regard to the choice of spike added before separation of uranium and thorium to determine recovery. The modifications are aimed at reducing the need for complex corrections, which are required when using spikes of short half-life, and to reduce contamination of detector surfaces by alpha recoil mechanisms. From each of the fractures, cores or blocks studied, 'slices' of rock at increasing distance from the fracture surface have been obtained for analysis, allowing the construction of disequilibrium profiles from fracture faces into the rock matrix.

A system for the ion exchange separation of uranium and thorium from rock samples, their electrodeposition onto steel planchettes, and the recording of alpha particle energy spectra has been assembled, and some samples prepared for alpha spectrometric analysis. U-236 (half-life 2.39×10^7 years) and Th-229 (half-life 7340 years) are being used as spikes. Solutions with activities of 100 disintegrations per minute per gram (dpm/g) have been prepared from 5 ml standards containing 370 Bq of U-236 and 950 Bq of Th-229. The spike solutions have been made up in 0.1 M hydrochloric acid to prolong their storage life.

Existing computer programs URAN and THOR, which convert the raw data from the multi-channel analyser to the required isotopic ratios (U-234/U-238, Th-230/U-234 and Th-230/U-238) are being modified to accommodate the new spikes and other changes in technique.

List of publications

BRIGGS, G.A.D.; DAFT, C.M.W.; FAGAN, A.F.; FIELD, T.A.; LAWRENCE, C.W.; MONTOTO, M.; PECK, S.D.; RODRIGUEZ REY, A. & SCRUBY, C.B. Acoustic Microscopy of old and new materials. Meeting on Applications on Acoustic Microscopy. Sendai, Japan (1988).

RUIZ DE ARGANDOÑA, V.G.; CALLEJA, L.; MONTOTO, M.; SUAREZ DEL RIO, L.M. & RODRIGUEZ REY, A. Petrophysical interpretation of the Acoustic Emission/ Microseismic Activity in heated crystalline rocks". Proc. 9th Int. Acoustic Emission Symposium, pp. 282-291. Kobe, Japan. (1988).

MONTOTO, M. Curso "Análisis Cuantitativo de Imágenes Microscópicas" (3er Simposium Nacional de Reconocimiento de Formas y Análisis de Imágenes). Oviedo, Spain (1988).

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/1/ HOWARTH, D.F. and ROWLANDS J.C. (1987). Quantitative assessment of rock texture and correlation with drillability and strengh properties. Rock Mechanics and Rock Engineering, 20, 57-85.

MIGRATION OF URANIUM DAUGHTER RADIONUCLIDES IN NATURAL SEDIMENTS

Contractor:	Natural Environment Research Council of the United Kingdom
Contract No.:	FI1W/0146.UK
Duration of Contract:	1 December 1987 to 31 May 1990
Period covered:	1 January 1988 to 31 December 1988
Project leader:	Dr. J. Thomson

A. OBJECTIVES AND SCOPE

This project utilises a characteristic uranium profile shape developed in deep-sea turbidite sediments to examine the behaviour of uranium and its daughter isotopes over time-scales desired for the isolation of radioactive waste. The geochemical background is that homogeneous organic-rich turbidites, emplaced in the deep-sea, experience oxidation from the top downwards by bottom water. Uranium redistributes in the turbidite in response to the oxidation front, but this redistribution ceases when the turbidite is isolated from bottom water on emplacement of the next turbidite. Two long cores (MD24 and MD10) are available from a 1985 NEA cruise with several such units up to 750 ky old preserved. Good stratigraphic, geochemical and geotechnical data are available, and pore water advection in the cores is believed to be negligible.

The goals of the project are:

Determination of uranium and its longer-lived daughter radionuclides in individual turbidite units with ages between 250 and 750 ky to compare the observed profiles with those predicted by radioactive ingrowth systematics.

Utilisation of the experimental data to estimate effective diffusion coefficients of the different elements for modelling purposes. Such data will be relevant to the in-situ geochemical conditions of the sediments over the long time scales indicated.

B. WORK PROGRAMME

2.1 Development and verification of a radiochemical analytical scheme for the analysis of uranium-238, uranium-234, thorium-232, thorium-230, protactinium-231, radium-226 and polonium-210.

2.2 Determination of activity versus depth profiles of the above radionuclides for different turbidites of different ages (250-750 ky) in core MD24.

2.3 Determination of a gross uranium profile for core MD10 to guide sampling and comparison of corresponding turbidite unit profiles with those of core MD24.

2.4 Chemical partitioning studies of selected samples (if necessary).

2.5 Model data obtained.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

A suitable radiochemical analytical scheme has been calibrated and is established in routine use.

Eighteen samples from turbidite t and forty samples from turbidite w and surrounding units have been analysed for alpha-emitting nuclides with the above scheme in core MD24 and analysis of turbidite s is underway.

A gross uranium profile has been obtained for samples from the lower 13 m of the second core, MD10.

PROGRESS AND RESULTS

2.1 Development of radiochemical scheme

An analytical method for radium-226 has been established based on lithium metaborate fusion of a separate sub-sample and subsequent measurement via radon-222 emanation. The overall analytical method employing this radium-226 analysis and the uranium and thorium isotope and polonium-210 analysis reported last year has been calibrated using two separate uraninite standards known to be at secular equilibrium, and has been applied to selected samples of core MD24 on a routine basis. This aspect of the work programme is now completed.

2.2 Uranium, thorium, polonium-210 and radium-226 analysis of core MD24

Good radiochemical closure has previously been demonstrated for the decay sequence uranium-238-uranium-234-thorium-230 in turbidite t from MD24. Figure 1 demonstrates that this closure breaks down at radium-226, which is deficient where thorium-230 is at a maximum in turbidite t. Conversely, a depth profile of radium-226/thorium-230 (Fig. 2) indicates that radium-226 is in excess where thorium-230 has its lowest values. Similar results have been found for turbidite w. Turbidite s has the largest maximum/minimum uranium differences in core MD24, and is expected to show an even larger radiochemical disequilibrium. This turbidite is now being analysed.

2.3 A uranium profile for MD10

Figure 3 shows the uranium profile, obtained commercially, for the lower 13 m of MD10, along with the uranium profile for MD24. MD10 contains additional small turbidites, two of which have affected the uranium redistribution pattern in turbidite t. Their incursion restricted the time available for bottom water oxygen penetration into the top of turbidite t in MD10 relative to MD24, and a uranium peak was not developed in turbidite t in MD10. The additional turbidite s1 was exposed to bottom water for a longer time and here a uranium peak and colour change were developed contemporaneously with oxidation in turbidite t in MD24. This aspect of the work programme is now completed.

The large uranium contrast between turbidite t and the overlying unit in MD10 suggests a promising zone for further investigation because of the large local gradients in uranium concentration.

LIST OF PUBLICATIONS

THOMSON, J. 1987. Migration of uranium daughter nuclides in natural deep sea sediments. In: HOOKER, P.J. & CHAPMAN, N. (eds.), UK Natural Analogue Co-ordinating Group: First Annual Progress Report. DoE Report No.: DoE/RW/88.036.

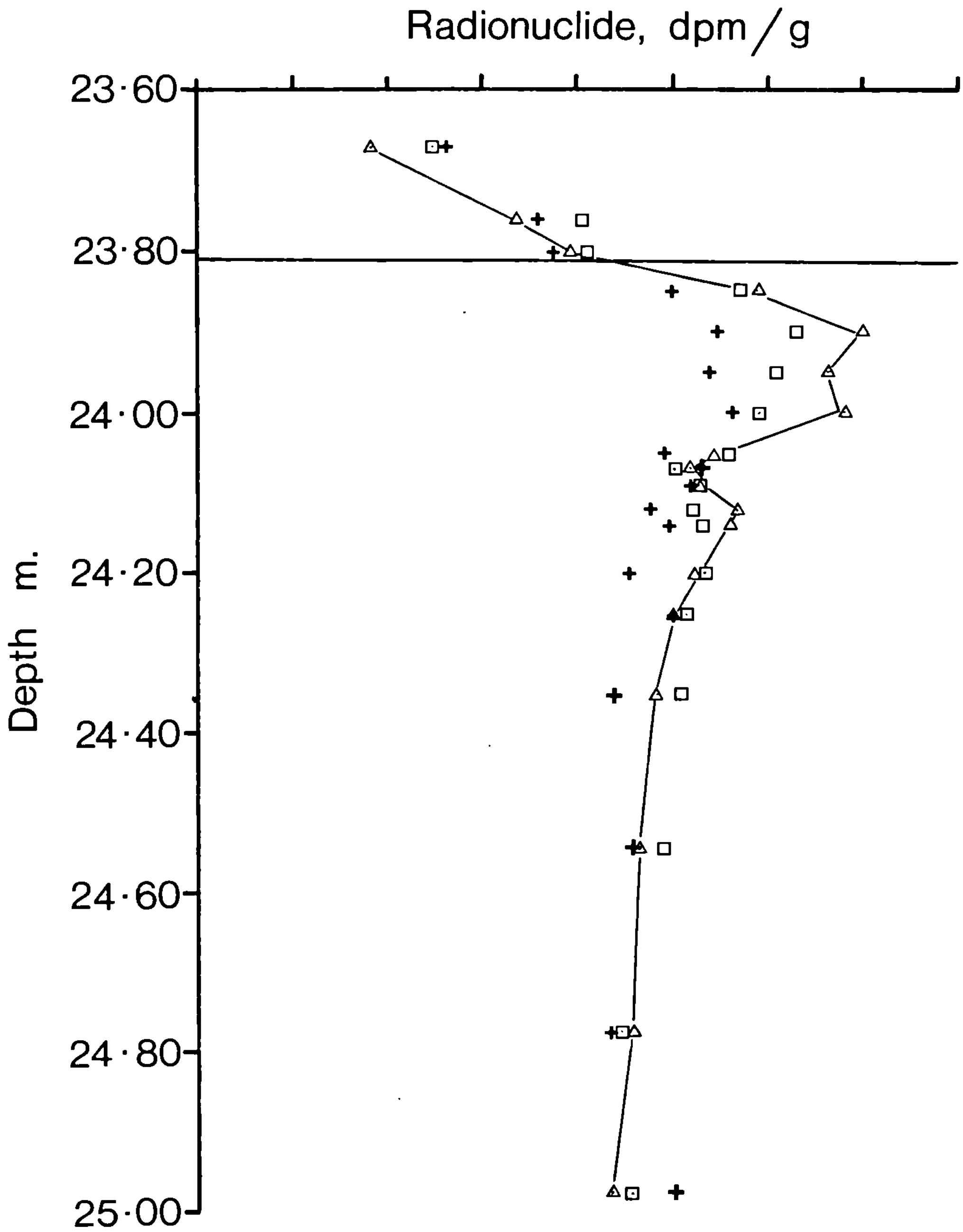


Figure 1: Thorium-230 (triangles), radium-226 (boxes) and polonium-210 (crosses) profiles versus depth for turbidite t from core MD24. The position of the fossil oxidation front is marked by the horizontal line at 23.81m. The thorium-230 data points are joined together to illustrate the profile shape.

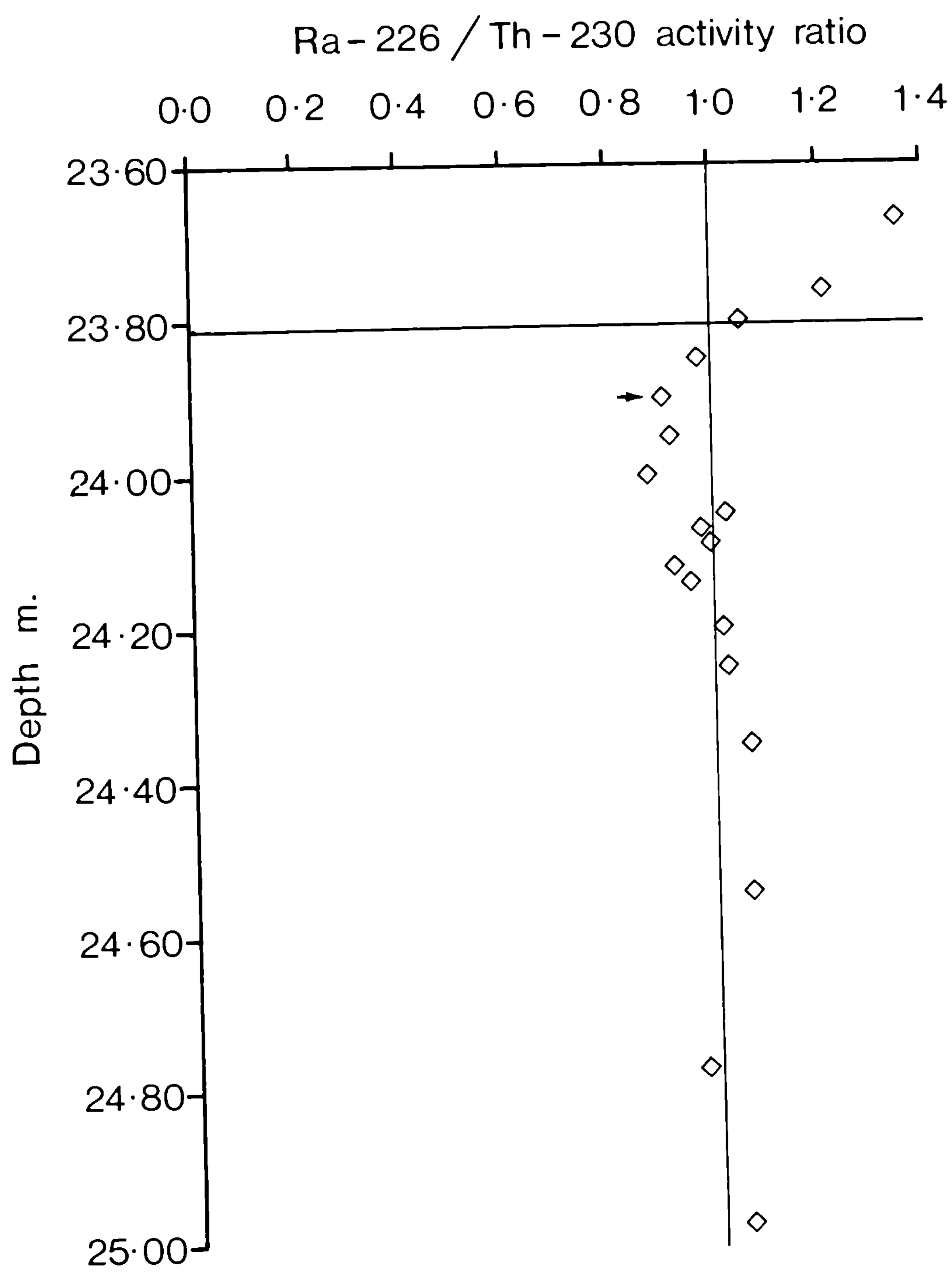


Figure 2: Radium-226/thorium-230 ratio versus depth for turbidite t from core MD24. The position of the fossil oxidation front is marked by the horizontal line at 23.81m. The arrow points to the analysis with the highest uranium-238 value. The vertical line is the secular equilibrium value of this ratio.

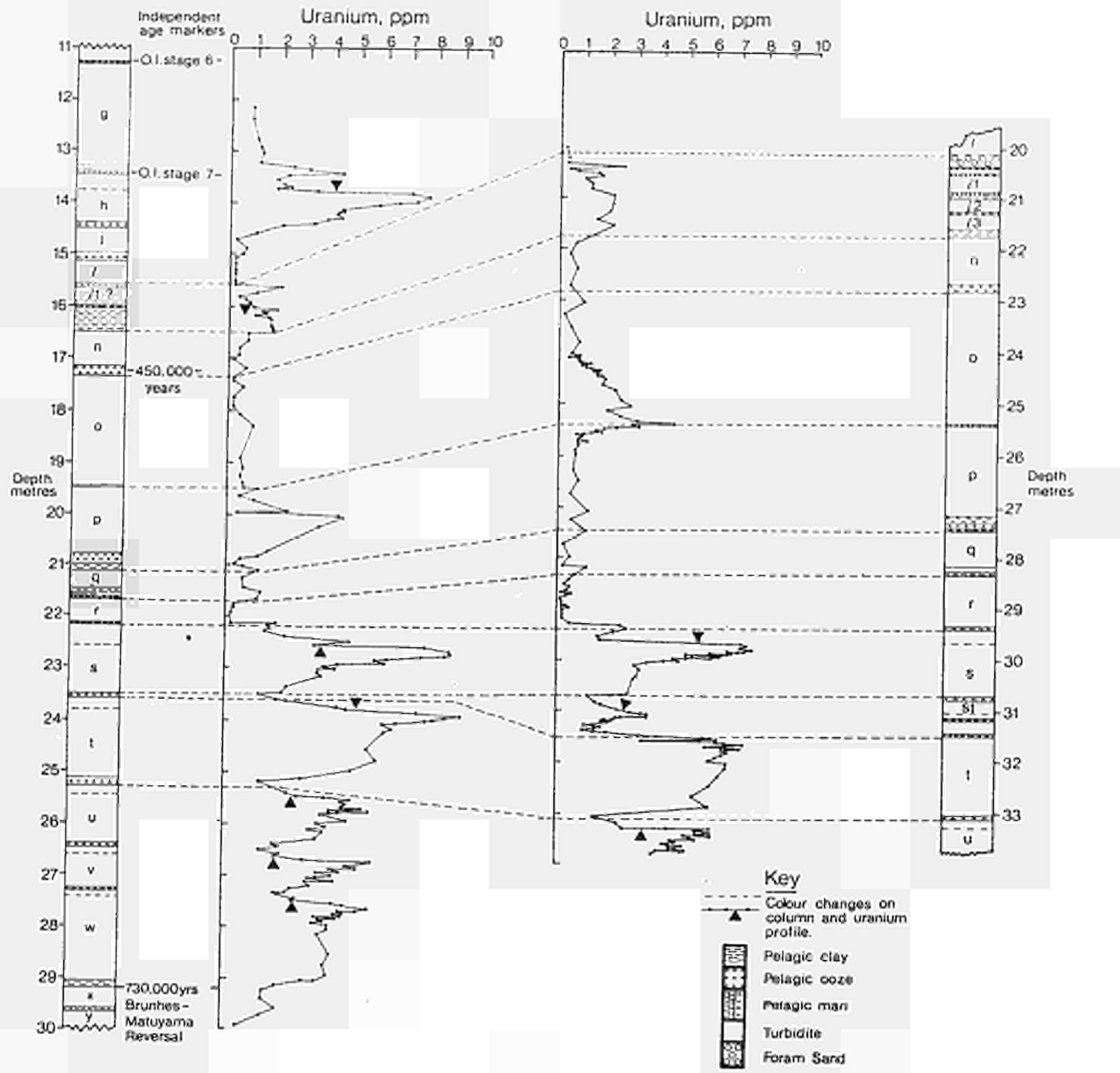


Fig. 3 : Statigraphic logs and solid phase uranium profiles for core MD24 (left hand side) and core MD10 (right hand side). Equivalent units are correlated between cores using dashed lines.

STUDY OF THE MIGRATION OF U, TH AND REE IN AN INTRAGRANITIC URANIUM DEPOSIT

Contractor: CEA-IRDI/DRDD/SESD/SCPCS/LECALT-Fontenay-aux-Roses-France
Contrat n°: FI1W/0149
Duration of contract: from December 1987 to December 1989
Period covered: January 1988 to December 1988
Project Leader: M.T. MENAGER

A. OBJECTIVES AND SCOPES

The intragranitic uranium deposit of the Jalerys (Granite de Grury-Morvan-France) is used a natural analogue of a high-level radioactive waste repository for studying the physico-chemical processes of transport for some trace elements of interest. The project is focussed on near-field migrations.

More specifically, the aims of this investigation are:

* To obtain information on the elemental mobilisations of U, Th and REE through the argillaceous and granitic barriers which surround the ore body considered as a highly concentrated source-term.

* To determine the geochemical mechanisms involved during the transport of radionuclides and to evaluate, as far as possible, their migration rates.

* To confirm the analogy in the geochemical behaviour of transuranic elements and their natural counterparts.

* To characterize, the retention capacity of a repository, that would be realized in a similar geological formation.

* Finally, to model the interaction between the granite, the fluid and the ore body by means of the geochemical code EQ3/6.

This project is conducted in collaboration with COGEMA and CREGU.

B. WORK PROGRAMME

B.1. Sampling of the Grury granite intruded by uraniferous mineralizations (the Jalerys mine and the Jacquot open quarry)
B.1.1. Samples are taken at regular intervals along a direction roughly perpendicular to the vein, from the mineralized body to the fresh granite.

B.2. Mineralogical studies at the laboratory
B.2.1. Petrographical and mineralogical works on primary and secondary parageneses, specially on U-Th-REE-rich ones, with optical and electron microscopy (scanning and transmission coupled with X-rays analysis) as well as fission track micromapping.
B.2.2. Study of the geochemical properties of the granite and of their evolution during alteration (major and trace elements analysis by ICP and neutron activation analysis).
B.2.3. Determination of the physico-chemical properties of fluids responsible for the alteration by the study of fluid inclusions (microthermometry, Raman microprobe).

B.2.4. Evaluation of temperatures active during the functioning of the system by the study of the thermal annealing of fission tracks registered in appropriate minerals.

B.3. Isotopic studies at the laboratory
B.3.1. Study of radionuclide mobilizations using uranium and thorium-series disequilibria for dating the last opening of the system (α and γ spectrometries, ion microprobe).
B.3.2. Dating of alteration minerals, whenever feasible.
B.3.3. Evaluation of the sorption efficiencies for U, Th and REE of granite-forming minerals for various degrees of alteration corresponding to different surface states (Rutherford backscattering, specific nuclear reaction analysis).

B.4. Modelling
B.4.1. Modelling of interactions between solids and solutions by means of the EQ3/6 geochemical code and comparison with data of the natural system.
B.4.2. Simulation with this code of the evolution with time of a high level radioactive waste intragranitic repository.

C. PROGRESS OF WORK AND RESULTS OBTAINED

STATE OF ADVANCEMENT

During this year, petrographical and mineralogical determinations have been performed by means of optical microscopy, X-ray diffraction, scanning and transmission electron microscopies coupled with energy dispersive x-ray analysis. Concentrations of major, minor and traces elements were obtained by inductively coupled plasma spectrometry and neutron activation analyses for the whole rock and by the latter method only for separated alteration phases. The study of fluid inclusions is in progress.

PROGRESS AND RESULTS

Alteration of granite

Four successive alteration stages were identified in the granite and two main periods can be distinguished:

* a high temperature period (400-200°C) including two stages with no major U-remobilization. The U-deposit occurs at the end of this period.
* a low temperature period (180-25°C) including two other stages with major U-remobilization.

In the first period, µm to mm-wide fissures of limited density occurred during a tardimagmatic event. A paragenesis of orthoclase-quartz-pyrite fills these fractures. The literature data allow us to determine that this event took place at a temperature of about 400°C.

The following phase is associated with major fractures and to the formation of dm to m-wide brecchia veins. We can observe a preferential alteration of biotites and of plagioclases in the granite: from the brecchia vein to 10 meters away biotites are altered into white micas whereas they are transformed into chlorite between 10m and 30m. Plagioclases are transformed into white micas over the all profile. A temperature range of 300-200°C can be assumed for such parageneses. A decreasing intensity of alteration with distance from the vein can be inferred.

During the low temperature period, a third event is associated to either fractures of limited intensity or recurrent faulting (?). Illite-smectite mixed layers (illite-smectite) are formed in plagioclases and fissures. For this phase a 180-100°C temperature range is suggested by literature data.

The last alteration phase, characterized by the development of micrometric veins filled by calcite and/or kaolinite, occurs for a temperature lower than 100°C.

All these steps are associated with haematization.

U, Th and REE mobilizations

During the high temperature period, the mobilization of U, Th and REE is limited and strictly associated to the alteration of granite accessory minerals. For example, the U released during alteration of biotites is trapped by Ti-oxides. The Th and REE released by the corrosion of monazite show a differential behaviour, as they are incorporated in silicate and carbonates respectively.

On the other hand, during the low temperature period, alteration processes affect both the granite and the U-ore body. Large mobilizations of U occurred and a gradient of U dispersion can be evidenced. For example, the alteration products in plagioclases like illite/smectite mixed-layer minerals and haematite trapped respectively $\simeq$15 ppm and $\simeq$27 ppm of U. In veins, mixed-layers, calcite and haematite show respectively $\simeq$30 ppm, $\simeq$15 ppm and $\simeq$30 ppm of U.

Distinctive retention scales occur: in "near field" ($\lesssim$ 10m), U is trapped in secondary minerals such as phyllosilicates, Fe-oxi-hydroxides and U-secondary minerals, in contrast to the "far field" ($\gtrsim$ 10m) where U is preferentially associated with carbonates.

Different retention mechanisms have been identified: incorporation in phyllosilicates, sorption on Fe-oxi-hydroxides, Ti-oxides, phyllosilicates, and precipitation of thorite and REE-carbonates.

REFERENCES

DRAN, J.C., DELLA MEA, G., PACCAGNELLA, A., PETIT, J.C., MENAGER, M.T., Sorption of actinides analogues on granite minerals studied by MeV ion beam techniques. Radiochimica Acta, 44/45, 299-304 (1988).

MENAGER, M.T., PARNEIX, J.C., PETIT, J.C., DRAN, J.C., Migration of U, Th and REE in a fossil intragranitic geothermal system: implications for the mobility of actinides around a radwaste disposal. Radiochimica Acta, 44/45, 291-297 (1988).

PARNEIX, J.C., MENAGER, M.T., TROTIGNON, L., PETIT, J.C., Hydrothermal alteration in the Auriat granite (Massif Central, France): analogy with a radwaste disposal. CEC Report n° EUR 11037 EN (1987) 13pp.

MENAGER, M.T., PETIT, J.C., MENET, C., Elemental remobilizations around the U-mineralized vein of the Jalerys (Morvan). Proceedings of the International Congress of Geochemistry and Cosmochemistry, Paris, August 29-September 2 1988, Chemical Geology pp 136.

4.3.D. Development of calculation tools for the description of radionuclide migration

COUPLING BETWEEN A GEOCHEMICAL MODEL AND A TRANSPORT MODEL OF DISSOLVED ELEMENTS

Contractor : Centre d'Etude Nucléaires de Fontenay-aux-Roses, CEA/IPSN/DAS/SAED - FRANCE
Contract n° : FI1W/0075
Duration of contract : from december 1986 to december 1988
Period covered : january 1988 - december 1988
Project leader : P. JACQUIER

A. OBJECTIVES AND SCOPE

In order to assess the safety analysis of an underground repository, we have to calculate the transport of radioelements in groundwater and their interactions with the geological medium. So the objective of this study is the elaboration and experimental validation of the coupling of a geochemical model with a transport model of dissolved elements.
Flow-through experiments were conducted, last year, on columns filled with crushed limestone, by the Département d'Etudes et de Recherches en Sécurité of the CEA in Cadarache.
The mathematical codes have been set up by the Centre d'Informatique Géologique, with purpose of interpretation of these experiments and validation of the coupled code. We have reached the last phase of the contract.

B. WORK PROGRAMME

B.1. Analysis of the results of elements chemical analysis at the outlet of the column for the eleven experiments.
B.2. Screening of these results, with computation of the electric imbalance, by the geochemical code CHIMERE.
B.3. Attempt of interpretation of these results, aiming at coupling geochemical and transport phenomena with a coupled code STELE.
B.4. First conclusions.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The study is achieved.

Progress and results

The item B.1. /1/ displays some abnormalities, concerning, for instance, the electric imbalance for some experiments. Nevertheless, it appears that, by computation of the saturation index of calcite or strontianite, the values are not very different with or without correction of the electric imbalance.
The item B.2. /1/, /2/ displays interesting results like the connection between the calcium behaviour, the injected $SrCl_2$ amount and the other species behaviour like magnesium, sodium (table I).

Evolution of the concentration of strontium and chloride at the outlet of the column, with respect to the inlet concentration

We can notice two types of curves for the strontium :
1) When the inlet concentration of $SrCl_2$ is 10^{-3} mole per liter, during thirty hours the outlet concentration of strontium remains equal to about the hundredth of its inlet level. Then its concentration increases continuously. In the experiment with distilled water at 38° C, it reaches the inlet concentration at the end of eighty hours (figure 1).
2) When the inlet concentration of $SrCl_2$ is 10^{-1} mole per liter, the outlet concentration of strontium is on the same order of magnitude as the inlet one, from the beginning of the curve.
To explain the two behaviours of strontium, we can invoke precipitation of strontium carbonate at the surface of the calcium carbonate particles ; this is visible for low concentrations (10^{-3} molar).

Conclusion

This is the first attempt to explain experimental results with a coupled model.
A first order rate kinetic is not enough to explain phenomena of strontium carbonate precipitation.
Now, there are two research ways which complete one another :
- improving models in order to take into account more phenomena,
- pursuing the identification of chemical mechanisms.

/1/ VINSOT A. - COUDRAIN-RIBSTEIN A. (1988)
Elaboration et validation expérimentale d'une méthode de résolution du couplage entre un modèle géochimique et un modèle de transport d'éléments en solution.
Rapport d'avancement LHM/RD/88/39

/2/ VINSOT A. - COUDRAIN-RIBSTEIN A. (1988)
Elaboration et validation expérimentale d'une méthode de résolution du couplage entre un modèle géochimique et un modèle de transport d'éléments en solution.

Commission des Communautés Européennes
Contrat avec le CEA/IPSN/DAS n°FI1W/0075
Rapport final LHM/RD/88/107

/3/ JACQUIER P. - COUDRAIN-RIBSTEIN A. - VINSOT A. - VINSON J.M.
Coupling between a geochemical model and a transport model of dissolved elements
Twelfth international symposium on the scientific basis for nuclear waste management
(Berlin, 10-13 octobre 1988)

Table I : Concentration of calcium at the end of the experiment

Experiment	Measured Calcium at the end
Distilled water	$0,2 \times 10^{-3}$ M
Equilibrated water	$0,3 \times 10^{-3}$ M
Distilled water + $SrCl_2$ 10^{-3} M	$0,9 \times 10^{-3}$ M
Distilled water + $SrCl_2$ 10^{-1} M	33×10^{-3} M
Equilibrated water + $SrCl_2$ 10^{-1} M	$0,03.10^{-3}$ M at room temperature 60.10^{-3} M for T = 38° C

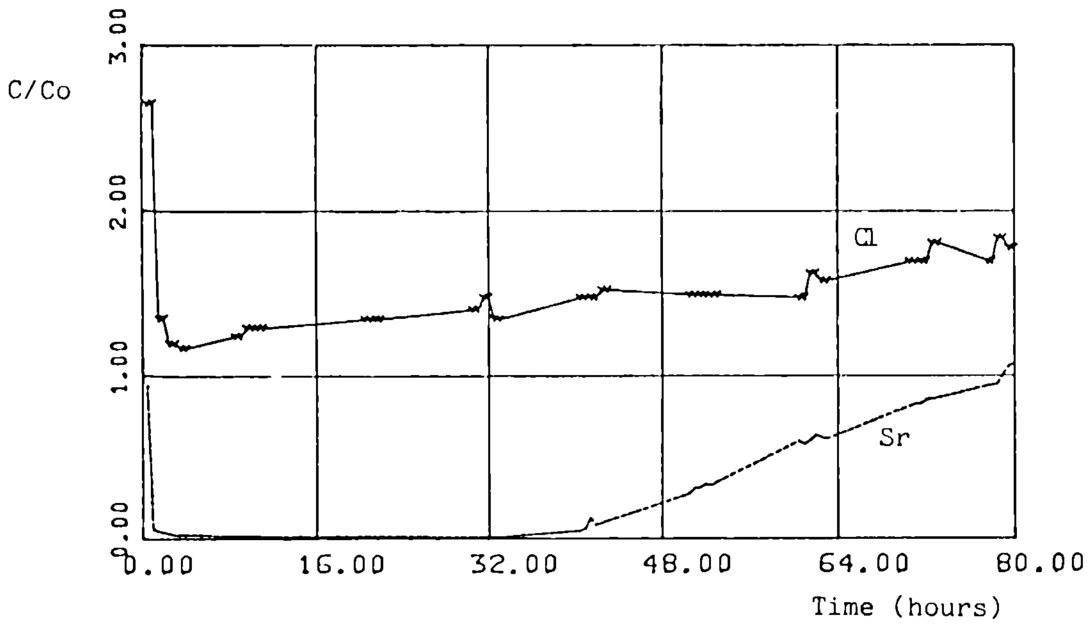


Figure 1 : Evolution versus time of the concentration of strontium and chloride at the outlet of the column, with respect to the inlet concentration ($SrCl2$ 10^{-3} M distilled water, T = 38°C)

Modelling of migration phenomena in the Boom clay and of heat dissipation from a HLW repository in the multi-layered hydrogeological systems surrounding the Boom clay

Contractor : SCK/CEN, Mol, Belgium
Contract No : FI1W/0055/B
Duration of contract : October 1986 to December 1989
Period covered : January 1988 - December 1988
Project leader : A.A. Bonne

A. OBJECTIVE AND SCOPE

In 1974 SCK/CEN launched a R&D-programme concerning the possibilities for disposal of high-level solidified and alpha-bearing radioactive wastes in a continental stratiform clay formation (Boom clay) situated below its own site. Several specific investigations still need to be further undertaken in order to characterise more accurately the argillaceous formation in view of assessing its appropriateness for hosting radioactive waste as well from engineering point of view as for long term safety and performance evaluations. In support of these also further modelling efforts are required in order to improve and confirm our prediction capability.

B. WORK PROGRAMME

B.4.1. Implementation and adaptation of the analytical code MICOF

B.4.2. Evaluation of the thermal impact due to HLW disposal into a stratified clay formation in a multi-layered aquifer system (Mol site) (completed and reported)

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

In addition to the updating of the MICOF-code in order to allow 3-dimensional calculations for discrete sources and different boundary conditions, a coherent conceptual model for the calculation of the transport through the Boom clay, and for the interpretation of migration experiments has been developed.

Concerning the performance of the research related to the heat dissipation in a multi-layered sedimentary sequence the work have been completed end 1987 and reported previously according to the contractual time-schedule.

Progress and results

For the modelling of migration phenomena in the Boom clay, the concept of the "diffusion accessible porosity" has been further developed /1/. This gives a coherent conceptual model which, up to now, fits all the results of our experiments. The model describes the transport of sorbed and nonsorbed species through a liquid saturated porous medium. As far as ion exchanging species are concerned, there is experimental evidence that K_D values from batch experiments may be used in the model for the calculation of the transport of retarded species, and this without any need for a so called "surface diffusion".

For the interpretation of the diffusion experiments, the calculations has been further refined to describe more precisely the experimental conditions (e.g. the inclusion of the filterplates in the calculations of the "flow-through type" diffusion experiments). This gives a better consistency of the obtained data. The interpretation of the results of the diffusion experiments on reconsolidated clay plugs has given a better understanding of the diffusion mechanism and a basis for the concept of "diffusion accessible porosity".

LIST OF PUBLICATIONS

M. PUT, P. HENRION
Radiochimica Acta 44/45, 343-347 (1988)

INTERCOMPARISON OF PREDICTIVE COMPUTER PROGRAMS FOR RADIONUCLIDE MIGRATION IN THE GEOSPHERE

Contractor:	W.S. Atkins Engineering Sciences, Epsom, U.K.
Contract No:	FI1W/0077
Duration of Contract:	February 1987 - January 1990
Period Covered:	January 1988 - December 1988
Project Leaders:	T W Broyd, D Read

A. OBJECTIVES AND SCOPE

Prediction of the transport of radionuclides through the geosphere requires detailed knowledge of aqueous geochemistry, host-rock mineralogy and the specific mechanisms of solid-solution interaction. Such knowledge often needs to be obtained using computer models and associated databases, which must be shown to provide accurate results, i.e. are validated for use. International code verification and proving exercises have become accepted as an important means of assisting the validation process.

This contract will establish an international project called CHEMVAL, aimed at reviewing current progress and establishing research needs in the areas of chemical and chemical transport modelling. The objectives of CHEMVAL are as follows:

i) to produce the best possible overall thermodynamic database for use with aqueous speciation and coupled chemical transport codes, consistent with project resources and time-scales.

ii) to apply aqueous speciation computer models to a range of realistic waste disposal situations, and hence to establish and/or confirm areas of research requirement.

iii) to provide validation both for aqueous speciation models and coupled chemical transport codes.

B. WORK PROGRAMME

1. Comparison, sensitivity and extension of databases. The results of other parts of the work will be used in determining the most appropriate course of action.
2. The application of available aqueous speciation programs and databases to a range of hypothetical, though realistic, waste disposal situations.
3. The application of available programs coupling chemistry and waste transport to a range of test cases.
4. Validation of aqueous speciation and coupled codes by comparison, where possible, with a) experiments, b) field tests and c) natural analogues.

The exact scope and nature of work will be as agreed at plenary meetings of CHEMVAL participants, to be held throughout the duration of the project.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

CHEMVAL may be considered in terms of two related subject areas. The first deals with aspects of computer program verification/ validation and is designed to increase confidence in chemical speciation modelling. It involves the active participation of 14 research organisations within EC member states, Finland, Switzerland and Sweden. The remainder of project resources are devoted to reviewing, extending and improving the body of thermodynamic data available to participants, concentrating on those elements of greatest perceived relevance to radiological assessment. Within CHEMVAL, aspects of thermodynamic data are being considered primarily by the University of Manchester and the University of Wales. The organisation structure of the project is shown in Figure 1.

The modelling components of CHEMVAL comprise four main stages:-

STAGE 1 : application of aqueous speciation codes to representative cement water and groundwater compositions.

STAGE 2 : attempts at validation of aqueous speciation models by comparison with experimental field and laboratory data

STAGE 3 : verification of coupled chemical transport codes

STAGE 4 : attempts at validation of coupled models by comparison with experimental field and laboratory data.

At the end of 1988, Stage 1 has been completed, and Stages 2 and 3 are well underway. Database activities will continue throughout the project.

D. PROGRESS AND RESULTS

STAGE 1 - The objectives of CHEMVAL Stage 1 may be summarised as:-

- to assess the status of computer codes employed within Europe and to verify them on a variety of problems relevant to radioactive waste disposal.
- to highlight differences in thermodynamic data used at each organisation and the importance of such differences in determining radioelement speciation and solubility.
- to confirm areas where further work is required.

An agreed methodology was adopted for the verification study in order to distinguish variation in results caused by differences in thermodynamic data from the caused by other factors, such as coding/operator error or decisions made by the user at input. Participants were asked to perform calculations first with their own "in-house" database and then to repeat the simulations with a standard CHEMVAL database, issued by Atkins ES. This Stage 1 database contained formation constants for 376 aqueous complexes and solubility products for 259 solids.

Test cases were based on actual field or laboratory analyses and were constructed to reflect the various disposal options envisaged by EC member countries. They comprise a cementitious "near-field" and two alternative "far-fields"; clay aquitard - sandstone aquifer and granite host - limestone aquifer. Roughly six simulations were devised for each of the five systems, ranging from relatively simple base case problems of aqueous speciation to complex reaction path calculations involving solution mixing and "irreversible" oxidation or reduction. The radioelements included were those considered important during earlier radiological assessment programmes, namely americium, plutonium, uranium and technetium, plus possibly, caesium, strontium and cobalt in the case of rapid release.

All of the codes used during the exercise were capable of solving the wide range of geochemical problems set (Figures 2 and 3). Excellent agreement has been obtained not only for straightforward speciation calculations but also for conceptually difficult reaction path simulations (e.g. Fig 4). This provides confidence in the data used and is also encouraging in the sense that little misinterpretation occurred on the part of individual participants. Other cases, including those concerned with redox reactions and organic complexation, displayed poorer agreement (e.g. Fig 5), however, and the lack of consistency especially in predicted trends requires further investigation.

A report has been produced on Stage 1 activities /1/ which will be published within the CEC's EUR series.

STAGE 2 - Whereas Stage 1 of CHEMVAL has considered realistic but hypothetical test cases, Stage 2 is concerned with the performance of models and databases for simulating real field or experimental datasets. Four sites are being studied:

Mol - representing a clay groundwater surrounding a repository.
Gorleben - simulating a saline water above a repository in salt.
Maxey Flats, Kentucky - an existing, near-surface repository.
Oman - natural, highly alkaline spring water providing an analogue to a cementitious near-field environment.

Test cases of varying complexity have been derived for each of the sites in question. The modelling work is proceding in two phases. In the first, participants have been asked to provide "blind" simulations, that is in the absence of field results. The second phase will occur early in 1989, and will entail participants trying to match field results, and reporting on the methodologies employed.

STAGE 3 - Only a relatively small subset of participants have the capability to simulate coupled chemistry and transport. An initial phase of Stage 3 has shown that candidate codes of a variety of complexity and solution algorithms can produce very similar results for uncoupled cases of aqueous speciation and solute migration. Test cases fully coupling the chemistry and transport will be approached early in 1989.

STAGE 4 - This has yet to commence.

Database Activities - Work on the production of a reviewed thermodynamic database will continue throughout the project. A comparison of some widely used databases has been prepared /2/ which will be published as a CEC EUR report. An extensive literature review of thermodynamic constants is being backed up by the use of model control cases and sensitivity studies, aimed at establishing the effect of proposed modifications to the database. The CHEMVAL database issued in Stage 1 is thus being updated throughout the project, and will be issued to participants at the beginning of each subsequent Stage of work.

References

/1/ READ, D. and BROYD, T.W., Report on Stage 1 of project CHEMVAL/ MIRAGE: Verification of speciation models. 1989.

/2/ CHANDRATILLAKE, M., NEWTON, G.W.A., ROBINSON, V.J., Comparison of thermodynamic databases used in geochemical modelling. EUR11891EN, 1988.

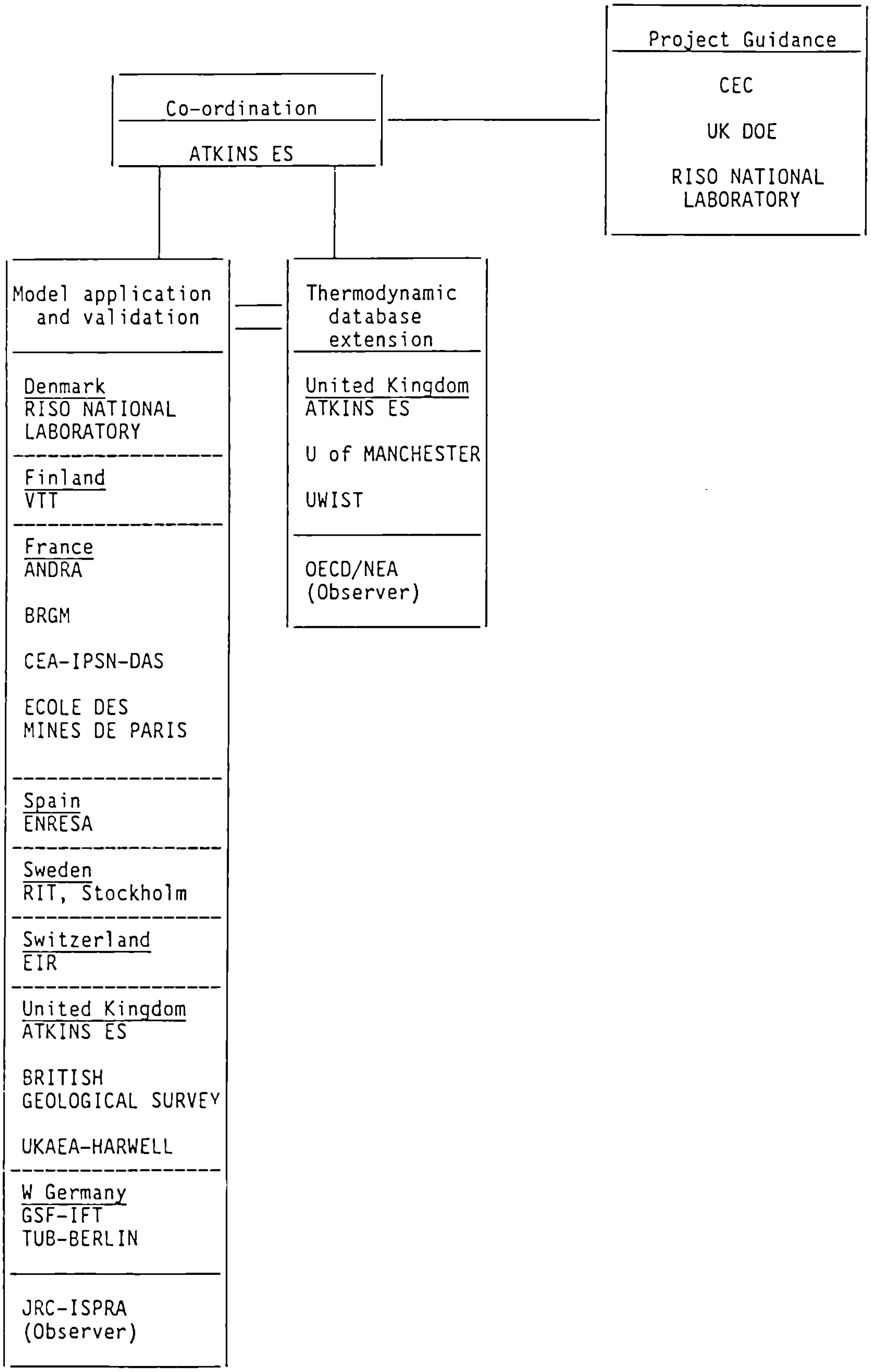


FIGURE 1 ORGANISATION OF CHEMVAL

FIGURE 2 - CHEHVAL STAGE 1 : VERIFICATION "IN-HOUSE" DATA

Results recieved

	DATABASE SUPPLIED	A CEMENT 1	2	3	4	5	6	7	B CLAY 1	2	3	4	5	6	C SANDSTONE 1	2	3	4	5	D GRANITE 1	2	3	4	5	E LIMESTONE 1	2	3	4	5
BGS	YES	*	*	*	*	o	*	*	*	*	*	*	*	*															
RISO		*							*						*		*			*		*			*				
AERE	YES	*	*	*					*	*	*	*	*		*	*	o	o	*										
BRGM/ANDRA	YES	*	*	*	*		*		*	*	*		*							*	*	*	*						
EMP/CEA		*							*						*														
GSF	YES	*	*	*	*	*	*	*	*	o				*	*	*	*	*	*										
TUB	YES	*	*	*	*	*	*	*	*	*	*	o	o	o	*	*	*	o	*	*	*	o	o	o	*	*	o	*	o
UWIST	YES	*	*	*	*	*	*	*	*	*	*	*	*	o	*	*	*	*	*	*		*	*	o	*	*	*	*	*
AES	YES	*	*	*	*	*	*	*	*	*	*	*	O	*	*	*	*	o	*	*	*	*	*		*	*	*	*	*
RIT		*	*	*	*		*		*	*	*				*	*	*			*	*								
PSI		*	*	*	*		*	*	*	*										*									
VTT	YES	*	*		*																								

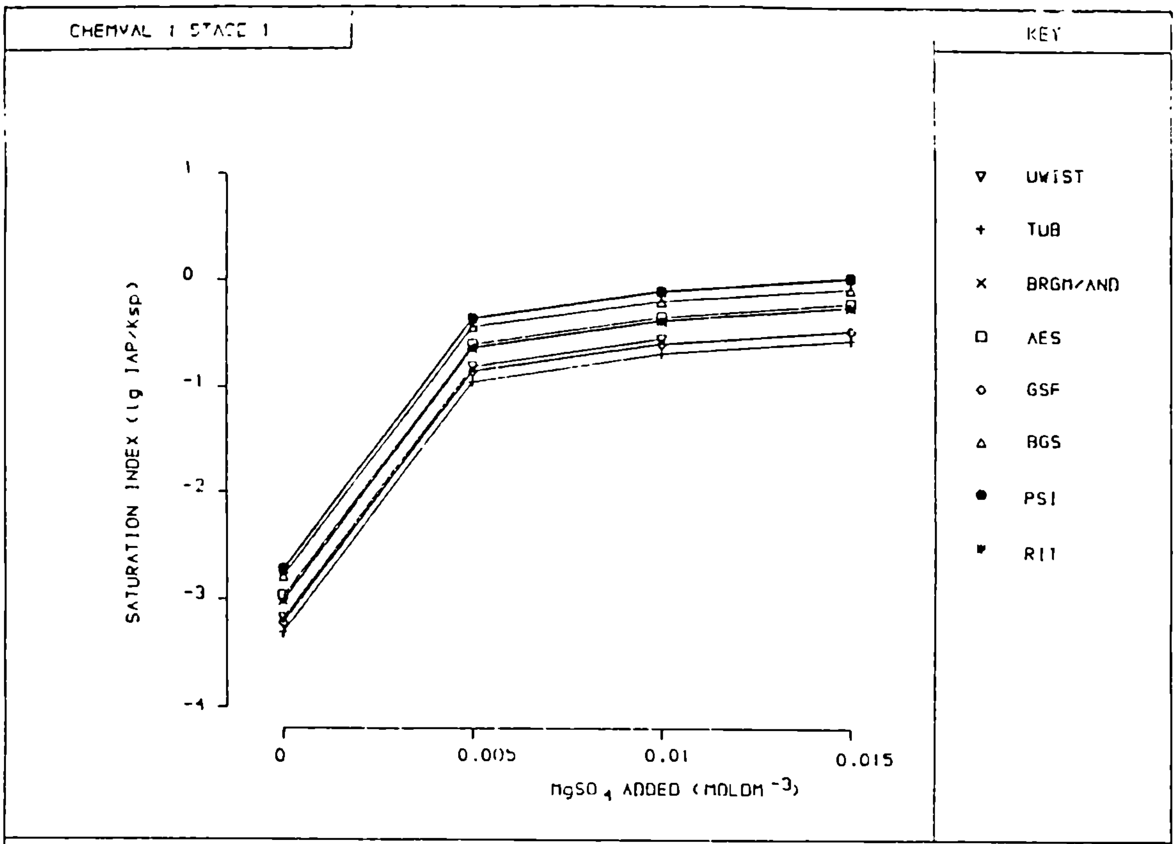
* run completed   O run attempted

FIGURE 3 - CHEHVAL STAGE 1 : VERIFICATION CHEHVAL DATABASE

Results recieved

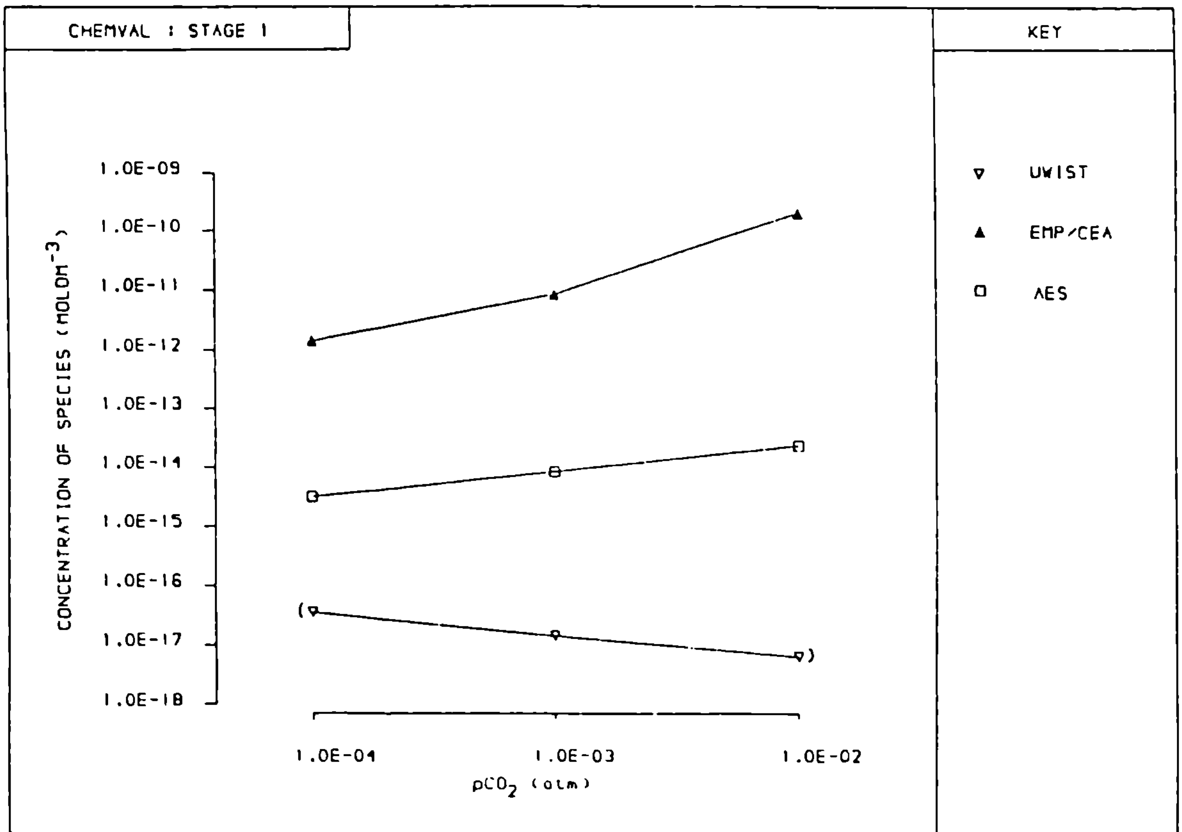
	A CEMENT 1	2	3	4	5	6	7	B CLAY 1	2	3	4	5	6	C SANDSTONE 1	2	3	4	5	D GRANITE 1	2	3	4	5	E LIMESTONE 1	2	3	4	5
BGS	*	*	*	o	o	*	*	*	*	*	*	o	*															
RISO																												
AERE								*	*	*		o	o	*			*	*										
BRGM																												
EMP/CEA	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*		*	*	*	*	o
GSF	*						*	*						*														
TUB	*							*						*					*					*				
UWIST	*	*	*	*	*	*	*	*	*	*	*	*	o	*	*	*	*	*	*		*		o	*	*	*	*	o
AES	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*	*		*	*	*	*	*		*	*	*	*	O
RIT	*	*	*	*	*	*		*	*	*		*	*	*	*		*	*	*	*	*	*		*	*	*		*
VTT	*	*	*	*		*													*	*	*	*						

* run completed o run attempted



TEST CASE A.6. SULPHATE ATTACK
SATURATION INDEX - GYPSUM

FIGURE 4



TEST CASE E.4. ORGANIC COMPLEXATION (CHEMVAL DATABASE)
CONCENTRATION OF U-EDTA SPECIES

FIGURE 5

FAR-FIELD MODELLING OF RADIONUCLIDE MIGRATION

Contractor : Harwell Laboratory, United Kingdom
Contract No. : FI1W/0078
Duration of contract : January 1987 - December 1989
Period covered : January 1988 - December 1988
Project leader : J D Porter, K H Winters

A. OBJECTIVES AND SCOPE

Numerical simulation of groundwater flow and solute transport is an essential element of any assessment of the performance of a proposed repository for nuclear waste. The accuracy and realism of such simulations must be clearly demonstrable and also defensible against critical public scrutiny. Detailed simulations are expensive so any increased computational efficiency offers the opportunity for more extensive sensitivity analysis as part of a comprehensive repository asessment.

The project aims to improve the capability, efficiency and realism of the NAMMU code, which simulates groundwater flow and solute transport through a porous medium. Our detailed objectives are to discover and exploit superior methods for solving significantly non-linear problems; to introduce into the code a capability for modelling chemical reactions; to identify and test improved techniques for simulating the progress of sharp fronts in solute concentration; and to identify better-founded representations of solute dispersion. The last two topics will be covered by sub-contracts placed at Universities in the United Kingdom.

The NAPSAC code presently calculates flow through a three-dimensional network of fractures. Our aim is to decide upon and implement the most appropriate method for calculating solute transport through a three-dimensional network, so NAPSAC could be used to investigate radionuclide migration from a repository in hard fractured rock.

B. WORK PROGRAMME

1. Improved methods for solving significantly non-linear groundwater-flow problems will be implemented in NAMMU because those currently available, although robust, are expensive.
2. The NAMMU code will be enhanced to provide a means of modelling the impact of chemical reactions on solute transport in a porous medium.
3. Methods will be assessed and tested that offer prospects of simulating efficiently the advance of sharp fronts in solute concentration, without spurious dispersion.
4. Models of inhomogeneous materials will be explored to provide better understanding and descriptions of solute dispersion.
5. The NAPSAC fracture-network code will be enhanced by incorporating a judiciously chosen technique for calculating solute transport.

C. PROGRESS OF WORK AND RESULTS OBTAINED

State of Advancement

There has been substantial progress in all the work areas in 1988. A number of solution methods for non-linear problems (Newton Raphson, Broyden and BFGS methods with and without line searches) have been investigated and results indicate that the iterative method due to Broyden is robust and gives significant savings in computer time for problems of interest.

Modifications to NAMMU to allow for concentration dependent or non-linear sorption have been tested and work is now in progress on the inclusion of a ternary heterovalent ion exchange model within NAMMU. The model is currently being validated by comparison with examples in the literature.

A sub-contract has been established with Cranfield Institute of Technology to investigate numerical methods for sharp fronts and a number of techniques have been tested on one-dimensional problems. The most promising of these is now being tested on two-dimensional problems on both structured and unstructured grids.

A sub-contract has been established with the University of Newcastle upon Tyne to investigate models of solute dispersion. A review of the literature in this area has been carried out and a computational study based on a Monte Carlo approach is underway.

The NAPSAC network flow code is being verified for use in the STRIPA project and a review of methods for representing transport processes is underway.

Progress and Results

Improved Solution Methods for Non-Linear Problems (Task 1)

The aim of the work on this topic is to identify and evaluate methods for solving strongly non linear groundwater flow problems, such as those involving the transport of saline waters of variable density. The Newton-Raphson technique currently employed in NAMMU is generally successful in finding a solution to such problems but the calculation can be very expensive, particularly when a parameter - stepping technique is required to reach the solution of interest /1/. In the fields of optimisation and structural engineering quasi-Newton and modified Newton methods, with and without the use of line searches, have been found to give good results /2-7/. The performance of such techniques was therefore investigated in the context of non-linear groundwater flow. The main test problems concerned examples of saline groundwater flow, ie convection cells driven by density differences. Figure 1 illustrates a typical solution for the concentration field for one of these cases. This problem was solved with three different mesh sizes.

As one would expect, the modified Newton method in which the Jacobian matrix is never updated failed to converge when applied to these problems.

The Broyden and BFGS (Broyden Fletcher, Goldfarb, Shanno) /2,4,7/ methods, both with and without line searches were then implemented. Both of these techniques make a low rank update to the Jacobian matrix.

Contrary to the experience of those working in the fields of optimisation or structural engineering it was found that the Broyden method without line searches was by far the most successful of the methods tried. The BFGS method (which uses a rank two matrix update), both with and without line searches, failed to converge to a solution. By contrast the Broyden method (using a rank one matrix update) gave successful convergence in all cases and, as can be seen from Table I, gave considerable savings in CPU time on the finest mesh. Note that the savings of CPU time increase as the size of the problem increases. Application of the method to a problem of coupled groundwater flow and heat transport showed similar efficiency. The use of the Broyden method in conjunction with parameter stepping is currently under investigation.

Incorporating Chemical Reactions in the NAMMU Code (Task 2)

Prior to the current work the NAMMU code could simulate solute transport in one, two or three dimensions, in either a saturated or unsaturated medium, with retardation by sorption modelled by a linear equilibrium isotherm (K_d) approach. As described in a previous report NAMMU has been modified to allow the sorption of one species to depend on the concentration of a second and also to allow for the modelling of non-linear isotherms (where the concentration sorbed onto the solid is non-linearly related to the concentration in solution). Both of these modifications have now been tested by comparison with the predictions of a simple one-dimensional finite difference code, for simple test problems.

More detailed modelling of the sorption process requires one to take some account of real chemistry. The extensive literature on the modelling of transport with chemical reactions has been examined and it is clear that a useful representation of competitive sorption processes can be obtained from a relatively simple ion exchange model. A ternary heterovalent ion exchange model has therefore been incorporated into NAMMU. This involves the modification of NAMMU to allow the simultaneous solution of three transport equations and the provision of a routine to calculate the sorbed concentrations of the ions, given their aqueous concentration, and the derivatives of the sorbed with respect to the aqueous concentrations. The model is currently being validated against test cases available in the literature. Figure 2 shows a comparison between the results obtained from NAMMU and the results of Valocchi /9,10/. It can be seen that the two are in very good agreement, especially given that the calibration of the flow equation was not carried out to great precision. Further validation of the model and its extension to two and three dimensions (a straightforward, but time-consuming programming task) is currently in progress. A more detailed treatment of the chemistry would require the coupling of NAMMU to a chemical equilibrium code. There is experience at Harwell of coupling the chemical code PHREEQE /11/ to transport processes in one dimension, implemented in the CHEQMATE /12/ code. Further work in this area will investigate possible methods for coupling PHREEQE to NAMMU for far field modelling.

Developing Numerical Methods to simulate the Advance of Sharp Fronts (Task 3)

A sub-contract to investigate this topic was established with Professor P Roe at the College of Aeronautics, Cranfield Institute of Technology, UK. Work at Cranfield started on 22 February 1988 upon the appointment of Dr D Vasilic-Melling as a Senior Research Fellow. After an initial period of familiarisation, various numerical schemes were compared (Lax Wendroff, van Albada, van Leer, Roe's Superbee) /13-15/ as applied to one-dimensional pure advection and advection-diffusion problems. These tests showed Roe's Superbee method to be very promising, giving accurate results without spurious oscillations even in cases with mesh Peclet numbers of 200. More recently a computer code has been written to implement the Superbee method in two dimensions, on a regular mesh. This code has been used to solve a 2D test problem which represents the transport of a concentration 'hump' in a shear flow /16/. The method again gives results in excellent agreement with the known analytical solution.

Theoretical work is also underway at Cranfield on the use of such methods on the unstructured meshes commonly used in NAMMU calculations.

Further development of this area will deal with the application of these techniques on such unstructured meshes and to cases including media with sharply contrasting hydraulic properties.

Methods for the Representation of Solute Dispersion (Task 4)

A sub-contract to investigate this topic was established with the Department of Civil Engineering, University of Newcastle upon Tyne. Work started on 1 February 1988 upon the appointment of Dr R Glendinning as a postdoctoral research associate. In the first four months of the study Dr Glendinning undertook a review of the available literature on stochastic models of solute transport in porous media. This work drew together material from a wide range of disciplines and will be reproduced as a separate report. Following the completion of the literature review the software to be used in the study was tested and revalidated. A pilot study was then carried out to establish the scope of the full Monte-Carlo simulation exercise to be undertaken. On the basis of the pilot study the number of scenarios to be investigated and the number of simulations in each scenario were established. Representations of dispersion in each simulation and the statistical analysis to be performed on the results were also considered. The full Monte-Carlo simulations are now in progress, based upon parameter values appropriate for a sand system. Work is also in hand on the problem of identifying effective hydraulic conductivities which could be used for 'nesting' simulations carried out at different length scales.

Transport in Fracture Networks (Task 5)

In order to account for the geometrical dispersion arising from the many different routes through a network of flow conducting fractures, it is appropriate to model the fractures directly with a stochastic fracture network model such as NAPSAC.

We have assessed the alternative approaches to developing a transport algorithm within the data structure of NAPSAC. We must be able to simulate networks of many thousands of fractures so that our results apply to representative volumes of rock and may be extrapolated to regional porous medium models. In order to model such a complex problem we need a very efficient algorithm. We intend to base our model on a particle following approach. We shall calculate pathlines on each fracture.

If we have a representative number of paths on each fracture plane, then we can track a swarm of particles through the network. Each particle traverses one fracture per step; incrementing distance, time and position, using the most appropriate of the previously calculated paths. The data structure for tracking the particles is available. Our next task is to write the code to calculate the paths, and the relative fluxes to the different possible routes from intersections. A simple pathline algorithm will be written for the regular finite-element meshes for the planar fractures in NAPSAC.

Our approach is particularly appropriate if we should wish to implement a one-dimensional channel model of flow in a fracture plane.

Table I : COMPARISON OF TIME SPENT IN SOLVER FOR NEWTON RAPHSON AND BROYDEN METHODS

Mesh	Number of Unknowns	Solver Time (Newton)*	Solver Time (Broyden)*	T(Broyden) / T(Newton)
5x5	484	1.1×10^7	1.1×10^7	1.0
10x10	1764	9.8×10^7	6.2×10^7	0.63
20x20	6724	7.6×10^8	3.0×10^8	0.39

* Times in microseconds.

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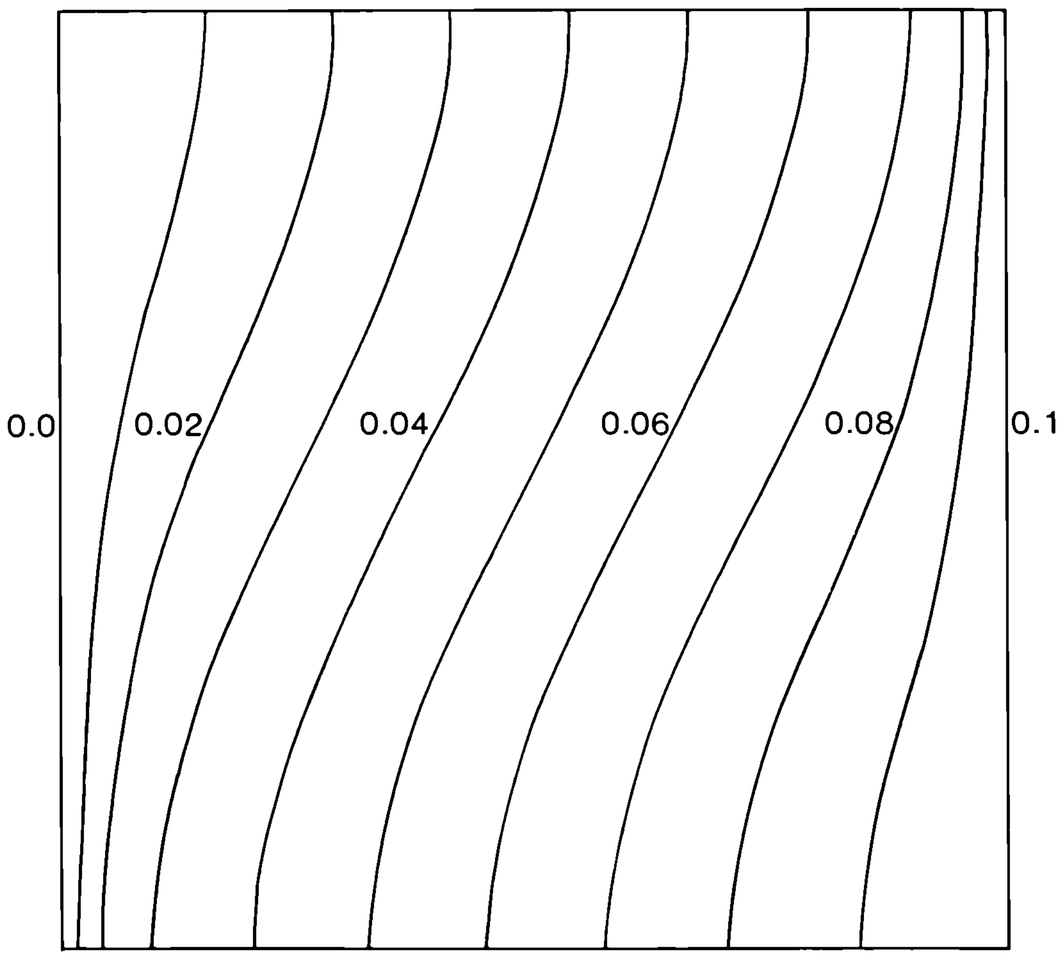


FIG.1. CONCENTRATION CONTOURS FOR SALINE CONVECTION CELL TEST PROBLEM

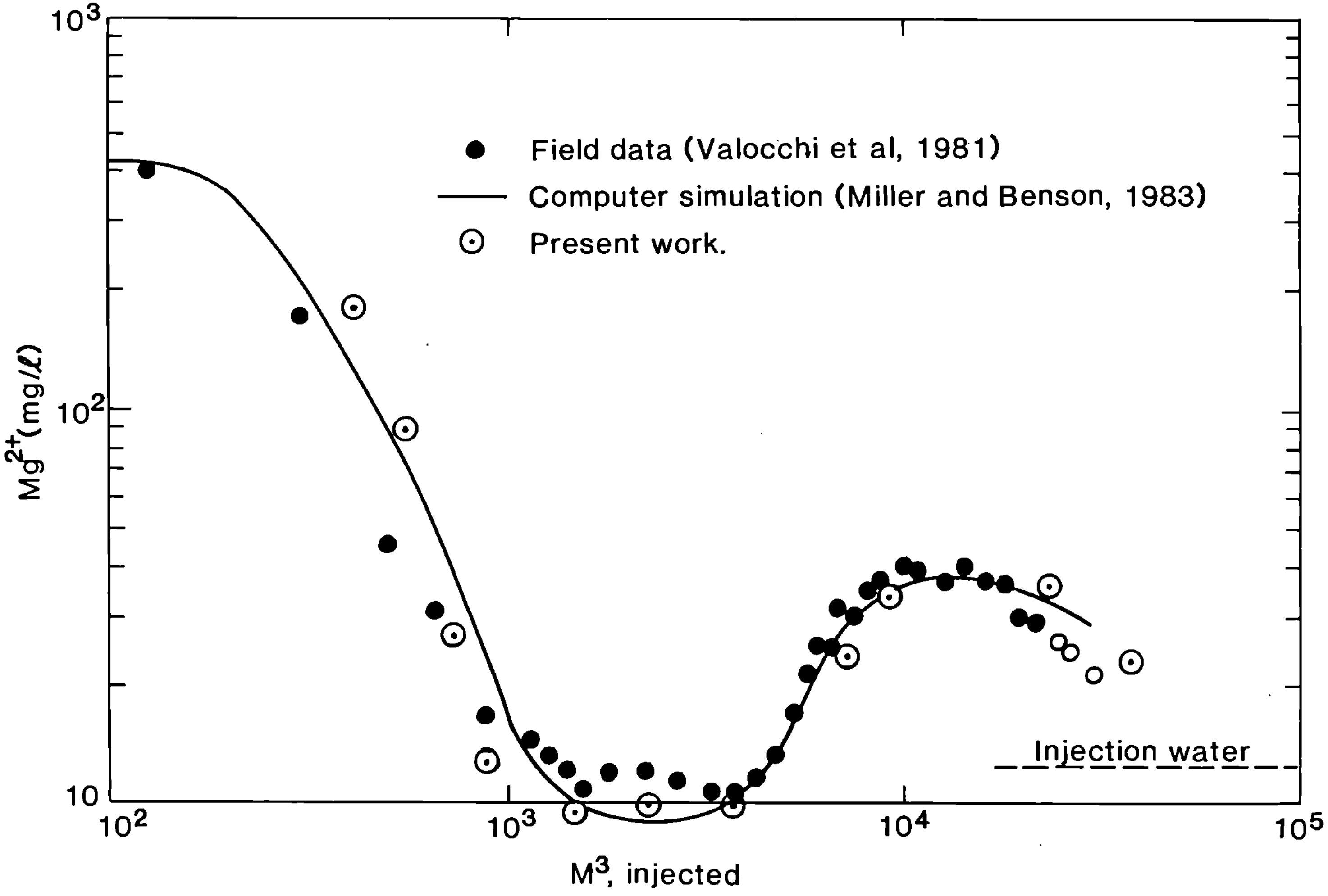


FIG.2. COMPARISON OF FIELD TEST (VALOCCHI ET AL., 1981) WITH NUMERICAL SIMULATION USING CHEMTRN (MILLER AND BENSON, 1983) AND NUMERICAL SIMULATION USING NAMMU, FOR THE EXCHANGING ION Mg^{2+}.

GEOCHEMICAL MODELLING

Contractor : Chem. Dept., Risø National Laboratory, Denmark
Contract No. : FI1W/0079
Duration of contract : August 1986 - December 1989
Period covered : January 1988 - December 1988
Project leader : Bror Skytte Jensen

A. OBJECTIVES AND SCOPE

To develop a versatile and userfriendly program-package for geochemical modelling. The programs are intended for PC's and should have a wide applicability and take advantage of new and recent insight in fundamental processes.

B. WORK PROGRAMME

2.1 To translate the earlier WHATIF-programs into PASCAL. The PC version Turbo Pascal 4.0 has been found convenient.

2.1.1 Simultaneously the limitations of the earlier version are removed to secure the versatility and increased scope for applications.

2.1.2 Algoritms for the calculations on phenomena like multielement ion- exchange and coprecipitations are constructed and added to the main program.

2.2 Experimental work on adsorption phenomena on Calcite are undertaken. The formation of solid solutions 'in' surface layer is considered a possibility.

C. PROGRESS OF WORK AND OBTAINED RESULTS
PROGRESS AND RESULTS

2.1, 2.1.1, 2.1.2

The translation of the speciation program, WHATIFAQ, into PASCAL is finished. The core of the program structure with associated database is working fast and reliable. To secure the userfriendliness of the program, it has been extensively 'menu-terised' both for database maintenance and for chosing calculation modes.

The program handles multielement ion-exchange directly, equilibria with solid phases, a.o., but coprecipitations can only be handled correctly with the mass-transfer program, WHATIF-PW, which soon will be translated into PASCAL.

However, a routine for the handling of precipitation and dissolution reactions by the speciation program is considered at present, and if it works well, then the classical time consuming mass-transfer calculations can be avoided.

Simultaneously a super-structure for handling processes like mixing, evaporation a.o. by directing a sequence of calculations is under consideration. A minor model system has been found to work well.

In **figs. 1, 2** and **3** are shown some examples of the used menu's and an example of output from a calculation.

2.2

The first part of the experimental work followed the long term variations in concentrations when mixtures of solid carbonates were equilibrated with water. During these studies several experimental difficulties associated with CO_2 were encountered, but the measurements disclosed the expected trends.

In a study of the adsorption of Eu on calcite in the presence of varying concentrations of NaCl, an immediate effect indicative of competition for adsorption sites like in ion-exchange was observed. In addition to this effect a very slow adsorption of Eu, which was only slightly influenced by the presence of salt, was observed - in some cases first after several months.

In **fig. 4** are shown some of the experimental results for Eu.

Fig. 1. Example of a DataBaseRecord.

```
                                                        120 records
                            Data Record
  CompCode:  Fe_3H2PO4                  Classific:  Fe_3

   Species:  Comp
                                           Component:       CompFac:
 ReacConst:    24.9500                  [1]: Fe_3       [1]:     1.0

 Enthalpy:     -4.3200                  [2]: PO4        [2]:     1.0

  FactorG:    -16                       [3]: H          [3]:     2.0

   Charge:     2                        [4]:            [4]:     0.0

 ProbForm:     1                        [5]:            [5]:     0.0

    OpVal:     3

      Update        Delete        Next         Previous          Exit
Change fields in this record
```

Fig. 2a. Example of WHATIFAQ input data.

```
                         INPUT: Conditions

  Headline:  WHATIFAQ-TEST                                  Date:  18/01/1989

  Temp Celcius:       25.0

  Fxd pH   Y/N ? ..... N     Fxd pe   Y/N ? ..... Y     Fxd Eh   Y/N ? ... N

  HS/SO4   Y/N ? ..... Y           More than one RedoxPair   Y/N ? ... Y

  Fxd P_CO2   Y/N ? .. N

  IonExchange   Y/N ?   Y

     ←┘-Next Field    ESC-Exit         ↓-Next            ↑-Previous
     ursor Left     →-Cursor Right   ^Y-Delete To EOL   DEL-Delete Char
```

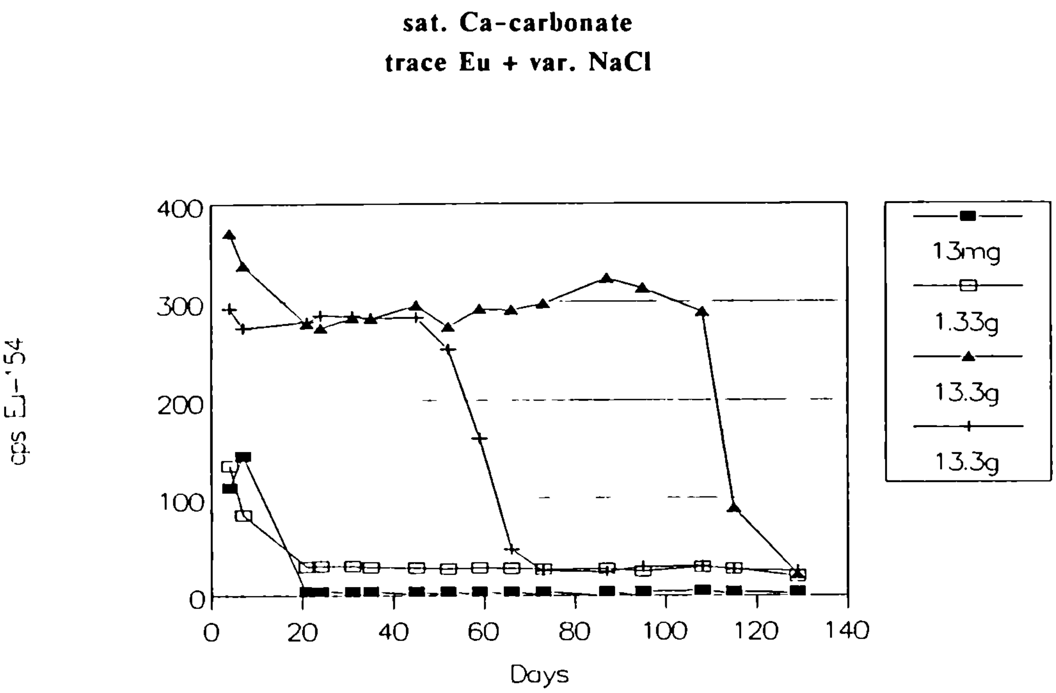
Fig. 2b.

```
                        INPUT: Total Concentrations M/L

 Ftot:      0.002           Natot:    0.232

 Cltot:     0.2             Ktot:     0.02

 SO4tot:    0.03            Mgtot:    0.01

 Stot:      0.0             Catot:    0.01

 CO3tot:    0.01            Altot:    0.0001

 Sitot:     0.0001          Fe2tot:   0.01

 PO4tot:    0.01            Fe3tot:   0.0

     ←┘-Next Field    ESC-Exit         ↓-Next            ↑-Previous
     ursor Left     →-Cursor Right   ^Y-Delete To EOL   DEL-Delete Char
```

Fig. 3. Example of WHATIFAQ output.

```
WHATIFAQ-TEST
-------------

Temperature =  25 Celcius


pe(fxd): 0.00

Ion-Exchanger Capacity = 0.020 Eqv./L


OUTPUT:
-------

pH =  7.7261
pe =  0.0000; Eh =  0.0000 Volts

H2O........  0.000 e..........  0.000 H.......... -7.726 OH......... -6.009

HIonB...... -9.804 NaIonB..... -1.767 KIonB...... -2.566 MgIonB..... -4.624
CaIonB..... -4.330 AlIonB.....-14.707 Fe2IonB.... -4.608 Fe3IonB....  -----

F.......... -2.792 HF......... -7.612 HF2........ -9.824

Cl......... -0.709 HCl........-14.854

SO4........ -1.728 HSO4....... -7.994

S..........  ----- HS.........  ----- H2S........  -----

CO3........ -4.179 H2CO3...... -3.741 HCO3....... -2.101

H4SiO4..... -4.004 H3SiO4..... -6.048 H2SiO4..... -9.759 HSiO4......-13.506
SiO4.......  ----- SiF_6......  -----

PO4........ -6.406 HPO4....... -2.572 H2PO4...... -3.625 H3PO4...... -9.324

Na......... -0.690 NaOH....... -7.164 NaF........ -4.065 NaCl....... -2.562
NaSO4...... -2.244 NaCO3...... -4.855 NaHCO3..... -2.874 NaHPO4..... -3.138

K.......... -1.789 KOH........ -8.562 KCl........ -3.461 KSO4....... -3.193
KHPO4...... -4.601

Mg......... -2.206 MgOH....... -6.163 Mg_4OH_4...  ----- MgF........ -3.724
MgCl....... -3.542 MgSO4...... -2.757 MgCO3...... -7.038 MgHCO3..... -3.884
MgH3SiO4... -7.577 MgH2SiO4... -9.811 MgH2PO4.... -5.217 MgHPO4..... -2.921
MgPO4...... -3.722

Ca......... -2.212 CaOH....... -7.449 CaF........ -4.430 CaCl....... -3.367
CaSO4...... -2.673 CaCO3...... -4.291 CaHCO3..... -3.740 CaH3SiO4... -7.833
CaH2SiO4...-10.897 CaH2PO4.... -4.963 CaHPO4..... -3.102 CaPO4...... -3.738

Al.........-11.684 AlOH....... -9.475 AlOH_2..... -6.322 AlOH_3..... -4.296
AlOH_4..... -4.438 Al_2OH_2...  ----- Al_3OH_4...  ----- AlF........ -8.346
AlF_2...... -6.045 AlF_3...... -5.090 AlF_4...... -5.502 AlF_5...... -6.560
AlF_6...... -8.346 AlSO4......-11.102 AlSO4_2....-12.327

Fe_2....... -2.390 Fe_2OH..... -4.227 Fe_2OH_2... -7.801 Fe_2OH_3...-11.912
Fe_2F...... -4.479 Fe_2Cl..... -3.306 Fe_2Cl_2... -3.788 Fe_2SO4.... -2.971
Fe_2HCO3... -3.918 Fe_2HS_2...  ----- Fe_2HS_3...  ----- Fe_2H2PO4.. -3.838
Fe_2HPO4... -2.419

Fe_3.......-14.742 Fe_3OH..... -9.732 Fe_3OH_2... -5.384 Fe_3OH_3... -5.453
Fe_3OH_4... -5.832 Fe_3_2OH_2.  ----- Fe_3_3OH_4.  ----- Fe_3F......-12.324
Fe_3F_2....-10.822 Fe_3F_3....-10.697 Fe_3Cl.....-14.761 Fe_3Cl_2...  -----
Fe_3Cl_3...  ----- Fe_3Cl_4...  ----- Fe_3SO4....-14.009 Fe_3SO4_2..-14.934
Fe_3H3SiO4.-12.506 Fe_3H2PO4..-13.756 Fe_3HPO4...-13.504


Ionic Strength  =  2.935E-01
```

Fig. 4. Adsorption data for trace amounts of Eu.



The kinetics of adsorption of trace-Eu onto calcite in the presence of variable concentrations of NaCl. An immediate competitive process seems to be followed by a very slow adsorption with a K_D of approximately 10, whose magnitude is practically independent of the presence of neutral salt. The slow adsorption might, until further evidence disproves it, be interpreted as due to the formation of solid solutions of Eu-carbonate in a finite surface layer on the calcite crystals.

GEOCHEMICAL DATABASES

Contractor : Chem. Dept., Risø National Laboratory, Denmark.
Contract No. : FI1W/0080
Duration of contract : August 1986 - December 1989
Period covered : January 1988 - December 1988
Project leader : Bror Skytte Jensen

A. OBJECTIVES AND SCOPE

In the prediction of the extent of migration of pollutants into the environment the confidence to the calculations is primarily dependent on the validity of the model used, i.e. the geochemical calculations and the handling of the convection dispersion phenomena. In geochemical calculations it is usual practice to assume that local equilibria are established. This may well be true in many cases, but that does not automatically mean that overall chemical equilibrium is prevailing in a formation.

Apart from these difficult considerations, the geochemical calculations may be in error or screwed because the thermodynamic data used are incorrect or internally inconsistent.

The present project has the following goal:
-To improve the geochemical databases, such that they may be used to handle a wider range of problems.

B. WORK PROGRAMME

2.1 To develop a database management system which can be used to make available data internally consistent, i.e. to refer to the same set of fundamental standard data.

2.2 To review and compare the most widely used databases.

2.3 To classify minerals according to their 'probability of formation' or other criteria relating to this.

C. PROGRESS OF WORK AND OBTAINED RESULTS

2.1 The Program to MAnage ThermoCHemical Data, **PMATCH**, has been developed by the collaborators, F. J. Pearson, Jr. and John D. Avis from Intera Technologies, and has been tested at Risø. The final report will be presented soon.

The program has several options which are chosen by sets of commands.

Initially the program is in the DATABASE MAINTENANCE MODE, with a series of possible commands which are used when the database is increased, revised etc. In the EDIT- and OUTPUT MODES additional commands are available, among which the UNITS-command present data calculated in chosen units. The units may be Calories or Joules and the CALC-command calculates and fills out missing fields in the data record considered. The available commands are shown in **fig. 1** and an example of the use of the CALC- and UNITS-commands is shown in **fig. 2.**

2.2 This task is mainly done within the CHEMVAL group, although additional data are considered when relevant problems arise. As an example has the need for using data for the formation of soluble, polymeric silicate species been considered. It has been found, that except for special conditions one may safely neglect the formation of these polymers in most geochemical calculations.

2.3 At present prompt precipitations are characterised as simple carbonates, sulphates and amorphous hydroxides. The relevant data are being compiled for introduction into the database. Theoretical considerations on rates of nucleation[1] are considered as a means of making further classifications of crystalline minerals.

References:

1) Nielsen, A.E.: Kinetics of Precipitation.
Pergamon: New York, 1964.

Fig. 1.

AVAILABLE COMMANDS

NAMES
MEMORY
INITDB
OPENDB
DELETE
EDIT
READ
OUTPUT
UNITS
QUIT

SHOW
ALTER
ERASE
CHECK
MEMORY
SAVE
UPDATE
NAMES
ENTER
UNITS
CALC
QUIT

FORMAT
COMMENT
DELFORMAT
OUTDEV
SHOW
BACKUP
NAMES
SCRIPT
QUIT

STOICH
REFERENCE
REAL
CLOSE
WRITE
MEMORY
UNITS
OFFSET

Fig. 2a.

```
Rec # 2              Product Species Data Entry

    NAME                  CAOH+              CHARGE                   1.0
    OPV                     0.0              ALK                      1.0
    DHA                     4.0              DHB                      0.0 *
    GF                      ---              HF                       ---
    SF                      ---              CPAF                     ---
    CPBF                    ---              CPCF                     ---
    VF                      0.0 *CM3         CP25F                    ---
    ALGK                    ---              BLGK                     ---
    CLGK                    ---              DLGK                     ---
    ELGK                    ---              LGK25                 -12.85
    DGR                     ---              DHR              6.08103E+01    kJ
    DSR                     ---              D25CPR                   ---
    DACPR                   ---              DBCPR                    ---
    DCCPR                   ---              DVR                      0.0 *CM3

    STOICH        3     +1.0 CA+2 +1.0 H2O -1.0 H+
    COMMENTS      1     logk25 from Ref 1; dHr from ?
    REFERENCES    1     1: Baes and Mesmer, 1976

ENTER record entry/editing command
```

Fig. 2b.

```
Rec # 2              Product Species Data Entry

    NAME                  CAOH+              CHARGE                   1.0
    OPV                     0.0              ALK                      1.0
    DHA                     4.0              DHB                      0.0 *
    GF             -1.71453E+02 * kC         HF                   -183.52 * kC
    SF             -4.04738E+01 *C/K         CPAF                     ---
    CPBF                    ---              CPCF                     ---
    VF                      0.0 *CM3         CP25F                    ---
    ALGK           -2.19631E+00 *            BLGK                     0.0 *
    CLGK            -3.1764E+03 *            DLGK                     0.0 *
    ELGK                   -0.0 *            LGK25                 -12.85
    DGR             1.75303E+01 * kC         DHR                   14.534   kC
    DSR            -1.00495E+01 *C/K         D25CPR                   0.0 *C/K
    DACPR                   0.0 *            DBCPR                    0.0 *
    DCCPR                   0.0 *            DVR                      0.0 *CM3

    STOICH        3     +1.0 CA+2 +1.0 H2O -1.0 H+
    COMMENTS      1     logk25 from Ref 1; dHr from ?
    REFERENCES    1     1: Baes and Mesmer, 1976

 Thermodynamic data units are Calories. H and G data are KiloCalories
ENTER record entry/editing command
```

Fig. 2c.

```
Rec # 2              Product Species Data Entry

    NAME                  CAOH+              CHARGE                   1.0
    OPV                     0.0              ALK                      1.0
    DHA                     4.0              DHB                      0.0 *
    GF             -7.17358E+02 * kJ         HF              -7.67848E+02 * kJ
    SF             -1.69342E+02 *J/K         CPAF                     ---
    CPBF                    ---              CPCF                     ---
    VF                      0.0 *CM3         CP25F                    ---
    ALGK           -2.19631E+00 *            BLGK                     0.0 *
    CLGK            -3.1764E+03 *            DLGK                     0.0 *
    ELGK                   -0.0 *            LGK25                 -12.85
    DGR             7.33466E+01 * kJ         DHR              6.08103E+01   kJ
    DSR            -4.20471E+01 *J/K         D25CPR                   0.0 *J/K
    DACPR                   0.0 *            DBCPR                    0.0 *
    DCCPR                   0.0 *            DVR                      0.0 *CM3

    STOICH        3     +1.0 CA+2 +1.0 H2O -1.0 H+
    COMMENTS      1     logk25 from Ref 1; dHr from ?
    REFERENCES    1     1: Baes and Mesmer, 1976

 Some MST thermo data missing; Calc type #16
ENTER record entry/editing command
```

MIGRATION OF RADIONUCLIDES BY HIGH-DENSITY BRINES: FINALISATION OF THE METROPOL CODE

Contractor: RIVM, Bilthoven, The Netherlands
Contract No.: FI 1W/0081
Duration of contract: October 1986 - October 1988
Period covered: January 1988 - October 1988
Project leaders: P.Glasbergen, F.Sauter

A. OBJECTIVES AND SCOPE

Transport of radionuclides, possibly released from a salt dome repository to the surrounding aquifers, is a key problem in the safety analysis of waste disposal in salt. For the numerical solution of the set of equations (Darcy's, Fick's law and extensions of these laws for high density brines) which describe the transport, a family of computer codes, called METROPOL, has been developed.

So far METROPOL-1, 2 and 3 have been developed. They deal, respectively, with steady state 3D flow, transient 3D flow and transient 3D flow of fluid with high density differences.

The objective of the present contract is to complete the development of the computer code METROPOL-4 which simulates the transport of low concentration dissolved species, including such processes as adsorption and decay.

In order to make METROPOL easily accessible to other users documentation will have to be written. Also for an easy interpretation of calculated results much effort will have to be put into the development of post-processing facilities.

B. WORK PROGRAMME

1. Inclusion of physico-chemical processes, such as dispersion, diffusion, adsorption, desorption, decay, dissolution/precipitation of salt, chemical reactions, in the METROPOL-code.
2. Inclusion of thermal effects.
3. Code development. New numerical techniques should be studied for the solving of large sets of linear equations as well as for the integration in time (adaptive time integration scheme).
4. Testing and validating the METROPOL-3 code for the case of the flow of high-density brine.
5. Preparing user manuals for METROPOL.
6. Development of post-processing facilities such as particle tracking, contour plotting.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

During the working period covered by this report, the transport code for low concentration species, Metropol-4, has been further tested and enhanced with the possibility of mass flux computations. Metropol-4 is now being used in the international study for the validation of transport codes INTRAVAL /16/ and in the Dutch safety assessment.

For the transport code for high concentration species (density dependent flow), Metropol-3, the effect of new boundary conditions for the interface groundwater - rock salt has been studied (/4/, /6/). Validation of Metropol-3 involves the participation of this code in the international hydrological intercomparison study HYDROCOIN (/12/, /13/) and the above-mentioned INTRAVAL study. Laboratory experiments designed and carried out at RIVM have been used for the purpose of (partial) validation of Metropol. In these experiments the breakthrough curves for a salt solution flowing through a vertical column and displacing low concentration solution are measured. These experiments are being used as a test case for the INTRAVAL study. Metropol-3 was able to successfully simulate these displacement experiments for the case of low and medium-high salt concentrations /9/. For high salt concentration, near saturation, simulation results are not available yet.

The particle tracking program Metropart has been revised in order to avoid problems with velocity fields which are discontinuous across a grid cell boundary. A new time integration scheme has made it possible to accurately approximate trajectories with a strong curvature.

User manuals are available for most parts of the Metropol-package now or will become available in the near future.

Progress and results

1. Inclusion of physico-chemical processes.

In Metropol-4 the transport of low concentration species by groundwater is simulated. The code Metropol-4 includes the processes of advection, diffusion, dispersion, adsorption and decay. The transport of several dissolved species, which are members of a decay-chain can be computed. In the period covered by this report mass balance and mass flux computations were added to Metropol-4. Several tests, comparing Metropol-4 with analytical solutions, have been completed. Metropol-4 is also used in the international study for the validation of transport codes INTRAVAL (/16/), but results from this study are not yet available.

Theoretical work has been done on the boundary conditions for high concentration solute /4/, /6/. A more realistical form for the boundary condition for the interface groundwater-rock salt has been proposed in these references. Numerical simulations with Metropol-3 show that the effect of these new boundary conditions can be significant.

2. Inclusion of thermal effects.

No work has been done on thermal effects.

3. Code development/new numerical techniques.

Metropol-4 has been implemented and tested on a Cray-XMP in Bracknell (UK). Testing of Metropol-3 on a parallel vectorcomputer (Alliant) showed that Metropol performs very well on a parallel computer, due to the specifically suited algorithm used in building the matrix of coefficients in the finite element method.

Research has been done with respect to the adaptation of preconditioning algorithms for large sparse matrices on vector and parallel computers.

4. Testing and validating the Metropol-3 code in the case of the flow of high density brine.

In order to assess the validity of the Metropol-3 code for the simulation of the flow of high density brine, it has been used in the international hydrological model intercomparison project HYDROCOIN (/10/-/13/, /17/-/19/). However, there was severe doubt about the usefulness of a heat experiment (the so-called 'Elder-case', level 2, case 2) for the validation of codes simulating density dependent groundwater flow (/12/). Therefore it was decided to set up laboratory experiments, with the help of the Technical University of Delft in order to directly investigate the transport of brine. In the working period covered, these column experiments, where fresh water or low concentration brine in a porous medium was displaced by high concentration brine, have been continued and results have become available /9/. These experiments are included as a test case in the international study for validation of transport codes INTRAVAL /16/.

Two types of experiments have been done:

(a) a resident salt concentration of 1 kg/m3 was displaced by a salt concentration of 1 kg/m3 was displaced by a salt concentration of 3 kg/m3 ('low concentration')

(b) a resident salt concentration of 1 kg/m3 was displaced by a salt concentration of 100 kg/m3 ('medium-high concentration')

The boundary conditions were: specified pressure on both boundaries, specified salt mass fraction for the inlet and no dispersive salt flux for the outlet.

Both types of experiment have been simulated successfully with Metropol-3; the breakthrough curve in some intermediate point of the column could be approximated with a root mean square error of less than 3%.

After moving the experimental set up from the Technical University of Delft to RIVM in Bilthoven and after recalibration, additional experiments were done:

(c) a resident salt concentration of 2 kg/m3 was displaced by a salt concentration of 4 kg/m3 ('low concentration')

(d) a resident salt concentration of 2 kg/m3 was displaced by a salt concentration of 275 kg/m3 ('high concentration')

The boundary conditions for (c) and (d) were: specified flux for flow and transport equations at the inlet, no dispersive salt flux and specified pressure at the outlet.

No results from Metropol-3 simulations for these experiments are available yet.

5. User manuals for Metropol.

A user manual for Metroplot, the post-processing facility for Metropol which produces plots of mesh, contours and trajectories in 2D cuts through the flow domain, has been completed /5/. A user manual for Metropart, the particle tracking programme, is available in draft version /14/. A user manual for Metropol-4 has been prepared and will be included as an annex to the final report of the current contract /15/.

6. Post-processing.

The post-processing package Metroplot has been adapted to the new plotting standard GKS (Graphical Kernel System). With GKS it is very easy to switch to different plotting devices (e.g. graphical terminal, plotter). Furthermore, it makes Metroplot more easily transferrable to other users. The post processor Metropart, which performs particle tracking, has been completely revised. It was felt necessary to revise the old version, since often problems arose due to discontinuities in the velocity field across grid cell boundaries. In the current version a continuous velocity field is used, which does not involve much 'smearing' of the velocity field. Furthermore, an adaptive time integration method, which uses a combination of first and second order time integration schemes and automatic time step selection, was implemented in the new version. This allows for accurate tracking of trajectories with strong curvature.

<u>List of publications related to Metropol or to Metropol applications, published in 1988</u>

/1/ PRAAGMAN, N., SAUTER, F.J.: Metropol, een softwarepakket voor driedimensionale simulatie van grondwaterstroming, H_2O, 21e jaargang, nr. 3, 4 februari 1988.

/2/ GLASBERGEN, P., HASSANIZADEH, S.M., NOORDIJK, H., SAUTER, F.J.: Geosphere migration studies as support for the comparison of candidate sites for disposal of radioactive waste in rock-salt, Radioactive Waste Management and the Nuclear Fuel Cycle, vol. 10 (1-3), pp. 179-195 (1988).

/3/ HASSANIZADEH, S.M.: Modeling species transport by concentrated brine in aggregated porous media, Transport in Porous Media, 3, pp. 299-318 (1988).

/4/ HASSANIZADEH, S.M., LEIJNSE, A.: On the modeling of brine transport in porous media, Water Resources Research, vol. 24, no. 3, pp. 321-330, March 1988.

/5/ DE VRIES, W.J., SAUTER, F.J.: User's manual Metroplot, RIVM report nr. 728514004, Bilthoven, april 1988.

/6/ LEIJNSE, A.: On the boundary conditions for high concentration solute transport. Contribution to expert meeting on "New developments in groundwater modeling", Delft, 14-16 september 1988.

/7/ HASSANIZADEH, S.M.: On basic equations of density dependent flow and laboratory experiments at RIVM. Contribution to expert meeting on "New developments in groundwater modeling", Delft, 14-16 september 1988.

/8/ LEIJNSE, A., GLASBERGEN, P., SAUTER, F.J., NIJHOFF-PAN, I.: Calculation of groundwater flow and particle tracking for the Gorleben site with Metropol. RIVM report 728516003, Bilthoven, 1988.

/9/ HASSANIZADEH, S.M., LEIJNSE, A., DE VRIES, W.J., GRAY, W.G.: Experimental study of brine transport in porous media, RIVM report 728514005, Bilthoven, 1988.

/10/ SAUTER, F.J., HASSANIZADEH, S.M.: Verification of the Metropol code for groundwaterflow in inhomogeneous porous media. Hydrocoin project, level 1, cases 1 and 2. RIVM report 728528001, Bilthoven, 1988.

/11/ SLOT, A.F.M., NIJHOFF-PAN, I.: Metropol application, simulation of regional groundwaterflow in rocks of low permeability (Piceance Basin, USA), Hydrocoin project, level 2, case 4, RIVM report 728528002, Bilthoven, 1988.

/12/ LEIJNSE, A., HASSANIZADEH, S.M.; Verification of the Metropol code for density dependent flow in porous media, Hydrocoin project, level 2, case 2, RIVM report 728528003, Bilthoven, 1988.

/13/ LEIJNSE, A., HASSANIZADEH, S.M.: Verification of the Metropol code for density dependent flow in porous media, Hydrocoin project, level 1, case 5 and level 3, case 4, Bilthoven, to appear.

/14/ SAUTER, F.J.: User's manual Metropart, RIVM report, Bilthoven, to appear.

/15/ SAUTER, F.J.: User's manual Metropol-4, RIVM report, Bilthoven, to appear.

References

/16/ SKI (Swedish Nuclear Power Inspectorate): INTRAVAL project proposal, SKI 87:3, Stockholm, July 1987.

/17/ SKI (Swedish Nuclear Power Inspectorate), NEA (Nuclear Energy Agency): The international HYDROCOIN project, level 1 report, Paris, 1988.

/18/ SKI (Swedish Nuclear Power Inspectorate), NEA (Nuclear Energy Agency): The international HYDROCOIN project, level 2 report, to appear.

/19/ SKI (Swedish Nuclear Power Inspectorate), NEA (Nuclear Energy Agency): The international HYDROCOIN project, level 3 report, to appear.

STUDY OF THE COUPLED THERMO-HYDROMECHANICAL EFFECTS DUE TO A HLW REPOSITORY IN A GRANITE GEOLOGICAL FORMATION

Contractor : COMMISSARIAT A L'ENERGIE ATOMIQUE
31/33 rue de la Fédération
75752 PARIS
FRANCE

Contrat N° : FI 1 W 0148 - F

Duration of contrat : October 1987 - May 1989

Periode covered : October 1987 - December 1988

Projets leaders : M. DURIN - A. MILLARD - P. OUSTRIERE - JM. PERES

A. OBJECTIVES AND SCOPE

For the evaluation of the performance of radioactive waste geological storage, the description of hydraulic, thermal, chemical and mechanical phenomena is necessary in the near field and in the far field. The difficulty of this description rests, on one hand in the definition of behaviour laws in accordance with time and space that we consider, on the other hand in the connection of phenomena.

The studies already achieved on this subject in the different countries are concerned with either thermo-mechanical effects or thermo-hydraulic effects.

The main purpose of this study is to consider together the thermo-hydro-mechanical effects due to a HLW repository in a granite site. In order to do so two models available now are coupled : CASTEM (thermo-mechanical model) and TRIO (thermo-hydraulic and migration model). Studies of radionuclides migration in the granite rock are conducted with the coupled models in order to evaluate the influence of the different phenomena.

B. WORK PROGRAMME

1 - Thermal calculations

2 - Thermo-mechanical computations

3 - Hydraulic and migration calculations

3.1 - Reference calculation without thermal and mechanical coupling

3.2 - Coupling with thermal effects

3.3 - Full coupling (thermal and mechanical effects)

C. PROGRESS OF WORK AND OBTAINED RESULTS

This study is performed on a reference granite site. The two dimensional calculation is on a 8 km long and a 1.5 km deep domain. The information used are the topography of the ground surface, the estimation of the bedrock boundaries and the hydrological properties.

1 - THERMAL CALCULATIONS

The repository is modelled by an homogeneous heat source of 1 500 x 60 m at - 500 m corresponding to 10^5 tUMI. Isothermals in the vicinity of the waste give a maximum temperature of about 90 °C, temperature remaining above 60 °C until 1 500 years (figure 1).

2 - THERMO-MECHANICAL COMPUTATIONS

The above mentioned temperature fields are used as input in the thermo-mechanical computation. Initial lithostatic stresses, have been assumed. On the lateral boundaries, horizontal displacements have been set to zero.

- The granite has been assumed without individualised cracks, and with a limited traction resistance (10 MPa). The material model used in the calculation is elastic-brittle, with a possibility of strain-softening when the traction resistance is reached. Only orthogonal cracks are allowed. Once cracked, the material can offer some stiffness when compressed, after the closure of the cracks.

The computation has shown a beginning of cracking at both ends of the repository, very early (around 15 years) and then a sudden cracking of the granite located above the wastes (at 27 years). This part of the granite behaves more or less like a beam subjected to a pressure due to the thermal strains. The greatest number of cracks is reached in about to 200 years. Then some cracks close behind the heat front. Figure 2 shows respectively the cracks patterns at 200 years and 1 000 years.

3 - HYDRAULIC AND MIGRATION CALCULATION (FIGURE 3)

This task is subdivided in three steps :

3.a - Reference calculation without thermal and mechanical coupling. We used classical hypothesis ("no flow" boundaries conditions on bottom and lateral limits, head equal to elevation on top boundary) and a permeability function of depth (betwen 10^{-9} m/s at $z = 0$ and 10^{-11} m/s at $z = -1\ 500$ m).

3.b - Coupling with thermal effects ; we used Boussinesq approximation and a permeability taking into account or not the evolution of viscosity with temperature. It is also assumed that the infiltration flow rate in the top boundary is not modified. The evolution of the velocity field as a function of time (figure 3) indicates important near field modifications until 5 000 years. The flow rate crossing the waste area increases quickly and then go back to a value lower than in initial case 3 (figure 4).

Due to the very low velocities, the trajectories of particles are slightly modified by thermal effects, but in some cases a decrease in travel time by a factor of two is obtained.

3.c - Full coupling (thermal and mechanical effects). This part is still in progress.

FIGURE 1 : ISCTHERMALS AS A FUNCTION OF TIME

A = 20.00000 B = 30.00000 C = 40.00000 D = 50.00000 E = 60.00000 F = 70.00000 G = 80.00000

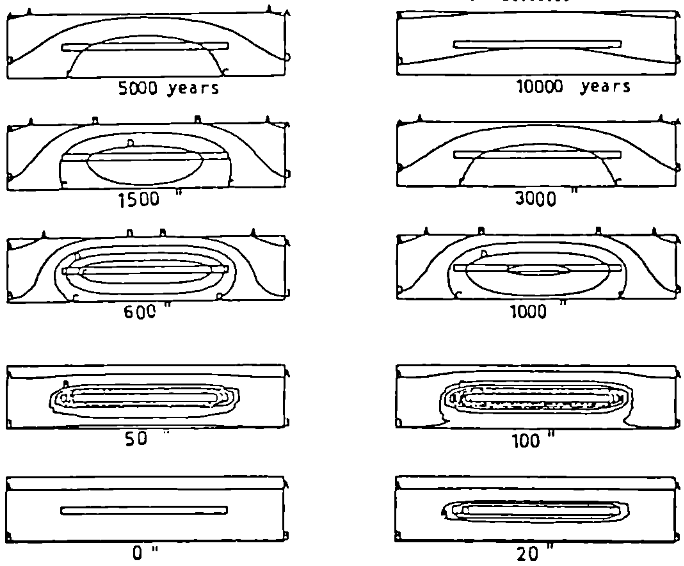


FIGURE 2 : DIRECTION OF FISSURES

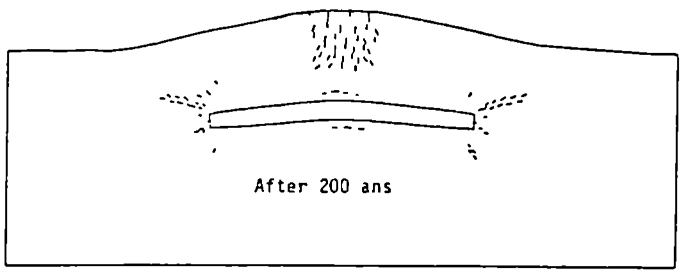


FIGURE 2

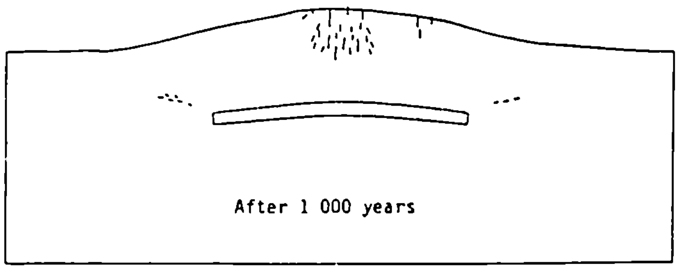
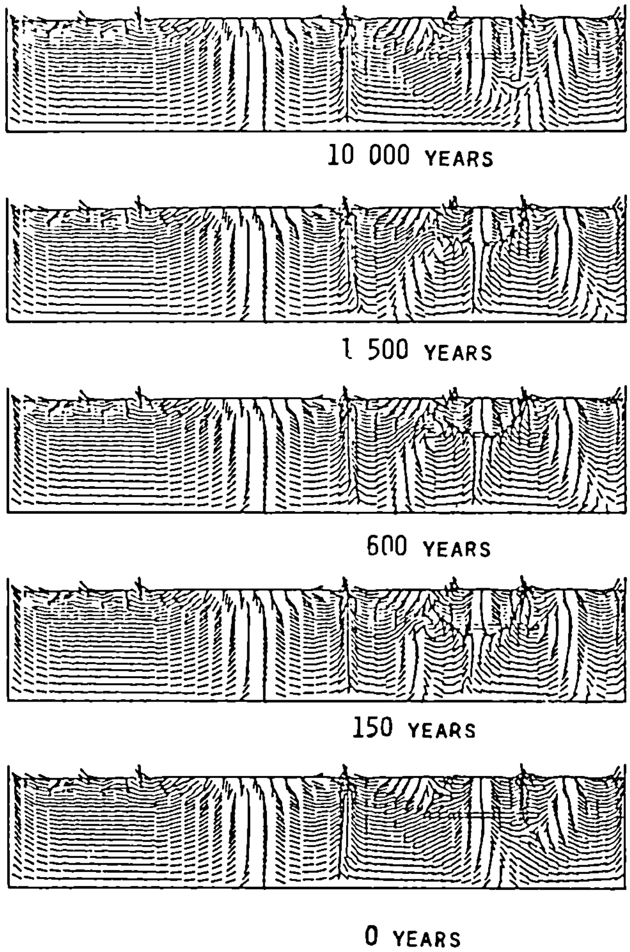


FIGURE 3 : VELOCITY FIELD AS A FUNCTION OF TIME



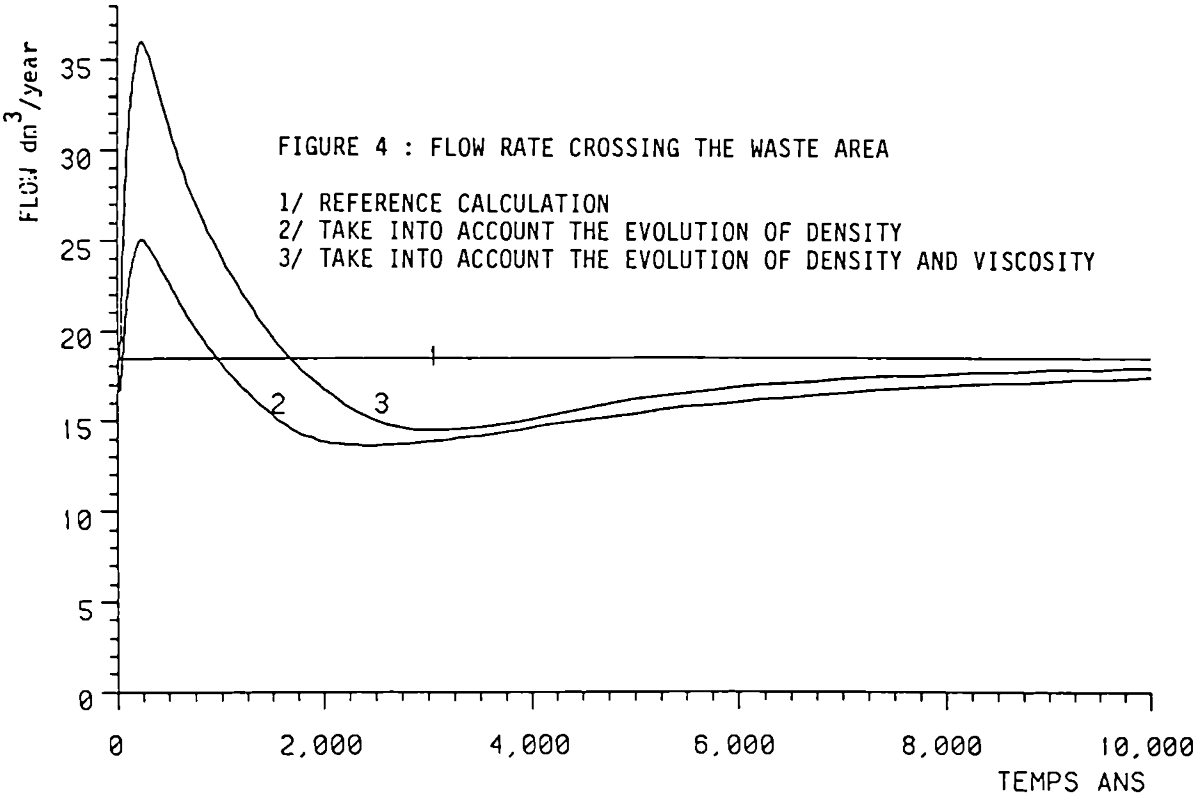


FIGURE 4 : FLOW RATE CROSSING THE WASTE AREA

1/ REFERENCE CALCULATION
2/ TAKE INTO ACCOUNT THE EVOLUTION OF DENSITY
3/ TAKE INTO ACCOUNT THE EVOLUTION OF DENSITY AND VISCOSITY

TRANSFER MECHANISMS OF RADIONUCLIDES IN THE GEOSPHERE

Contractor : Centre d'Etudes Nucléaires de Fontenay-aux-Roses, CEA/IPSN/DAS/SAED - FRANCE

Contract : FI1W/0167

Duration of contract : October 1987 - March 1989

Period covered : January 1988 - December 1988

Project leader : A. CERNES

A. OBJECTIVES AND SCOPE

It is generally agreed that the main processes governing solute transport in the geosphere are : convection, hydrodynamic dispersion, molecular diffusion (Fick's law) and interaction with the rock matrix (adsorption - desorption). In most cases, when mathematical models are designed to take into account only these physical phenomena, or even just a few of them, they will give a true representation of reality.

In the science of non-equilibrium thermodynamics, which describes the evolution of the systems caused by thermodynamic forces, this method does away with non-diagonal couplings of the forces and fluxes. However, this simplification is only valid if one can be sure that the effects of the coupling is negligible. It is probable that the special conditions prevailing in the vicinity of radioactive waste, in particular the high temperature gradients, exclude the use of the simplified approach.

The objective of this study is therefore to investigate the effects of these physical phenomena and to express them mathematically as well as to evaluate the importance of their role in the aforementioned special conditions.

B. WORK PROGRAMME

1 - Theoretical research

This phase makes a review of the principal phenomena likely to play a role in the transfers occuring in natural media and to extract and adapt their mathematical expressions.

2 - Bibliographic study

The bibliography, as extensive as possible, in undertaken on these phenomena, in order mainly to have an idea of the scale of the parameters involved.

3 - Application

The results obtained will be applied to the radionuclides transfer in a geological medium, particularly to a porous medium. Calculations will be made (1D or 2D) to show the relative importance of the different transfers, specially regarding the thermal effect of the disposal.

C. PROGRESS OF WORK AND OBTAINED RESULTS

The two organizations conducting this study are the Paris School of Mines (Centre d'Informatique Géologique and Centre de Physique de l'Irréversibilité) and the Commissariat à l'Energie Atomique (IPSN/DAS).

To begin with, a wide-ranging review was made of already published theories and articles. The formulation of the phenomenological equations of non-equilibrium thermodynamics is particularly complex, at least when one attempts to find an analytical solution to them. The available literature sometimes contains incoherences which make it difficult not only to understand the phenomena but still more to quantify them. Nevertheless, it was possible to extract and adapt the mathematical expressions for the principal phenomena likely to play a non-negligible part in natural media, except for the electrical ones, which are considerably more complex than the others. The phenomena in question are principally : chemical sedimentation, the Soret effect and its corollary the thermogravitational effect.

From a rational point of view it seemed preferable to consider these phenomena separately and in their presumed order of importance. The efforts were therefore primarily concentrated on the thermogravitational effect, which is the cause of impressive overconcentrations in porous media subjected to laboratory constraints. This thermogravitational phenomenon is the result of the combination of the Soret effect and natural convection induced by a horizontal or sloped thermal gradient. The magnitude of the overconcentration depends on the establishment of the appropriate relation between the Soret flux and the velocity of the fluid, and the porous medium is useful as a means of adjusting this velocity. This research into earlier publications yielded a number of studies of the thermogravitational effect in porous media, in particular those by SCHOTT and COSTESEQUE of the laboratory of mineralogy and crystallography at the University of Toulouse. Their work contains valuable reports of experiments that show the magnitude of the phenomenon under high temperature gradients of the type prevailing in the vicinity of radioactive waste repositories. Furthermore, such experiments made with radionuclides, especially uranyle nitrates, show that these compounds are among the most sensitive to the Soret effect.

Since the phenomenology of the thermogravitational effect and the experimental results were known, we concentrated on solving the phenomenon numerically. It then became apparent that the particular formulation of the equations made it possible, at least as far as the above-mentioned experiments were concerned to include the phenomenon in the existing models.

For two different, not too unrealistic cases, the contribution of the "non-dominant" Soret effect to the global transport has been found to be quite substantial, as shown on the figure 1 (first case), 2 and 3 (second case). In consequence, such phenomena seems not to be a priori neglected in a quantitative evaluation of the radiological consequences associated to a nuclear waste repository.

On the other hand, a substantial amount of work has still to be done : the substantial discreparcy found, during validation tests, between the computed results and the experiences performed at Prof. Costesèque's laboratory show that the present modelling of this phenomenon is quite far from being satisfactory. In consequence, also the theoretical and the algorithmical (and, maybe the experimental one too) aspects of this study have to be refined before getting a consistent tool which could be used in the above-mentioned evaluations.

From another hand, we much also pay attention, for the studies of repositories in geological formations, to the electrical phenomena which may occur during the migration. This problem seems to be quite complicate, also for theoretical (i.e. because of the number of phenomena to be studied and consequently, of equations involved) and also for experimental reasons (lack of data).

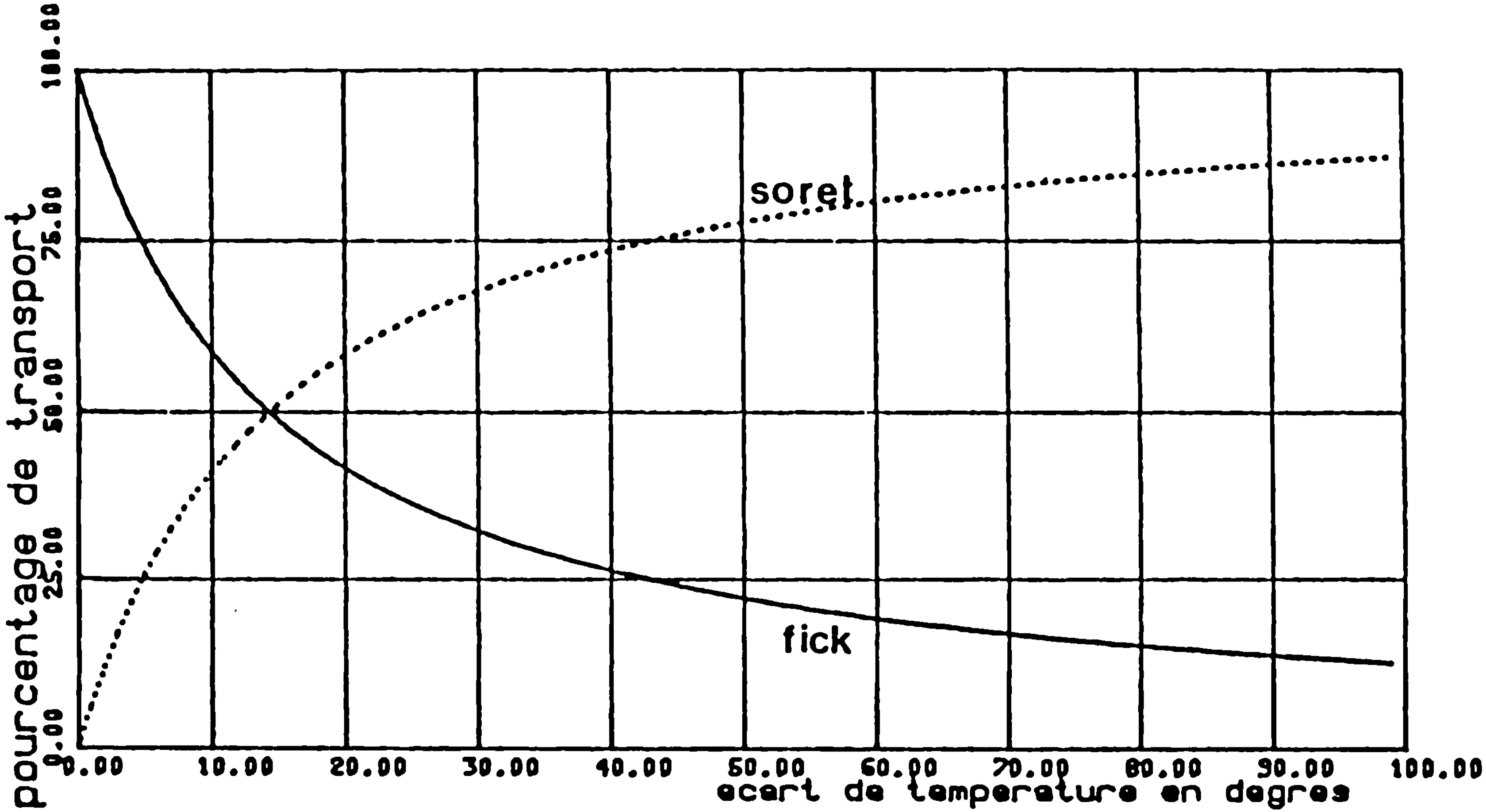


Fig. 1 : Comparizon of the SORET and of the FICK (classical diffusion) effect

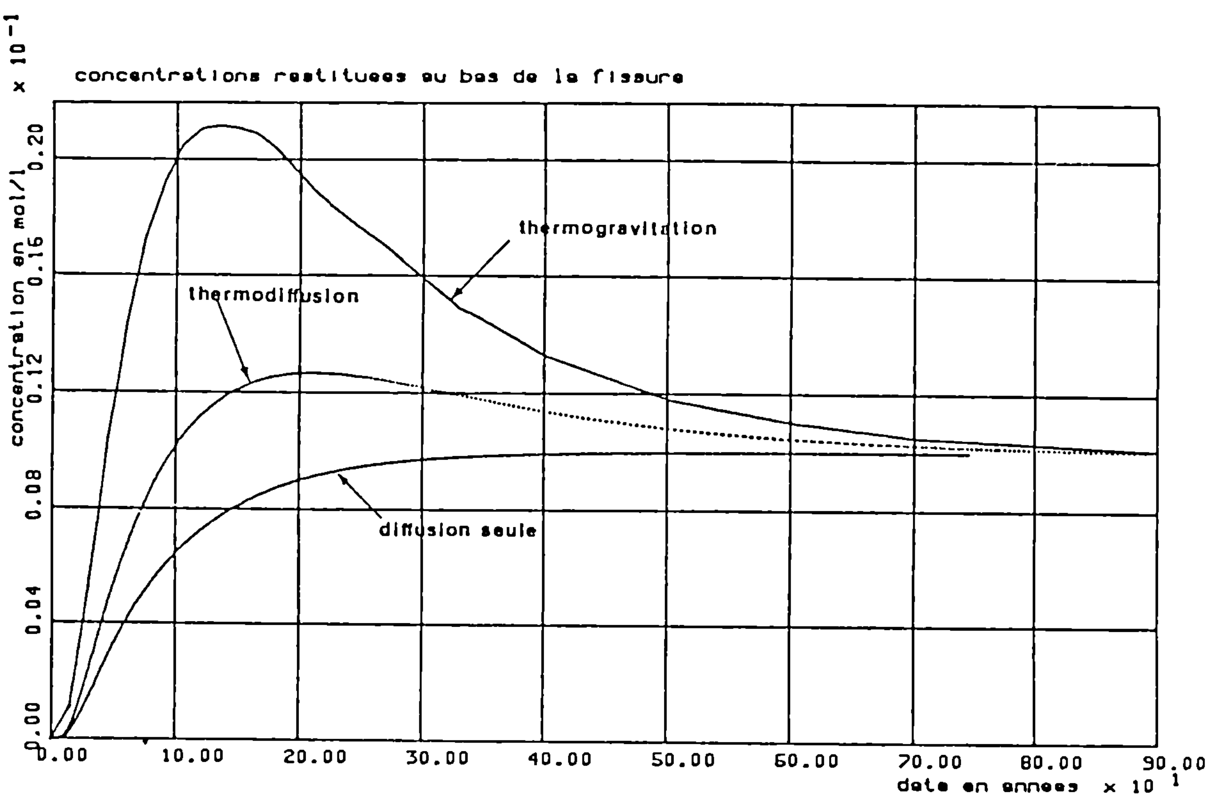


Figure 2

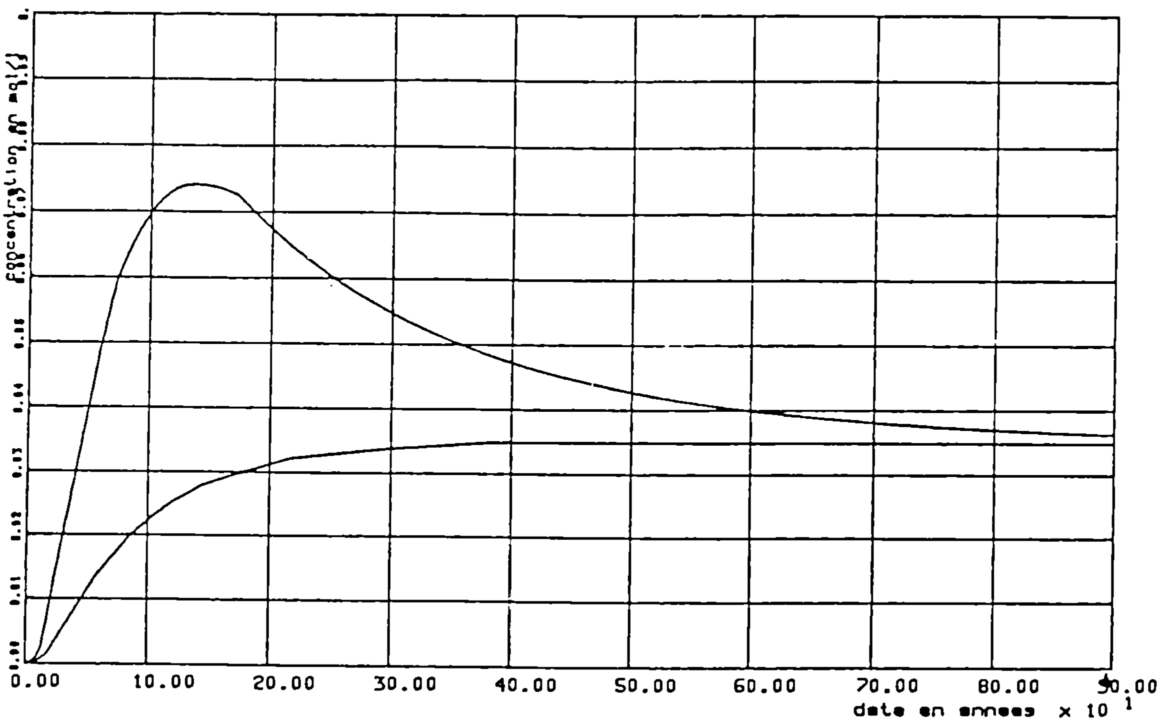


Figure 3 - Flux massiques en zone IV.

DEVELOPMENT OF THE CHEMTARD CODE

Contractor: W.S. Atkins Engineering Sciences, Epsom, UK
Contract No: FIIW/0232
Duration of Contract: October 1988 - December 1989
Period Covered: October 1988 - December 1988
Project Leaders: D Read, S K Liew

A. OBJECTIVES AND SCOPE

In order to evaluate the long-term performance of a geological repository, predictions need to be made of the likely extent and rate of radionuclide transport following contact with groundwater. Several chemical transport simulators have been written for this purpose and during previous development work the Lawrence Berkeley code CHEMTRN was substantially modified and enhanced. The enhanced code is called CHEMTARD (Chemical Transport Adsorption Redox and Decay) and allows the treatment of aqueous complexation, transport by advection and/or diffusion, reversible precipitation-dissolution, radioactive decay and sorption by ion-exchange or surface complexation. It may be used either as a stand- alone coupled transport program, or as part of an integrated radiological assessment methodology.

As with other programs capable of simulating reactive chemical transport, numerical problems may be encountered with CHEMTARD owing to unfavourable combinations of parameter values. Such difficulties are compounded by the long run times required for all but the simplest simulations.

This contract aims to address the above in order that the code enter efficient routine use. The objectives are as follows:-

i) enhancement of numerical methods
ii) development and testing of component models
iii) improvement of code documentation.

B. WORK PROGRAMME

1. The modular structure of CHEMTARD is to be improved and a separate executive routine will be constructed. Means of increasing the efficiency of the existing Newton-Raphson procedure and reasons for convergence failure will be explored.

2. The functional scope of the code will be extended to cover transport by pure diffusion. Current methods of treating oxidation-reduction and mineral precipitation will be improved, the latter to permit supersaturation of solids.

3. Interactive software will be written to facilitate construction of input files together with a graphics package to display CHEMTARD output. Comprehensive documentation will accompany the developed code.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State Of Advancement

During 1988, an outline schedule of tasks has been established and quality assurance procedures have been formulated for the duration of the project. The latter have been reviewed and accepted by the independent UK DoE Software Co-ordinator. Some stages of the development work are well advanced but since the project still has twelve of the total fifteen months to run, none of the items highlighted in Section B has yet been completed. Progress, primarily on task B1, is summarised below.

PROGRESS AND RESULTS

1. Numerical Methods

(i) Modularisation

Although the extant CHEMTARD code comprises twenty separate routines, storage requirements are dominated by the main calling routine as shown in Figure 1. This is regarded as unsatisfactory since the latter not only accesses, either directly or indirectly, all remaining subroutines but also performs some numerical calculations. The aim of the modularisation programme is to reduce dependence on the main routine and produce a more efficient code structure, including the use of a separate executive. Additional subroutines representing alternative or enhanced physical models may then be added more easily.

The intention is to subdivide the code into routines which can be classified according to the headings in the new CHEMTARD function tree, shown schematically in Figure 2. The main calling routine has been reformulated leaving the new subroutines to be written.

(ii) Efficiency and Stability

The efficiency, stability and convergence of a numerical code such as CHEMTARD are inevitably closely linked. These aspects have been investigated via a review of published work, principally in the fields of combustion and chemical physics, "walk-through" deciphering of the code, examination of the way in which component models are formulated and finally stability analyses. Emphasis has been placed on the Newton-Raphson based numerical solver and several techniques have been identified which could enhance the code substantially. These include:

- introduction of relaxation factors to "damp" rapid changes in calculated concentrations

- derivation of a functional relationship between time step and grid spacing, as in the case of an explicit scheme

- increased machine precision

- solution of the transport and mass action equations in two matrices.

The amount of time CHEMTARD spends in each routine for a given set of test cases has been determined and used to direct priorities. Those aspects which affect the numerics of the code are:

- the difference form of the Jacobian matrix
- the difference form of transport, source and sink terms, including those for radioactive decay
- convergence criteria and constraints on the chemical models.

Stability analyses have been carried out using the perturbation method. These demonstrate that the form of the transport equation without chemical coupling is inherently stable. Stability criteria have now been identified for the coupled equations in the absence of radioactive decay and the approach is being extended to address full hydrodynamic/chemical coupling with decay.

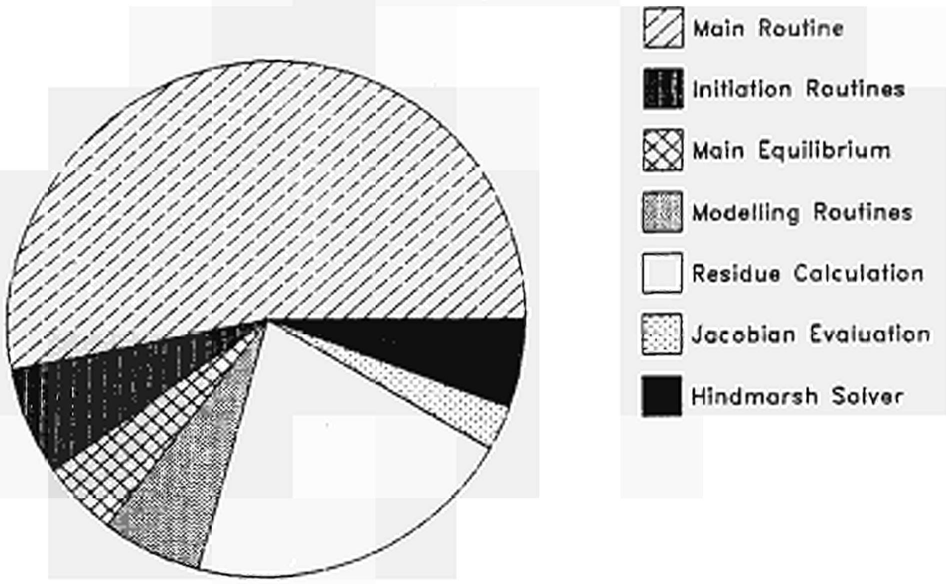
2. Physical and Chemical Models

Relatively little effort has been devoted to this area given the large scale up-grading of component models carried out during the previous phase of the work. To date, only the precipitation routine has been modified, to allow for mineral supersaturation. The accuracy of code changes has been confirmed by verification against the PHREEQE code. Additional testing includes participation in Stage 3 of project CHEMVAL/MIRAGE.

3. Input/Output

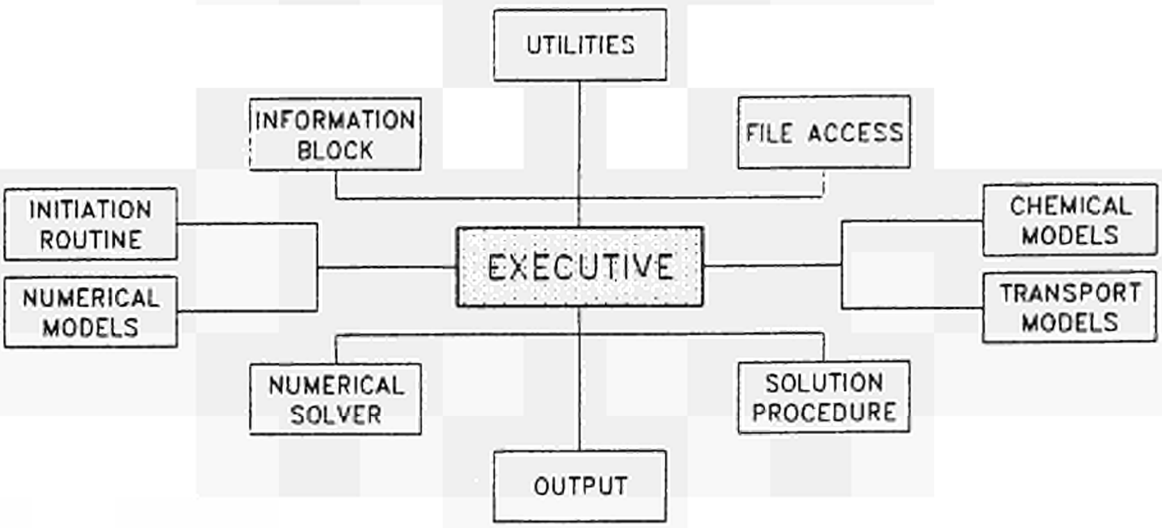
The development of a graphics facility to present CHEMTARD output is well advanced and should aid the user in interpreting results. Work on program input will commence shortly.

FIGURE 1 – CHEMTARD Subroutine Storage Requirements



CHEMTARD Numerical Function Chart

FIGURE 2 – New CHEMTARD Function Tree



4.4. SHALLOW LAND BURIAL

In situ study of the effect of a permanent artificial impervious screen on groundwater flow, in a porous medium, in order to improve the safety of a radioactive shallow disposal

Contractor : Commissariat à l'Energie Atomique, Fontenay-aux-Roses, France
Contract n° : FI.1W/0213/F
Duration of contract : July 1988 - December 1989
Period covered : July 1988 - December 1988
Project leader : J.Cl. GROS

A. OBJECTIVES AND SCOPE

This technique should allow to improve shallow land burial quality by reducing velocities while increasing the groundwater transfer times without interrupting its flow.

Flow velocities and dispersion coefficients are modified by the screen.

Pollution is thus confined downstream in a zone where velocities have been considerably reduced.

The experimental device is made of :

- 1 injection well,
- 18 piezometers in a fan array, down flow from the injection well,
- 5 pumping wells about fourty meters from the injection well.

The groundwater speed will be controlled and a parallel stream flow will be maintained in order to study the transverse dispersion.

A theoretical study has been carried out to determine the flow and the dimensions of the experimental device.

The grout diaphragm wall of cement-bentonit was been realized by SOLETANCHE.

The theoretical study, the "in situ" experiments and the calculations have been executed by the CEA/DERS/SERE of CEN/Cadarache (France).

B. WORK PROGRAMME

1. In situ study of the effect of a permanent impervious screen on groundwater flow.

1.1. Dimensions of the screen.

1.2. Selection of the impervious "setting slurry".

1.3. Character of grout.

2. Hydrodispersive studies of the aquifer ground in disturbed flow by the impervious screen.

2.1. Checking for leaks of the screen and studies of the flows around and downstream of the screen.

2.2. Hydrodispersive studies properly speaking.

2.2.1. Experiments with tracers.

2.2.2. Calculation of longitudinal and transversal dispersion coefficients.

2.2.3. Determination of the geometry of the radioactive confined area downstream of the impervious screen.

2.2.4. Evaluation of the velocity decrease and radioactive concentration decrease downstream of the screen.

C. PROGRESS OF WORK AND OBTAINED RESULTS

Works have begun on July 1st, 1988. The temporary pile-plank screen realised for an anterior study was removed.

It was replaced by a permanent "setting slurry" wall.

This thin vibrated screen is 19.80 m by 14.50 m, and was injected during September 1988.

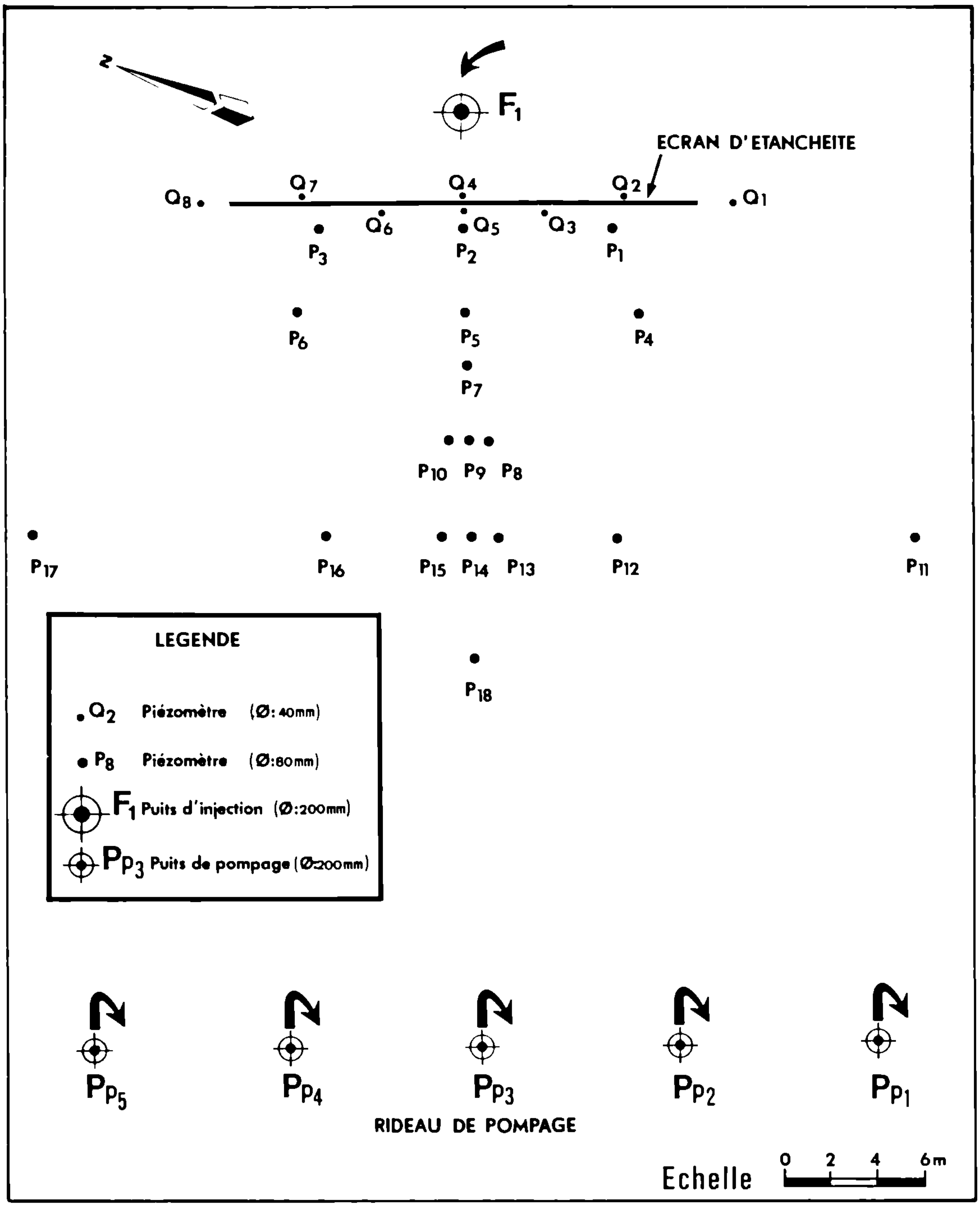
It was a "grout diaphragm wall" of cement-bentonit.

The composition for 1 m^3 of grout is :

- Cement : 200 kg
- Bentonit : 760 l
- Calcareous filler : 450 kg

Permeability K : 2.10^{-8} m/s

The experiments with tracers will begin on January, 1989.



Plan of the experimental device

TITLE: A Large Scale Laboratory Investigation into Gas and Water Permeability of Clay Barriers Exposed to the Environment

Contractor: Taylor Woodrow Construction Limited
Contract No: FI1W/0215
Duration of Contract:
Period Covered: Period to December 1988
Project Leader: Mr T P Lees

A. OBJECTIVES AND SCOPE

The objective of this proposal is to investigate the hydraulic and gaseous permeability of a barrier in the form of a top cap over a shallow land repository to provide essential data for the design and safety case.

B. WORK PROGRAMME

The work programme shall comprise:-

1. Literature survey of recent work relevant to the proposal. Material selection and characterisation.

2. Design and Construction of the test beds. Preparation and compaction of clay samples.

3. Subject test beds to environmental conditioning. Commence periodic sampling and testing programme.

4. Analyse results of testing programme and identify change in physio chemical properties with depth due to environmental conditioning programme.

C. PROGRESS OF WORK AND OBTAINED RESULTS

Contractural Agreement with the UK Department of Environment, who have agreed in principle to provide the balance of funding has been verbally agreed. Written confirmation is expected shortly. No technical progress within the project can be reported.

Discussion with a number of experts has been held to assist in the selection of suitable clay materials for testing. Drawings for the construction of the test beds have been prepared.

OPTIMISATION OF CONCEPTS FOR CONCRETE BARRIERS IN SHALLOW BURIAL FACILIITES

Contractor: ENRESA, MADRID (E)
Contract No: FI1W/0216
Duration of contract: 1 December 1988 - 1 April 1990
Period covered: 1 December - 31 December 1988
Project leader: P. Zuloaga

Progress report not yet available.

TASK No 5

Safety of geological disposal

TASK 5: SAFETY OF GEOLOGICAL DISPOSAL

A. Objective

Assessment of the performance of isolation systems for radioactive waste and of the corresponding radiological impact.

B. Research performed under the 1980-1984 programme

Initiation and implementation of the first phase of the PAGIS (Performance Assessment of Geological Isolation Systems) project, the purpose of which is to assess the capacities offered by the various geological disposal options (salt, clay, granite and marine sediments) for the containment of high level radioactive waste.

C. 1985-1989 Programme

The studies to be conducted concern:

a) Continuation and completion of the PAGIS project, in accordance with the plans and procedure adopted during the previous programme.

b) Initiation and implementation of the PACOMA (Performance Assessment of COnfinements for Medium-Level or Alpha bearing waste project): Safety assessment of disposal systems for alpha contaminated radioactive waste and for medium-level waste buried into geological formations (clay, granite and salt); evaluation of the corresponding radiological impact.

c) Support activities to PAGIS and PACOMA.

5.1 PAGIS Project

Title : Performance evaluation for HLW repositories in granite formations phase 2.

Contractor : Commissariat à l'Energie Atomique
31-33 rue de la Fédération - F 75752 PARIS

Contract n° : 427-84-9-WAS-F

Period covered : from 1.1.1988 to 31.12.1988

Project leaders : J. LEWI (IPSN) and F. ANSELIN (ANDRA)

A - OBJECTIVES AND SCOPE

The second phase of the PAGIS action is devoted to the evaluation of performances of repositories in geological formations for high level vitrified wastes.

For the granite option the approach consisted in :

- on one hand, studying each sub-system (near-field, far-field and biosphere) in detail. The corresponding calculations are performed by ANDRA,

- on the other hand, performing global assessment of radiological consequences with the MELODIE code. The corresponding calculations are performed by IPSN.

B - WORK PROGRAMM 1988

As requested by the Commission, all the calculations have been achieved at the end of 1987.

In consequence the work realized in 1988 has consisted in the preparation of the final comprehensive report, taking into account the CEC observations and the review of the summary prepared for the CEC by ANS.

In addition, the translation of the comprehensive report in English has begun in late 88.

For the deterministic computations (cf. table 1)

1) in all cases, for the "normal scenario", the maximum values of the dose rate are reached after at least 1 million years,

2) these maxima are far below the value of 10^{-4} Sv/year (which is one tenth of the 10^{-3}Sv/year ICRP recommended threshold), with one exception : the terrestrial discharge of the Barfleur site ($1,4.10^{-4}$ Sv/year), where the flow rate of the river in the immediate vicinity of the site (and in consequence the dilution effect) is specially low,

3) the value of the maxima depends on the disposal concept,

4) in addition, for the "britannic site", the effect of the sedimentary cover is quite important : the dose rate varies by one order of magnitude,

5) the most relevant radionuclides to the dose rate are : ^{237}Np, ^{229}Th, ^{99}Tc.

		AURIAT	Barfleur		Britannic site	
			Ruisseau de Barfleur	l'Estran	Without covering	With covering
Time of occurrence of the maximum (y)	Concept A	3.10^{6}	$2,7.10^{6}$	$4,7.10^{6}$	10^{6}	$2,8.10^{6}$
	Concept B	6.10^{6}	$6,5.10^{6}$	$9,9.10^{6}$	$1,7.10^{6}$	$3,4.10^{6}$
Maximum value (Sv/year)	Concept A	3.10^{-5}	$1,4.10^{-4}$	$4,7.10^{-9}$	4.10^{-5}	$2,8.10^{-6}$
	Concept B	$6,4.10^{-6}$	$3,3.10^{-5}$	$1,4.10^{-9}$	2.10^{-5}	$1,4.10^{-6}$

Table 1 : Global dose rate computations for PAGIS

The sensitivity analysis, which was performed for Np^{237} (representative for the actinides) and Cs^{135} (fission products), showed that the most important parameters were :

1) the retardation coefficient in the geosphere,

2) the characteristics of the geosphere : permeability of the deepest zone and dispersivity,

3) the solubility limit and the diffusion coefficient in the engineered barrier (bentonite) of Np^{237} or the retardation coefficient in the bentonite and the bentonite thickness of Cs^{135}.

For the two radionuclides, the uncertainty on the dose rate due to the uncertainty on the parameters is about equal to the mean value of the dose rate (for 200 runs).

2.2. Human intrusion

Without attempting to evaluate the probability of an human intrusion in a granitic formation (which should be rather small, the site beeing chosen in a region without natural resources), the study was aimed at the quantification of its radiological consequences for several cases : intrusion time between 1 000 and 100 000 years and different contamination scenarios. We assumed the intrusion led to the creation of a cubic 100 meter edge cubic cavity in the immediate vicinity of the repository [6].

Using the description of the Auriat site realized for PAGIS, the calculation was performed in three steps :

- calculation of the evolution of the repository until the intrusion time,
- computation of the supposed instantaneous new flow distribution after the intrusion,
- computation of the dose rate, using the mean volumic activity on the walls and the outgoing flow rate.

Three contamination scenarios were considered :

- A : a worker in the mine contamined by external irradiation and inhalation,
- B : an animal drinking in the vicinity during the mining operation,
- C : gardening after the banalization period following the closure of the repository.

With the exception of the worker scenario (the dose rate may reach $1,6.10^{-2}$ Sv/year, which is comparable with the normal exposure in a granite mine), the other dose rate are quite weak (see table 2).

	Sc. A	Sc. B	Sc. C
Maximal dose (Sv/year)	$1,6.10^{-2}$	$2,2.10^{-6}$	$1,1.10^{-3}$
Occurrence time (y)	10^5	3.10^4	3.10^4

Table 2 : Intrusion calculation (Auriat)

Performance Evaluation of HLW Waste Disposal in Geological Formations - PAGIS 2 Project: Sub-Seabed Option

Contractor:	NRPB, Chilton, UK
Contract No.:	WAS-430-84-9-UK
Duration of Contract:	Part 1 of Phase 2 : 1 July 1984 - 31 December 1984 Part 2 of Phase 2 : 1 July 1985 - 31 March 1987
Period covered:	January 1988 - December 1988
Project Leader:	Ms M D Hill

A. Objectives and Scope

The main objective of PAGIS is to evaluate the capacity of various geological formations to act as repositories for vitrified high-level radioactive waste, using common assessment methodologies where possible. This contract is concerned with the sub-seabed disposal option; others are assessing disposal in clay, granite and salt formations on land.

In Phase 1 of PAGIS, which was completed in June 1984, the sites and waste emplacement techniques to be considered were selected. The reference site is at Great Meteor East (GME) and two variant sites were selected - one in the Cape Verde Plateau (CV) and the other in the Southern Nares Abyssal plain (SNAP). All these sites are in the Atlantic Ocean, and have been investigated in international studies of the feasibility of sub-seabed disposal.

The scope of Phase 2 is i) to perform a preliminary radiological assessment using the models and data chosen in Phase 1 ii) to undertake sensitivity and uncertainty analyses iii) to reappraise the data and models and to perform an improved assessment of potential doses and risks to individuals and populations.

B. Work Programme

B.1 Preliminary 'best estimates' of doses and risks to individuals and populations.

B.2 Estimates of the probabilities of occurrence as a function of time of various scenarios.

B.3 Definition and selection of parameters for sensitivity and uncertainty analyses.

B.4 Investigation of the sensitivity of the model results to variations in the values of parameters.

B.5 Quantification of the uncertainty in the model output.

B.6 Improved assessment using the best available models and data which have been selected considering results from B.2 - B.5.

C. Progress of work and obtained results

State of advancement

The work has been completed and the final contract report has been sent to CEC in a form ready for publication. Comments have been made to CEC on the draft summary report for PAGIS Phase 1 and 2 and the final version has been agreed.

Progress and Results

The final contract report consists of a series of 8 topical reports. Final versions of these topical reports have been sent to CEC ready for publication in 1989. The 8 topical reports cover the following subjects:

Topical Report 1	Models and data.
Topical Report 2	Best estimate results for the reference case.
Topical Report 3	Best estimate results for variant cases.
Topical Report 4	Altered evolution scenarios.
Topical Report 5	Global Sensitivity analysis.
Topical Report 6	Uncertainty analysis.
Topical Report 7	Local sensitivity studies.
Topical Report 8	Summary and conclusions.

The first 7 topical reports will be published by CEC as the Comprehensive Report for the subseabed option (EUR 11779) and Topical Report 8 will be published separately as NRPB report number R218/EUR 11483.

NRPB has attended a number of meetings with CEC and the other PAGIS contractors to discuss the layout and content of the summary report of PAGIS phase 1 and 2. This report covers all four HLW disposal options considered in the PAGIS exercise and the final version has been agreed.

SUMMARY AND REVIEW OF PAGIS - PHASE 2

Contractor: Associated Nuclear Services Limited, UK
Contract No.: FI1W/0104-UK (H1)
Duration of contract: September 1986 to March 1988
Period covered: January 1988 to December 1988
Project Leader: G.D. Burholt

A. OBJECTIVES AND SCOPE

The objectives of the work are:

- to prepare a summary of the results and conclusions of the performance assessments of the repositories for HLW in deep geological formations covering four options (clay, granite, salt and the sub-seabed); and
- to review critically the basis, methods, models and data used for the performance assessments and to prepare a report setting out comments and suggestions relevant to further actions of the PAGIS project.

The 'Summary' report is intended for wide distribution and is to be written in relatively simple terms. The 'Comments and suggestions' report is for use by the Commission and the PAGIS Secretaries.

B. WORK PROGRAMME

B.1 Discussions with the nominated representatives of the CEC and agreement of the study basis, including identification of the reference project reports.

B.2 Collation and initial review of the PAGIS phase I, and as they became available, phase II documents and supporting material.

B.3 Discussions with PAGIS Secretaries and other relevant organisations, with the aims of assessing background and clarifying published material.

B.4 Preparation of a draft 'Summary' report of 200 to 250 pages, giving comprehensive coverage of the results and conclusions of the performance assessments for the four geological options.

B.5 Detailed critical review of the approach, the methodologies, models and data, and preparation of a draft 'Comments and suggestions' report.

B.6 Collation with the PAGIS Secretaries and the CEC representatives to produce final versions of the two reports.

B.7 Production of master copies of the final reports.

C. PROGRESS OF WORK AND OBTAINED RESULTS

Summary

Over one hundred documents concerning work carried out in Phase 2 of PAGIS were received and considered by ANS. Due to delays in receiving material from the secretaries and a desire to improve the consistency of presentation, a third draft of the Summary Report (Options section) was prepared for submission to the PAGIS Steering Committee. Further refinements were incorporated into the final fourth draft of the report.

A detailed critical review of PAGIS Phase 2 was prepared with the following agreed objectives:

- to show how far the approaches adopted for each option were appropriate for the objectives of the exercise;
- to verify that no major discrepancies in the assumptions and the data base existed between the evaluations on the four geologic options and to check that a reasonable degree of consistency had been attained;
- to suggest, if appropriate, areas where the similarities of the situations and/or the phenomena could allow improvements in the harmonisation;
- to suggest future complements to the study in order to cover those areas which are felt essential for a sound and exhaustive evaluation of the system performances within the scope of the PAGIS exercise.

Progress and results

1. Preparation of draft Summary Report

Draft 2 of the Summary Report (Option sections only) was prepared and submitted to a meeting of Commission staff and the PAGIS Secretaries at the end of December 1987 and further refined during January 1988, taking account of comments received and new documents received from the Secretaries. Draft 3 (Options section) was submitted to the Commission for inclusion in the draft document submitted to the PAGIS Steering Committee in February 1988. Assistance was also given with editorial improvements to other sections of the draft. As a result of further comments and documents received, the final Draft 4 (Options section) was prepared and submitted to the Commission during April 1988, together with draft Bibliography and Glossary sections.

2. Preparation of Review Report

In view of the delays to the Phase 2 programme and the deferment of Phase 3, a reduced Review of PAGIS Phase 2 was prepared and submitted to the Commission during July 1988. The salient features of each option are identified under the following headings and these features are compared across the options and with the initial objectives of the PAGIS programme:

- inventory assessment;
- radionuclide selection;
- definition and selection of scenarios;
- modelling approach;
- parameter values;
- stochastic modelling;
- radiological impact parameters and criteria;
- presentation;
- overall objectives and organisation;
- suggestions for improvements and further work.

EVALUATION OF THE RESULTS FROM THE PAGIS EXERCICE

Contractor : ONDRAF/NIRAS - Brussels, B
Contract n°: FI1W - 0201 - B (N)
Working period : januari 1988 - december 1988
Project leader : R. Heremans

A. OBJECTIVES AND SCOPE

In order to get the best profit from the PAGIS exercice in view of producing clear conclusions and set out guidelines for possible continuation of the work the Commission ask assistance for :

- examining in detail the various final topic reports of the clay option
- revising the final report of the clay option
- evaluating the most important results of the PAGIS action
- elaborating suggestions on possible futur research works related to performance assessments.

B. WORK PROGRAMME

To review and comment the two following documents.

- A document issued by the British Company : Associated Nuclear Services, (ANS) entitled "PAGIS" - Summary report of phases 1 and 2" - ref. R722-1.
  ANS was the CEC contractor for synthetising the works performed on the four options (clay, salt, granit and sub-seabed) and the results obtained for each of them in the framework of the CEC PAGIS action.
- The document issued by the Belgian Nuclear Research Centre (CEN/SCK) entitled "PAGIS (phase 2) - Disposal in clay formations - final report" - ref. EUR 11776 EN, preprint - CEN/SCK was the CEC contractor for the clay option in the framework of the same PAGIS action.

To formulate general proposals for future work and more particularly for the clay option.

C. PROGRESS AND RESULTS

State of advancement

The two documents were found ready for publication, the first one early July 1988 and the second one in Autumn 1988.
A final report including proposals for future work was transmitted to the CEC Staff.

Progress and results

1. Concerning the ANS document

The chapters A : Introduction, B : Common Basis, C1 : Clay and D : Conclusions were reviewed in detail. Chapters C1: Clay, C2 : Granite, C3 : Salt and C 4 : sub-seabed, were checked for mutual consistency and respective homogeneity in their general presentation.

- For chapter : A - Introduction, and B - Common Basis, more general comments were formulated to improve the text, in order to make it more comprehensible and more fluently readable for non specialists. Figures and tables were also reviewed and suggestions made in the same sense.
- For chapter : C1 - Clay option, more thorough modifications were suggested concerning the link between certain paragraphs and, a better systematic presentation of the results. Written concrete modifications were submitted to the Commission Staff and the authors of the document. These suggestions were afterwards discussed in a meeting during which a general agreement was reached.
- At another meeting with the same previously mentioned partners and the secretaries of the four PAGIS options, the figures and tables to be presented in the final document were discussed and selected.
- The proposed glossary of terms was reviewed and a comparaison with the existing IAEA "Radioactive Waste Management Glossary" second edition (ref.: TECDOC - 447), was also completed. The final glossary was discussed with the Commission Staff.
- A final discussion of the PAGIS action conclusions to be presented in this document took also place with the Commission Staff.

2. Concerning the CEN/SCK document

Written comments and suggestions to improve the text, figures and tables were transmitted to the Commission Staff for the 5 volumes (Basic Data - Methods - Input Data and Preliminary Calculation - Results - Conclusions).

In general it was proposed

- to present a table of contents in front of each chapter in each volume;
- to give a better circumscription of each chapter by
  * briefly introducing each of them
  * improving the titles
- to present more clearly several figures and tables.

As far as the variant sites (Harwell and Val d'Era) are concerned, it was decided to limit the presentation of the results obtained for the Val d'Era site to those giving a preliminary evaluation of the performance of a deep hole matrix.

During the page by page review, minor modifications were proposed in order to improve the text and its readibility. A reshuffling of some subchapters, paragraphs and alineas was also suggested in some cases.

A couple of meetings were especially devoted to the exchange of meanings on the content and the fullness to give to the conclusion part. It was finally agreed to include some comparison of the obtained calculation results with the ICRP criteria and to forward some recommendations for future work.

A final check of the preprint version of the document indicated that most of the suggestions made were taken into account.

3. Proposals formulated for future work.

Three items were identified : data acquisition, model development and validation, subjects to reflect about and to discuss.

3.1. Data aquisition - complementary site investigations, laboratory and in situ investigations were suggested in order to complete and to state more precisely data on the Boom clay in the Mol-Dessel area (e.i. in situ permeability, heat effets, retention of particular elements).

3.2. Model development and validation. This work has to be continued in the framework of a CEC action, examples are the modelling of mass transfer in alternated layers (sand-clay) or actinide retention by organic molecules. As far as validation is concerned a common approach could be developped.

3.3. Subjects to reflect about are : collective dose calculation, cut-off time, worst case scenario, etc...

5.2 PACOMA Project

Title : Performance evaluation of confinement for alpha waste repository in granite formations (Pacoma project).
Contractor : Commissariat à l'Energie Atomique
31-33 rue de la Fédération F75752 PARIS
Contract n° : FI1W/0040-F
Duration : 1/04/87 - 30/06/88
Period covered : from 1/01/88 to 31/12/88
Project leaders : J. LEWI

A - OBJECTIVES AND SCOPE

The general objective is the safety assessment of a deep repository in granitic formations for alpha wastes.
This work has consisted in a first time in the definition of a detailed waste inventory for alpha bearing waste and in the adaptation to the α case of the source-term model CONDIMENT developped for the PAGIS project (vitrified wastes).

B - WORK PROGRAMM

B.1.1 Basic data : a new waste inventory has to be provided. The sites are the same as in Pagis (Auriat, Barfleur, national Uk site). The repository design has to be defined for alpha-bearing wastes.
B.1.2 The methodology will be similar to what was done for the PAGIS project (deterministic, sensitivity and uncertainty analysis).
B.1.3 Model and calculations tools : as far as possible, the same as for PAGIS ; a new version of the source model (CONDIMENT) has to be realized.
B.1.4 Calculations : similar to what has been done for the PAGIS project.

C - PROGRESS OF WORK AND OBTAINED RESULTS

C.1.1 Inventory
A detailed waste inventory has been provided for α cemented wastes and for sludges. A list of selected radionuclides has been established on the basis of long half-lives and high toxicity factors.
C.1.2 The methodology is similar to the one used for the PAGIS project (best-estimate calculations, sensitivity and uncertainty analysis for the normal scenario), but the altered scenario choosen corresponds to a shaft sealing failure. The sites are the same as for the PAGIS project (Auriat and Barfleur in France, notional UK site) but a new repository design has been defined for alpha waste. The alpha waste repository is constituted of a regular system of galleries in which trenchs are realized. In these trenchs, regular cylindrical wells are dug and each borehole can receive a stack of 4 waste cemented drums. For the concept A, bentonite is compacted between the waste cemented drums and the concrete of the trench. For the concept B, concrete is poured between the waste cemented drums and the concrete of the trench, instead of the bentonite.
C.1.3 Models and calculations tools are the same as for the PAGIS project (MELODIE code) but adaptations of the source-term model CONDIMENT has been realized for taking into account the specific chemical behaviour of the concrete matrix. We focused in 1988 on the data collection aspects of the inventory (hulls, sludges, ...) and on preliminary calculations on the Barfleur site. The site represents an exemple of a granitic coastal one and the repository location leads to a release of radioactivity both in a territorial biosphere (La Saire ; ruisseau de Barfleur) and a marine biosphere (Estran). These calculations have been made for the concept A. The geosphere and biosphere data are the same as in PAGIS. The individual maximum dose rate are given in tableau 1 and figure 1 for the fission products Cs 135, Tc 99 and for the actinides of the Np 237 chain.

D - REFERENCES

C. BRUN-YABA, A. CERNES, CEA - CEA/IPSN
Exercice PACOMA - Option granite
Etat d'avancement au 1/12/88 - not published

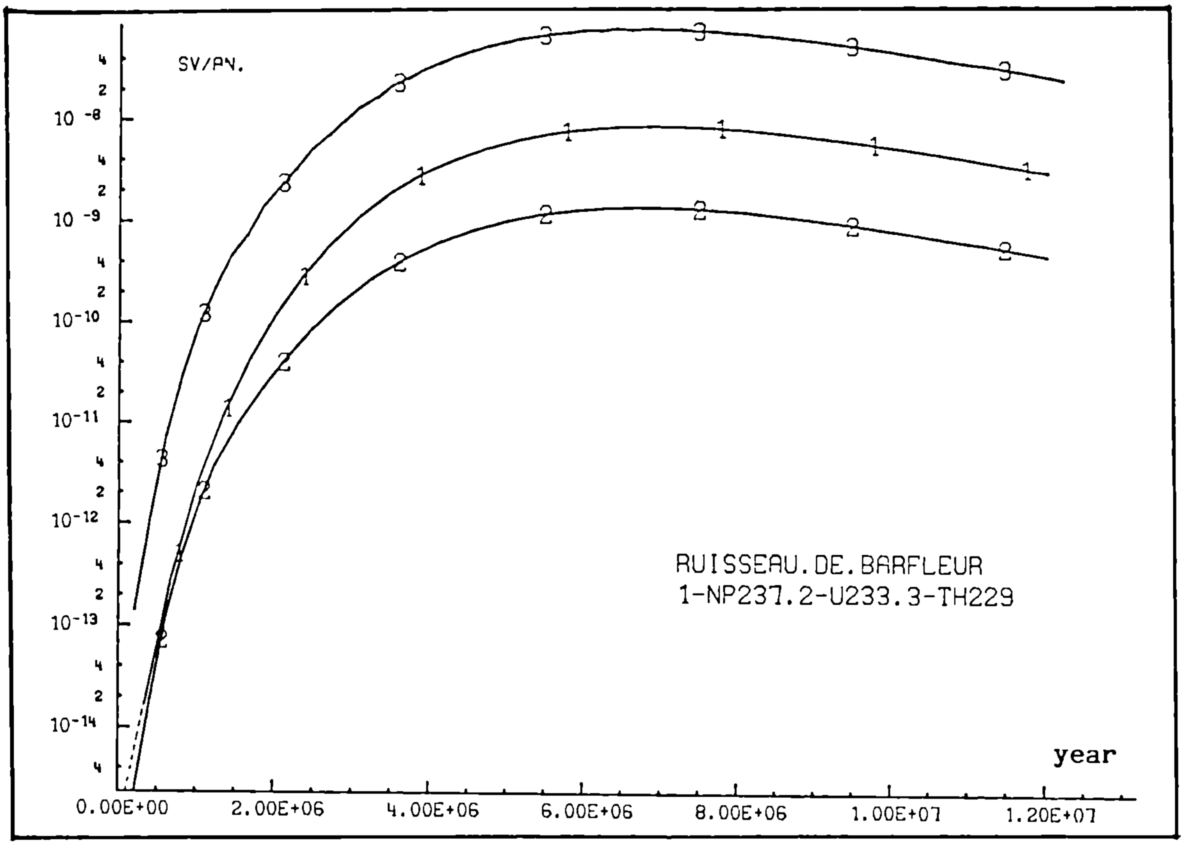


Figure 1.a : Individual dose rate as function of time
Barfleur site - actinides

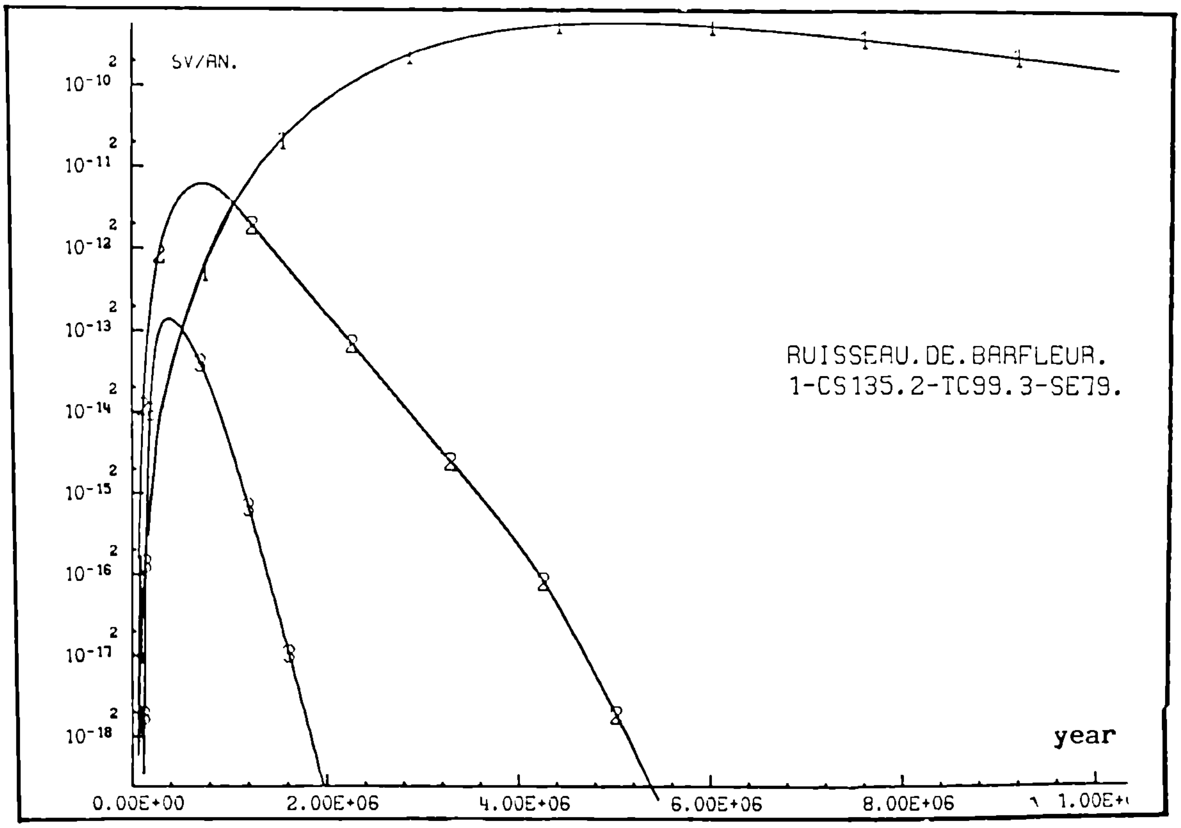


Figure 1.b : individual dose rate as function of time
Barfleur site - Fission products

Radionucléide	La Saire		Le ruisseau de Barfleur		L'Estran	
	Time of occurrence	Dose (Sv/an)	Time of occurrence	Dose (Sv/an)	Time of occurrence	Dose (Sv/an)
Cs 135	9 605 000	$1,4 \times 10^{-14}$	5 020 000	$5,7 \times 10^{-10}$	8 060 000	$1,0 \times 10^{-15}$
Tc 99	1 150 000	$4,8 \times 10^{-16}$	745 000	$6,1 \times 10^{-12}$	947 000	$1,1 \times 10^{-15}$
Np 237	11 800 000	$1,2 \times 10^{-13}$	6 830 000	$9,4 \times 10^{-9}$	10 200 000	$9,3 \times 10^{-13}$
U 233	11 600 000	$2,1 \times 10^{-14}$	6 760 000	$1,4 \times 10^{-9}$	10 300 000	$4,4 \times 10^{-15}$
Th 229	11 600 000	$1,2 \times 10^{-12}$	6 820 000	$8,7 \times 10^{-8}$	10 300 000	$5,2 \times 10^{-12}$

Tableau 1 : Individual maximum dose rate (normal scenario)
Barfleur site

Performance Assessment of Confinements for MLW and Alpha Waste (PACOMA Project)

Contractor: NRPB, Chilton, UK
Contract No.: FI1W/0041-UK(H)
Working Period: October 1986 - March 1989
Period covered: January 1988 - December 1989
Project Leader: Ms M D Hill

A. Objectives and Scope

The overall objectives of the UK contribution to the PACOMA Project are to develop and demonstrate procedures for assessing the radiological impact of disposal of intermediate level waste in a deep repository located in a clay formation. The hypothetical repository considered is assumed to be at Harwell in Oxfordshire. The research is co-ordinated by the UK Department of Environment and is being carried out by four organisations:

National Radiological Protection Board (NRPB)
Theoretical Physics Division, UKAEA Harwell Laboratory
Electrowatt Engineering Ltd
CAP Scientific Ltd

The NRPB work is in two phases. The objectives in Phase I are to establish a detailed methodology for the assessment, to collect data for biosphere modelling and to carry out preliminary calculations. In Phase II the aim is to carry out the assessment, using information provided by other UK contractors and in consultation with other participants in PACOMA, particularly CEN/SCK.

B. Work Programme

B.1. Adaptation of PAGIS methodology for use in the assessment of intermediate level waste disposal, identification of radionuclide release scenarios.
B.2. Review of available biosphere data, and preliminary calculations for typical releases.
B.3. Detailed planning of calculations to be carried out in the full assessment, finalising biosphere data and assumptions for each scenario.
B.4. Best estimates of doses and risks to individuals and populations for each scenario.
B.5. Sensitivity and uncertainty calculations.
B.6. Co-ordination of joint report by the four UK contractors.

C. Progress of work and obtained results

State of Advancement

Items B.1, B.2 and B.3 have been completed and work has started on items B.5 and B.6. Work on item B.4 and B.5 has been delayed awaiting finalisation of the inventory in November 1988. However, the best estimate models and data have been set up and the interface between the models at UKAEA and NRPB has been tested so the calculations can be started as soon as the input is provided. A version of BIOS suitable for uncertainty analysis (MINIBIOS) has been developed and benchmarked against BIOS for a number of release types. The database for the uncertainty analysis has been finalised and the geosphere model has been set up within the uncertainty analysis executive sampling program ESP.

Progress and Results

B.1. Assessment methodology and release scenarios

The PAGIS methodology will be adopted as the basis for the PACOMA study. Hence best estimate calculations of peak individual doses and risks, and of integrated collective doses will be performed for several scenarios. These will be supported by uncertainty and sensitivity analyses of the most probable scenario. The scenarios that will be studied relate to different assumptions for the biosphere. They are the normal scenario, intrusion by drilling an exploratory borehole, intrusion by abstracting water from a well and two future climatic states, a periglacial biosphere and a greenhouse biosphere.

B.2. Review of biosphere data and preliminary calculations

The biosphere data has been kept under continued review and a number of changes have been made since the preliminary calculations performed in 1987. The basic characteristics of the four river sections representing the Thames have been retained but additional data has been collected on the agricultural practices in the Thames Valley area allowing an improved characterisation of the present day biosphere. The data relating to the BIOS soil model has been reviewed and this has resulted in changes to the parameter values used to represent infiltration of rainfall and bioturbation. The preliminary best estimate calculations were repeated with the revised data set and a number of differences were observed. These were mainly related to the area of land irrigated and the changes in bioturbation rate. The importance of the marine pathways were found to be diminished due to revisions to the characteristics of the estuary. Preliminary geosphere calculations by UKAEA showed that the release from the geosphere could be split into several components. These components will be added together by UKAEA to give releases to two distinct areas of the biosphere, the chalk and the clay. BIOS has been modified to accept releases to more than one area of the biosphere simultaneously.

It was decided to restrict the variations in the biosphere data for the two future biospheres to variations in physical transport parameters since it is very difficult to characterise future agricultural practices. The data for a periglacial environment suggests a generally higher rainfall and cooler climate and this has been used to derive values for the river flow rate, irrigation rate and agricultural productivity. The drop in sea level that will accompany the increase in the size of the polar ice caps means that the North Sea will be considerably reduced in size and that the Thames would probably join another river flowing over the exposed part of the bed of the North Sea before reaching the marine environment. Preliminary calculations for the periglacial biosphere were performed for ^{135}Cs, ^{129}I and the ^{237}Np chain and these were compared with the up to date present day biosphere results. The peak individual doses for the periglacial biosphere were less than those for the present day biosphere for all three radionuclides.

Information on the number of wells, the abstraction rates and the purposes for which the water is used have been obtained from the local water authority. These will be used to formulate a well model.

B.3. Planning and finalisation of data and assumptions

The data for the best estimate calculations have been finalised for all scenarios except the greenhouse biosphere. The type of interface to be employed between the UKAEA geosphere model and the NRPB biosphere model has now been finalised, BIOS has been modified to read in flux histories in the agreed form and to use the groundwater velocity vector to determine the water movement in the deep soil boxes and a test flux history generated by UKAEA has been successfully read into BIOS. All the scenarios to be studied will use the normal evolution scenario flux histories and concentrations in the geosphere.

The organisation of the uncertainty and sensitivity analysis has been finalised. It was agreed that it would be more efficient if NRPB ran the entire uncertainty analysis using information provided by the other contractors. The fluxes from the geosphere will be provided to CAP Scientific for their model comparison work in January 1989.

B.5. Sensitivity and uncertainty analyses

A simplified version of BIOS (MINIBIOS) has been developed for use in the uncertainty analyses for PACOMA. This model retains many of the features of BIOS, the main differences being the absence of a pasture soil model, fewer ocean compartments and the omission of transport to land by seaspray since this would not be expected to be important for peak individual doses from an inland site. MINIBIOS has been benchmarked against BIOS for releases to river and deep soil and the two models generally give similar results. The parameters to be sampled have been selected and the data has been finalised. A simple nearfield model has been written which provides a reasonable fit to the best estimate results obtained by Electrowatt. The data for the model has been agreed and finalised. The geosphere model TROUGH-1D has been set up to represent the Harwell site following discussions with UKAEA. This has been interfaced with the nearfield model and set up within the executive sampling program ESP. The sampled geosphere parameters have been selected and the database has been taken from information provided by UKAEA and CAP Scientific. The system will be run using just the nearfield and geosphere models to provide the data required by CAP Scientific. MINIBIOS will then be interfaced into ESP and will use the geosphere fluxes produced by TROUGH-1D as input.

B.6. Co-ordination of joint report by the four UK contractors

An outline of the final joint report has been compiled and issued to all four contractors. Regular meetings have been held to discuss the format and content of the final report.

ACQUISITION OF SUBJECTIVE DATA FOR USE IN MODELS FOR WASTE SITE ASSESSMENTS (PACOMA PROJECT)

Contractor : PRINCIPIA MECHANICA LTD

Contract No : FI1W/0042-UK

Working Period : January 1988 - December 1988

Project Leader : J M Bealby

A. Objectives and Scope

In the modelling of radionuclide movement from a repository, a compromise must be achieved between accuracy and cost. For probabilistic site assessment using Monte Carlo simulation, one dimensional models are used due to their small computational cost whereas for deterministic modelling more detailed two- and three-dimensional models are used. The objectives of this programme of work are to develop and demonstrate consistent data acquisition and preparation techniques for probabilistic site assessment and for detailed deterministic modelling of radionuclide movement from a repository for intermediate level waste under the Harwell site. A comparison of radionuclide risks derived from both assessment procedures will be made and any inconsistencies in the input data and resulting inconsistencies in the risk estimates will be investigated.

B. Work Programme

B.1 Research Programme: development of methodology for comparing probabilistic site assessment codes and detailed deterministic models; planning for the data acquisition exercise.

B.2 Data Acquisition: the use of expert opinion to acquire data for 5 to 10 parameters.

B.3 Model Comparison: comparison of input data and expected risks.

C. Programme of Work and Results Obtained

Summary

The data acquisition exercise has been completed and the results passed onto Harwell and NRPB for use in their 2-D and 1-D modelling work, respectively. Discussions have continued between the UK contractors to establish a feasible modelling comparison approach.

Principia have received most of the results from NRPB and are aware of the form that the Harwell results will take. Work has begun on the model comparison phase.

The contract-end date for Principia, was extended from February 1989 until 30th June 1989.

Progress and Results

B.1 Research Programme

Task B1 is complete and a report on the work has been issued /1/.

B.2 Data Acquisition

Task B2 is complete and a report on the work has been issued /2/.

B.3 Model Comparison

Planning for task B3 has continued. The modelling approach and the methodologies for the comparison have been discussed between the relevant contractors.

This phase of the work requires input from the other UK contractors in the form of their modelling results. The results from the AERE Harwell 2-D detailed modelling will be compared with the results of the 1-D probabilistic approach being used by NRPB. The biosphere models used for the 1-D and 2-D models will be different. In order to simplify the comparison, we intend to compare the nuclide flux from the geosphere. This will avoid an unnecessary complication from using two different biosphere models.

The input data to be used by both parties, was checked to quantify any inconsistencies. The data from our report /2/, was checked against previous studies wherever possible (namely Dry Run 1, /3/. Dry Run 2 /4/ and MOL data /5/).

Principia have received most of the results from the 1-D probabilistic approach used by NRPB, and are expecting all the results from the UK contractors by March 1989. AERE Harwell have specified the format that their modelling results will take. Principia have begun the model comparison work by establishing the analysis to be performed.

References

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/2/ Dalrymple, G.J. and Phillips, L.D. Using a structured approach to the acquisition of probabilistic data from expert opinion. CAP Scientific draft report. 3409/TR.2 July 1987.

/3/ Thompson, B.G.J. (DoE) and Broyd, T.W. (Atkins R&D) 'DRY RUN 1': An initial examination of a procedure for the post-closure radiological risk assessment of an underground disposal facility for radioactive waste. TR-DOE-4 February 1986.

/4/ Gralewski, Z.A. Kane P and Nicholls, D.B. (Electrowatt Engineering Services (UK) Ltd) Development of a methodology for post closure radiological risk analysis of underground waste repositories. Illustrative Assessment of the Harwell site. DOE/RW/87.034 June 1987.

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PERFORMANCE ASSESSMENT STUDIES FOR MEDIUM LEVEL WASTE DISPOSAL AT HARWELL SITE (PACOMA PROJECT)

Contractor: Harwell Laboratory, United Kingdom

Contract No: FI1W/0043 - UK (H)

Duration of Contract: December 1986 - March 1989

Period covered: January 1988 - December 1988

Project leader: Dr K.H. Winters

A. OBJECTIVES AND SCOPE

We shall simulate the transport to the surface of radionuclides released from a hypothetical repository in a clay layer beneath Harwell Laboratory. The rates of release from such a repository, for various waste inventories, have been calculated by Electrowatt Engineering. CAP Scientific have derived probability distributions for the values of parameters involved in the relevant groundwater-flow and solute-transport calculations, by structured questioning of groups of experts. Radiological risks due to the migration of radionuclides from the repository will be calculated by the National Radiological Protection Board. The final stage of the project involves a comparison by CAP Scientific of radiological risks derived by probabilistic risk assessment and from deterministic simulation. This analysis should identify any significant differences in the approaches to safety assessment adopted by UK Nirex Ltd. and by the United Kingdom Department of the Environment.

B. WORK PROGRAMME

1. Review , and if necessary revise, existing calculations of groundwater-flow in the vicinity of the Harwell site, so as to establish numerical accuracy and to examine sensitivity to modelling assumptions.

2. Carry out radionuclide-transport calculations and determine how results depend upon uncertainties in the modelling assumptions and in the values of physical parameters.

3. Assist CAP Scientific Ltd. to compare safety assessments based upon probabilistic and deterministic simulations.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

We have carried out two-dimensional calculations of radionuclide transport from a hypothetical repository located in the clay layer under the Harwell site. The results show that the assumption of transport only along the flow pathline that passes through the repository is invalid. Transport in the clay is predominantly through diffusion, and release of nuclide into the aquifers above and below the clay occurs at much the same time; the concentrations at the interface with the chalk and Corallian tend to values of 2.5×10^{-4} and 5.2×10^{-2} respectively, relative to the source concentration. Radionuclide first emerges at the surface from the chalk directly above the repository, and only later at the clay outcrop where the flow pathline through the repository emerges. The surface flux varies with time in a complex way, but can be represented as a superposition of three one-dimensional transport models.

Progress and Results

The two-dimensional transport calculations described below were obtained for the transport of a single, fictitious nuclide having infinite half-life and negligible sorption. The concentration of the nuclide was assumed to be constant in time. The results so obtained are indicative of the spread of actual nuclides; since the equation representing the transport of a single nuclide is linear then the effects of total repository activity, radioactive decay and sorption can be assessed by appropriately scaling our predictions. Further, our computed quantities can be differentiated with respect to time to give a response function that may be used to predict the effect of an arbitrary time-varying source.

a) Transport in a two layer sub-grid

In view of the substantial cost of simulating the transport of nuclide by the groundwater flow, we performed preliminary calculations on a sub-grid using pressure values interpolated from the grid employed for the flow calculation. We considered the particular sub-grid that covers part of the route taken by the groundwater pathline from the repository, down through the clay and along part of the Corallian aquifer. It was found necessary to use a much finer grid than was required for the flow calculation, to suppress the unphysical spatial oscillations of nuclide concentration that occurred.
The results show that within the clay layer the transport upwards towards the chalk aquifer is as important as that downwards towards the Corallian demonstrating the dominance of diffusion. Thus the assumption of transport along the flow pathline only is invalid.

b) Transport in a three layer sub-grid.

The above results suggest that release of nuclide into both upper and lower aquifers occurs at much the same time. Thus we are led to consider a more complete two-dimensional calculation of the transport over the entire region. To achieve such a simulation with adequate mesh resolution we have improved the efficiency of the time-stepping algorithm in NAMSOL by a factor of 20. The grid used is shown in Figure 1 and illustrates the fine mesh required in the neighbourhood of the repository. Figure 2 shows the superposition of the contour of nuclide concentration at a level of 10^{-7} relative to the source concentration at the repository, at successive times after release from 10^{11} seconds to 5.1×10^{12} seconds in steps of 10^{12}s. Thus it represents the dispersal of the cloud of radionuclide released from the repository. The boundary conditions assume a unit concentration at the repository and zero concentration at the surface. From the figure it is apparent that nuclide first emerges at the surface from the chalk above the repository, not from the clay outcrop where the flow pathline emerges. In addition to the two pathways identified already, the figure reveals a third path that transports nuclide to the remaining surface chalk between the Harwell site and the clay outcrop, by back diffusion from the Corallian aquifer through the central clay layer.

c) Evolution of surface flux of radionuclides

We computed the variation with time of the flux of nuclide transported through the surface of the region shown in Figure 1. The time dependence is complex, but it can be broken down to three distinct contributions, each of which varies with time in a simple way. These contributions are the flux through: the surface clay; the chalk above the repository; and the remaining surface chalk. The temporal variation of the flux through each of these surface zones can be explained by a simple one-dimensional transport model. Thus the behaviour of the total surface flux can be represented by a superposition of three 1-D pathways.

We are assessing the effect on these results of a different boundary condition on the nuclide concentration at the surface. We have replaced the condition of zero surface concentration by one that equates the advective and diffusive surface fluxes. This changes the way in which the nuclide disperses in the chalk, but there remain three distinct pathways for release of the nuclide at the surface.

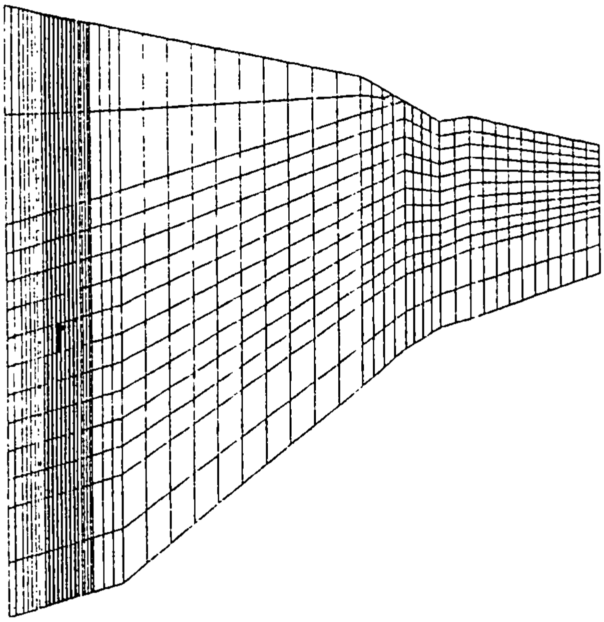

Figure 1
Three layer finite element
subgrid with refinement

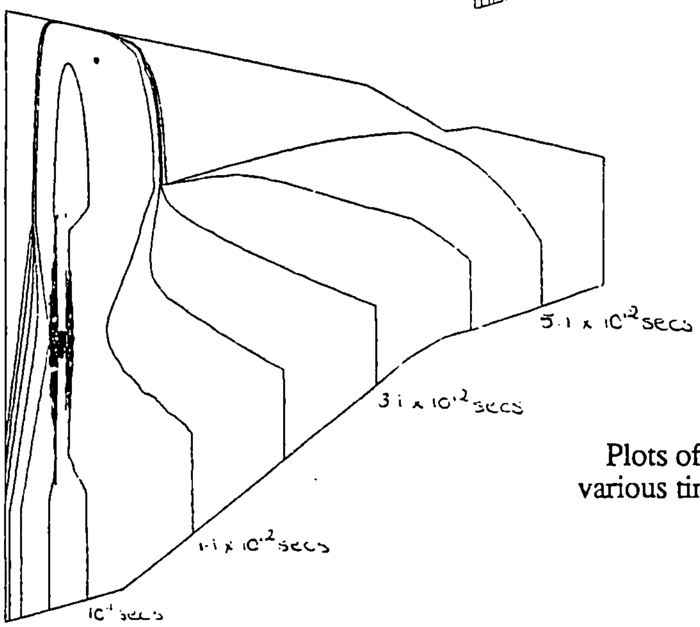

Figure 2
Plots of the 10^{-7} nuclide contour at
various times on the three layer subgrid

Safety Evaluation of Geological Disposal Concepts for Low and Medium Level Wastes in Rock Salt - PACOMA Project

Contractor:	GSF - IFT Braunschweig, FRG
Contract No.:	FI1W/0044-D
Duration of Contract:	April 1987 - December 1988
Period covered:	January 1988 - December 1988
Project leader:	R. Storck

A. OBJECTIVES AND SCOPE

The overal objectives of the GSF contribution to the PACOMA project are to develop and demonstrate procedures for the radiological safety assessment of a deep repository for α-bearing and medium-level radioactive waste in salt rocks.

The research covers the disposal in a repository mined in a salt dome. The reference repository design will be taken as a basis for the calculations. Design variants and alternative disposal concepts are investigated. The results are given in terms of release rates to geosphere and in terms of doses to the individuals and populations.

B. WORK PROGRAMME

B.1. Disposal site
B.2. Repository design
B.3. Scenarios
B.4. Models
B.5. Calculations

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The final repository design is established taking the plans of the national authorities as a guideline and reducing the repository to LLW and MLW disposal locations for the reference case.

The PACOMA reference inventory and the German variant have been obtained. Data for the determination of the source term for the reference repository of the salt option have been collected.

Scenarios will be the same as for PAGIS, i.e. a normal evolution scenario and the altered evolution scenarios accidental brine intrusion and human intrusion.

Progress and results

B.1. Disposal site

In PACOMA only one disposal site is considered which is identical to the reference disposal site of the salt option used in PAGIS. All geological and geochemical data are taken over with no changes, supplemented by sorption data for the additional elements C, Cl, and Ca. For a parameter variation study Kd-values of the Dutch site will be used.

B.2. Repository design

As a guideline for the repository design the repository used for the PSE study is used [1], disregarding the HLW wing. The size of the residual repository then is reduced according to the smaller number of MLW canisters compared to PSE which will be disposed in the PACOMA case.

The design of the reference repository is established and all design data are fixed. Design variants are additional dams, variants due to the different disposal concepts which will be investigated. These alternative concepts are disposal of Iodine filters into boreholes and/or disposal of LLW-α into boreholes. Additionally a repository including boleholes with HLW will be investigated as a variant.

B.3. Scenarios

A normal evolution scenario and two altered evolution scenarios will be investigated. The normal evolution scenario is the diapirism of the salt dome with subrosion at the top of the salt dome. The same model assumptions are used as for PAGIS. The altered evolution scenarios which will be considered are the following two scenarios:

- the combined intrusion scenario with a brine intrusion via the main anhydrite plus intrusion from undetected brine pockets into MLW boreholes,
- the human intrusion scenario, where a storage cavern is leached in a former repository area. The diameter of a cavern is approximately 50 m. Since the distance between MLW boreholes is 15 m (57 m between HLW boreholes in the PAGIS exercise) the canisters of about 4 to 9 boreholes may be laid open.

B.4. Models

New segment models for MLW/LLW chambers and MLW boreholes and the corresponding drift segments have been implemented into the EMOS code and tested during 1988. The same holds for mobilisation models and models for radionuclide transport due to exchange processes [2].

On a meeting of ECN and GSF held in December at ECN, Petten, The Netherlands, the models used for the calculation of doses were compared. The method of calculating the collective doses could not finally be committed.

B.5. Calculation

Calculations of reprocessing waste have been performed to obtain the PACOMA reference inventory. In addition, a German variant inventory has been calculated using 90% Uranium and 10% MOX fuel elements. In both cases the inventory of spent fuel before reprocessing are taken from results of burn-up calculations obtained by KFK using the KORIGEN and ORIGEN codes. A comprehensive list of all the nuclide waste inventories was distributed to all members of the PACOMA steering committee.

Preliminary best estimate calculations have been performed during the first half of 1988. Preliminary results were given in a draft topical report distributed in November 1988.

In order to start with the final calculations for the PACOMA project a meeting was held in June at GSF in Braunschweig, F.R. Germany, for coordinating the performance assessments to be made by ECN and RIVM, The Netherlands, and by GSF on the other hand. The discussion was on sites, Disposal concepts and scenarios to be investigated. Reference case and variants were defined.

End of this year results for the final best estimate calculations are obtained. A topical report covering the reference inventory used for PACOMA, the reference disposal concept, the design of the reference repository and all its design data, a scenario description, the modelling procedures and model data, and result of the best estimate calculation will be distributed in January 1989.

References

[1] Storck, R., et al.: PSE Abschlußbericht, Vol. 16, Projektleitung HMI, Berlin 1985

[2] Hirsekorn, R.P.: In Proceedings of the Workshop on Near Field Assessment of Repositories for Low and Medium Level Waste, OECD Nuclear Energy Agency, Paris 1988

SAFETY EVALUATION OF GEOLOGICAL DISPOSAL CONCEPTS FOR LOW AND MEDIUM LEVEL WASTES IN ROCK SALT (PACOMA-PROJECT)

Contractor	: ECN, Petten, The Netherlands
Contract No.	: FI1W-0045 NL
Duration of contract	: April 1987 - December 1988
Period covered	: January 1988 - December 1988
Project leader	: J. Prij

A. OBJECTIVES AND SCOPE

The research covers the disposal of all types of low and medium level waste in two types of caverns, mined by solution mining in three types of salt formations. The aim of the study is the comparison of the safety of these different concepts. The methodology used is the same as used for PAGIS. The work will be performed in cooperation with GSF in Braunschweig (FRG). A part of the analyses will be executed by the RIVM in Bilthoven (NL).

B. WORK PROGRAMME

1. Basic data

Data will be collected of all types of waste, including the radionuclide inventories and the immobilization models. Also the data defining, the geosphere and biosphere will be collected.

2. Disposal site

Three types of generic formations will be studied; a salt dome, a small pillow and a salt layer.

3. Repository design

Two types of solution mined caverns will be studied. In the first type the waste is dumped in the cavern which is still filled with brine while in the second type the waste is disposed after having removed the brine out of the cavern.

4. Scenaro's

The scenario consists of a water intrusion/extrusion type, while the evaluation is based on the best estimate of individual dosis and the probability of occurence.

5. Models

In principle, the computer codes to be used in PACOMA will be the same as for PAGIS. Where it is appropriate, GSF and ECN will use the same models.

6. Calculations

Best estimate calculations of the dosis to individuals will be performed for the different concepts. The sensitivity of the results will be investigated and a final assessment will be made.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

During this period the analysis of radionuclide transport in rock salt has been performed for the various disposal concepts. The computational results have been described partly and they will be applied to investigate the transport of the radionuclides in the overburden and the biosphere. The flow tubes in the geosphere have been selected and the parameters for the analysis of the nuclide migration are fixed.

Progress and results

The methodology applied to the PACOMA-study consists of a deterministic approach with so-called Best Estimate values for the model parameters and a statistical simulation with parameter values varying according to their probabilities.

To perform Best Estimate calculations for the transport of radionuclides from repositories with low and medium level waste in rock salt, the computer code EMOS (Endlager Model Scenario) has been used.

The analysis concerns the nuclidetransport of both leached caverns as well as mined cavities in three salt formations, i.e., dome, pillow and layer.

With respect to the leached caverns, the cavern sump can be removed before the waste is disposed of. These so-called dry caverns must be stable and closure must not exceed acceptable rates.

For this reason, dry caverns for final disposal of radioactive waste seems to be only feasible in salt domes and pillows.

In case of conventionally mined cavities it is assumed that excavation would be carried out from an accessible underground repository. The estabishment of these excavations assumes thick salt formations encountered in domes and pillows.

A safety assessment of a repository with radioactive waste is strongly determined by the relevant scenarios. The main mechanism for the breaching of the integrity of a repository in a salt formation is the intrusion of ground water or brine through a main anhydrite vein into the pores of the sealings and backfill after final closure of the repository.

The intrusion of a limited amount of brine is conceivable from brine pockets occuring in salt layers. Water or brine will leach radionuclides from the waste and contaminate the brine.

Due to the creep behaviour of rock salt, the contaminated brine in the inundated repository will be squeezed out and may find a release patheway via the main anhydrite. Hence, contaminated brine may reach the aquifers in the overburden and radionuclides will be transported with the groundwater and probably reach the biosphere.

The time dependent calculations carried out with the EMOS-code are related to the time of repository closure.

At the same time the onset of the scenarios is assumed. All calculations are performed with an initial radionuclide inventory (NL) specified in our previous annual report 1987.

The computational results for the disposal concepts under consideration are summarized in table I. It can be observed from table I that the initial state of the leached caverns, i.e., dry or wet, in both salt formation dome and pillow has had a major influence on the computational results.

Due to the initial dry state convergence of the caverns goes faster than in wet situations and results into a shorter duration of the scenario.

The effect of faster convergences, however, has resulted in a larger amount of activity release. In case of a leached cavern in a salt layer, the results in table I show a considerable amount of activity release during a relative short period of time. This result is due to the rather high convergence rate used in the calculations for salt layer.

Further shows table I that the scenario duration of mined cavities in both salt formations dome and pillow are roughly 3×10^4 and 2×10^4 years respectively. In spite of this difference in duration of the scenarios, the amount of radioactive release is in the same order of magnitude.

Table I. Summary of Results of Best Estimate Calculations

	Mined cavities and leached caverns in rock salt						
Salt formation	Dome			Pillow			Layer
Repository design	Leached		Mined	Leached		Mined	Leached
Initial state	dry	wet	dry	dry	wet	dry	wet
Nuclide inventory	NL	NL	NL	NL/3*	NL/3*	NL	NL
Scenario duration [a]	1.407×10^{4}	1.517×10^{4}	3.084×10^{4}	9.933×10^{3}	1.737×10^{4}	1.913×10^{4}	2.716×10^{3}
Activity release [Bq]	2.03×10^{15}	1.88×10^{15}	0.16×10^{15}	$3 \times 0.837 \times 10^{15}$	$3 \times 0.587 \times 10^{15}$	0.18×10^{15}	5.28×10^{15}
Mass release [kg]	1.18×10^{3}	2.75×10^{3}	3.75×10^{3}	$3 \times 0.509 \times 10^{3}$	$3 \times 1.23 \times 10^{3}$	3.41×10^{3}	1.50×10^{3}

*) Since the leached caverns in salt pillow contains one third of the total inventory NL, the computed results are multiplied consequently by a factor of three.

Assessment of radiological consequences and risk associated with the geological disposal of MLW and alpha-waste in clay formations (PACOMA project)

Contractor : SCK/CEN, Mol, Belgium
Contract No. : FI1W/0046-B
Duration of contract : March 1987 - March 1989
Period covered : January 1988 - December 1988
Project leader : A.A. Bonne, J.L. Marivoet

A. OBJECTIVE AND SCOPE

The aim of the PACOMA project is to carry out, as an extension of the PAGIS action on HLW, a comprehensive assessment of the radiological consequences and risks associated with geological disposal of alpha-bearing and medium level wastes. The methodology used in this performance assessment is the one that has been developed during the PAGIS exercise. The main objectives of the study are :

- best estimates of the dose rates and risks to individuals and populations for the normal and relevant altered scenarios ;
- studies concerning the sensitivity of the calculated results to variations of data bases, parameters and models ;
- analysis of the uncertainties.

B. WORK PROGRAMME

B.1. Data collection.
B.2. Adaptations of the computer codes.
B.3. Best estimate calculations.
B.4. Sensitivity study.
B.5. Uncertainty analysis.
B.6. Final assessment.

C. PROGRESS OF THE WORK AND OBTAINED RESULTS

State of advancement

The data collection has been completed. The computer codes developed for the PAGIS exercise have been adapted and new computer codes describing the near-field and the biosphere have been developed and tested.

Sensitivity studies have been elaborated to evaluate which near-field phenomena can contribute significantly to the confinement of the radionuclides in the geosphere. The selection of the radionuclides to be considered in the PACOMA study, is based on simplified pilot calculations of the radionuclide migration through the clay barrier.

Deterministic calculations of individual dose rates have been finished for a water well pathway in the case of the normal evolution scenario. The deterministic calculations for the other pathways and scenarios are in progress.

Progress and results

Simplified calculations of the radionuclide migration through the clay layer have been performed to evaluate which radionuclides can be expected to reach at significant levels the aquifer overlying the host formation. Drinking water dose rates have been estimated for a water well pathway. Two elimination criteria have been applied : a first criterion is based on a concentration limit and the second one on a dose rate limit. The following radionuclides have been selected for further calculations :

- the activation and fission products ; C-14, Cl-36, Ni-59, Se-79, Zr-93, Tc-99, Pd-107, I-129 and Cs-135 ;
- the actinides : U-236, Th-232, Np-237, U-233, Th-229, U-238, U-234, Th-230, Ra-226, U-235, Pa-231.

The influence of some near-field parameters on the radionuclide release rates from the near-field into the geosphere has been examined. The following parameters have been taken into account : the canister life-time, the diffusion coefficient in the waste matrix and in the backfill and the degradation with time of the waste matrix and the backfill. The results obtained show that the diffusion coefficients in the waste matrix and the backfill are influential parameters with respect to the radionuclide fluxes at the near-field clay interface. However none of the near-field parameters influences significantly the radionuclide fluxes at the clay aquifer interface. This last statement illustrates the dominant performance of the geological host formation compared to the engineered barriers in the case of a repository in clay.

The dose rates to a member of the critical group have been calculated deterministically for a water well pathway in the case of the normal evolution scenario. The on-set of the radionuclide release into the aquifer is at about 7,000 years. Contributors to that release are some non-retarded radionuclides present in the medium-level and alpha-bearing waste forms. The calculated dose rates are shown in Fig. 1. The maximum calculated dose rate is about 7E-7 Sv/y and it occurs after about 60,000 years. Other important radionuclides are Se-79 which yields a maximum dose rate of 3E-8 Sv/y after 220,000 years, Tc-99 with a maximum dose rate of 1E-8 Sv/y after 800,000 years and Np-237 with a maximum dose rate of 2e-8 Sv/y after 8 million years. The maximum dose rates due to the actinides from the decay series of U-235 and U-238 occur after about 100 million years ; their maximum total dose rate is about 1E-8 Sv/y.

A comparison of the dose rates calculated for the water well pathway in the PAGIS and PACOMA exercises shows that a noteworth dose due to I-129 occur in the case of the disposal of medium-level and alpha-bearing waste forms. The dose rates calculated for the other radionuclides are about one order of magnitude lower than the corresponding dose rates calculated in the case of high-level waste disposal.

LIST OF PUBLICATIONS

J. MARIVOET and A. BONNE
Performance assessment of geological isolation systems for radioactive waste - Disposal in clay formations
SCK/CEN, Mol, Report R-2748 (1988)
CEC, Brussels, Report EUR 11776 EN (1988)

J. MARIVOET, G. VOLCKAERT and A. BONNE
Performance assessment of a high level waste repository in clay
in W. Lutze (Ed.), Proc. of the 12th Symp. on Scientific Basis for Nuclear Waste Management, Berlin, (1988)

J. MARIVOET and C. VAN BOSSTRAETEN
Probabilistic performance assessment for radioactive waste disposal : a simplified biosphere model
Health Physics, Vol. 55(6), pp. 993-1000 (1988)

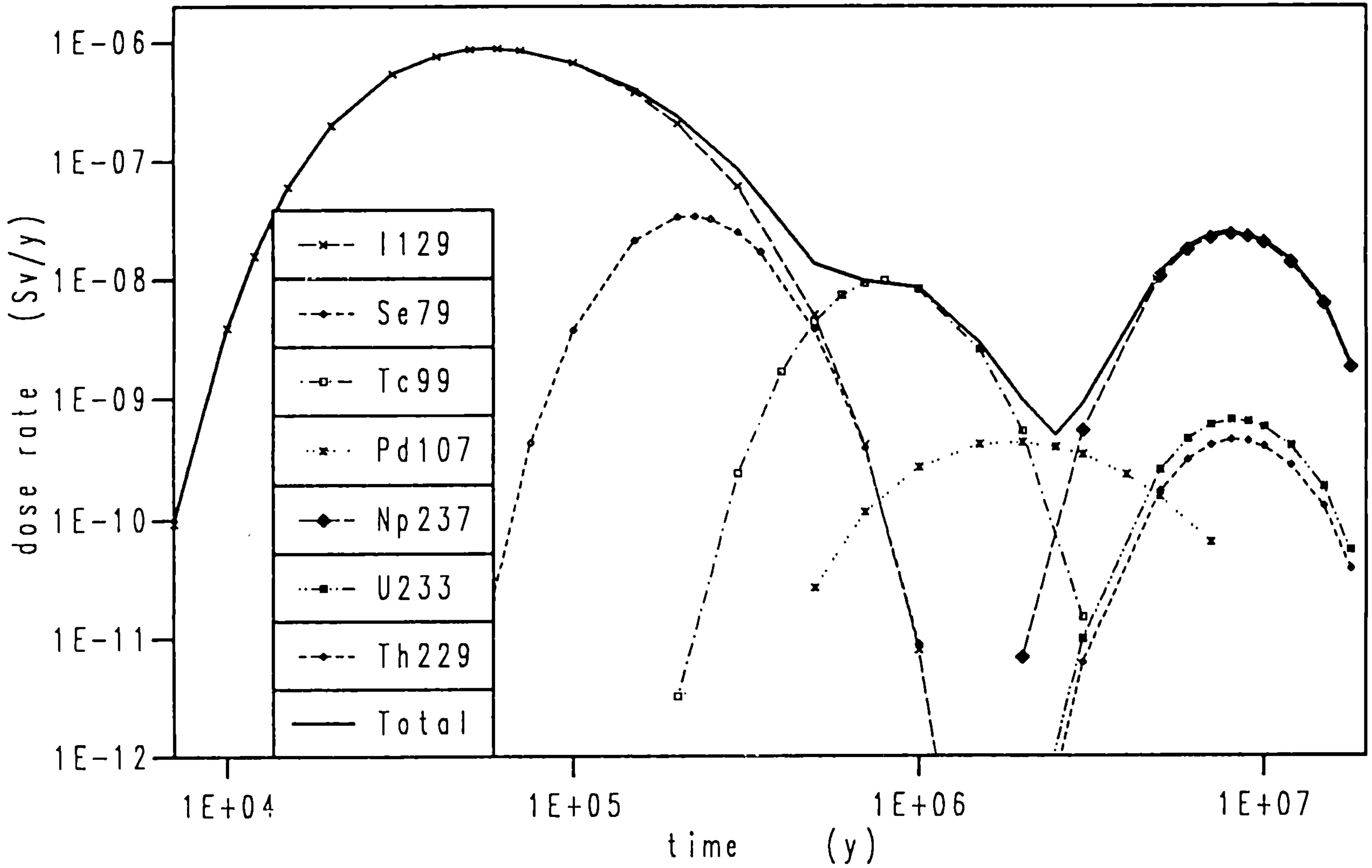

Fig. 1. - Individual dose rates calculated for a water well pathway in the case of the normal evolution scenario

5.3 Support studies

Definition of standards of quality assurance related to the development of disposal facilities for radioactive waste.

Contractor:	Cedar Design Systems Ltd, London, U.K.	
Contract No:	CEC -	FI1W/0039 - UK (H1)
	DoE (UK) -	PECD 7/9/384
	UKAEA -	H2C 618390 T

Duration of contract:	January 1987 - March 1989
Project Leader:	I.E. Hill
Date:	January 1989

A. Objectives and Scope

The research project will produce a definition of an acceptable and appropriate set of procedures for quality assurance, verification, and validation. These procedures would form the basis for establishing uniform standards for risk assessment software, which could be implemented throughout the European radioactive waste disposal programme.

The value and effectiveness of the procedures will be demonstrated by their practical implementation as part of the work of other contractors on the CEC PACOMA projects. Included in this practical implementation will be the introduction of software tools for automating quality assurance procedures, where such tools are available or can be developed.

The work will be subdivided into 4 stages covering respectively, analysis of existing procedures and definition of requirements; a case study of the use of existing procedures in the UK Department of the Environment; definition of procedures; and implementation of procedures.

In order to ensure that the work done is appropriate to the development of risk assessment codes, reference will be made to the other contractors for the CEC PACOMA project.

B. Work Programme

B.1. Review of existing software quality assurance standards and definition of requirements

B.2. Case study of existing UK DoE procedures

B.3. Definition of procedures

B.4. Implementation of procedures

C. <u>Progress of work and obtained results</u>

State of advancement

Work on this project started in January 1987 and is now in its final stages. All 4 tasks have been completed and the final project report is being written.

Progress and results
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1. Review of existing software quality assurnace standards

Interviews were conducted with a total of seven organisations responsible for repository performance assessments. This work has been consolidated by analysis of the requirements for software quality assurance procedures derived from the interviews, and from the case study of UK Doe procedures.

The major areas for improved procedures that were identified by participants in the interviews were:

- The overwhelming requirement was that quality assurance procedures should not slow down the software development process significantly;
- Documentation, particularly at the program structure level is a general problem. The time scales for waste disposal are sufficiently long (up to 20 years) for considerable changes in personnel to occur - this greatly increases the need for documented code and for documented quality assurance procedures. Aids for production of documentation were identified as important;
- Some organisations are interested in introducing quality assurance procedures as a way of ensuring code is more accessible to all members the group, and to remove the personal possessiveness associated with individuals code. This was not a general requirement of all groups;
- Related to the above is a need for a way of making general purpose subroutines more widely available within the group;
- There is a need for guidance on the extent to which verification and validation of a program transfer when the program is used on other computers, and by other groups;
- Any pressure that exists for quality assurance procedures comes from outside; the groups responsible for safety assessments are self-regulating;
- Most groups have standards (of varying degrees of formality) at the level of programming practice, but design or specification standards are not used. (There seems to be a need for confirmation that the coding standards used are appropriate);
- The need for configuration management procedures is generally recognised, but most groups only have procedures for control of the executable code for versions of the programs used for assessment purposes.

This work has been reported in a draft report [1] that will eventually become part of the final report for the project.

2. UK DoE case study

The case study of existing UK DoE procedures has taken the form of participation in the review stages of the VANDAL development project, as well reviews of programming level documents produced by various contractors for the full DoE risk assessment programme. This work has provided many useful examples of the sort of procedures that are suitable for development of performance assessment codes, and of the difficulties of introducing these procedures. This work has not been reported separately, but will be included in the final project report.

3. Definition of procedures

The actions needed to introduce more rigorous software quality assurance procedures have been described in [1] together with an outline description of the sort of procedures that would be appropriate. The actions required are:

- statement of the policy for achieving quality in code development and use. This should be documented at three levels, namely a quality manual covering management aspects, codes of practice for specific software development activities, and quality plans for each project;
- definition of an orderly sequence of events for development and use of software, ie a software lifecycle model, as an essential first step in introducing more formal procedures;
- choice of procedures to be followed for each of the lifecycle stages.

Procedures have been defined by describing the principles of the methods to be used; giving examples of the records or documentation to be used; and providing references to published sources of information. The topics covered are:

- project management;
- software development, including analysis and specification, design, coding, and testing;
- documentation;
- configuration management;
- verification and validation;
- use of software for performance assessments.

The procedures recommended will be described in a series of appendices to the final report.

4. Implementation

As a test of the feasibility of the procedures to be included in the recommendations, an outline of the procedures was discussed with the NRPB. They have introduced a version of the procedures for use on a single project, with only the minimum of input from us. This exercise has provided useful feedback to us on the sorts of procedures that can be introduced without making radical changes in existing working practices.

Part of the work specified for this stage of the project was an evaluation of software tools that would aid in introducing software quality assurance. Interim reports on design and analysis aids [2], and on aids for configuration management[3] have been produced, and will be incorporated in the final project report.

List of publications

[1] HILL, I.E., Requirements for software quality assurance procedures for radioactive waste risk assessment codes. (Jan 1988).

[2] HILL, I.E., A preliminary review of Analysis and Design tools as aids to improving software quality. (June 1987)

[3] MAYER, J., A preliminary review of configuration management tools as aids to improving software quality. (Feb 1988)

MODELLING THE LONG TERM EVOLUTION OF GEOLOGICAL RADWASTE DISPOSAL FACILITIES

Contractor:	Dames & Moore, Twickenham, United Kingdom
Contract No.:	FI1W/0169-UK
Duration of contract:	November 1987 - July 1989
Project Leader:	P.S. Ringrose

A. OBJECTIVES AND SCOPE

The primary objective of the study is to allow more realistic modelling of the effects of ice sheet advances and retreats on deep underground disposal facilities for radioactive wastes and their environments. This is to be undertaken by advancing the status of fundamental research in this area. In Northern Europe glacial conditions are expected to return within the next 20,000 to 30,000 years and cycles of ice sheet advance and retreat to continue for the next million years at least. The influences on disposal sites could be significant. It is expected that algorithms will be developed from the work which, if used in a suitable computer code, will allow realistic modelling of ice sheet erosion and deposition and the effects of ice sheet advance/retreat cycles (including the associated ground freezing/thawing) on groundwater flow.

The project is being carried out in collaboration with researchers at the Grant Institute of Geology, University of Edinburgh, United Kingdom.

B. WORK PROGRAMME

B.1 Research status review: a brief review of the status of research into the effects of long-term environmental changes on deep disposal facilities.

B.2 Background Research: this second task is the major aspect of the work programme and incorporates research into three aspects of glacial processes as well as development of algorithms, as detailed below.

B.2.1 Investigation into the combined processes of permafrost development/degradation and ice sheet advance/retreat on groundwater flow systems and the physical properties of geosphere media, together with approaches to modelling such processes.

B.2.2 Investigation of approaches to modelling erosion during ice sheet advances and the consequent alteration of topography in the area surrounding a disposal site.

B.2.3 Investigation of approaches to modelling sediment deposition by ice sheets and the dispersion patterns of eroded materials, which could include contaminated geosphere or waste.

B.2.4 Specification for further research requirements arising from the project and of outline approaches to addressing these requirements.

B.3 Specification for the incorporation of site evolution modelling into performance assessment of geological disposal of radioactive wastes, with reference to three suitable sites in Europe.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The research status review (item B.1 above) has been completed and investigation into permafrost development (under item B.2 above) has progressed into a 2nd phase, with initial results having been reported.

Progress and results

C.1 Research status review.

A draft report of this review has been completed /1/. The report covers the following aspects:

- A summary of the nature of environmental change and the processes which are significant for deep disposal.
- A review of worldwide research into environmental change.
- A summary of environmental change modelling work at Dames & Moore.
- A comparison of approaches to the incorporation of environmental change into performance assessment.

C.2 Background Research.

The most important element of this work involves the development of a fundamental model of the growth and effects of permafrost layers which accompany glaciation. This model involves the application of the continuum theory of mixtures to a three-phase medium (water/ice/rock) and at the scale of interest (depths of hundreds of metres and timescales of thousands of years).

The first phase of this work has been reported /2/ and work under phase II is underway. Phase I resulted in the formulation of a one-dimensional model (Figure 1) in terms of a set of constitutive physical equations. Phase II involves the application of this model, the development of 2-dimensional models, and the inclusion of mechanical effects.

It should be noted that this work involves development of fundamental equations describing ground freezing at a scale not previously considered in work on soil freeze-thaw effects in soils.

The other aspect of the background research involves modelling the effects of glaciers on erosion of glacier substrates and the deposition of material from glaciers. Fundamental relationships for this have been previously developed /3, 4/ and will be applied in example illustrations of these effects.

C.3 Incorporation of results into performance assessments.
This third aspect of the work involves specification for the use of site-evolution models in the various performance assessment situations within the EEC. This work is currently in progress.

List of publications

/1/ DAMES & MOORE, A Review of Research into the Effects of Long-term Environmental Change on Deep Land Disposal Sites for Radioactive Waste, Technical Report TR-D&M-12, Draft (1988).

/2/ DAMES & MOORE, Background Research: Permafrost Evolution and Groundwater Flow Beneath Glaciers, Technical Report TR-D&M-16, Draft (1989).

/3/ BOULTON, G.S., Processes of Glacier Erosion on Different Substrata. J. Glaciology, Vol. 23 (89), 15-36 (1979).

/4/ BOULTON, G.S., Development of a Theoretical Model of Sediment Dispersal by Ice Sheets In: Prospecting in Areas of Glaciated Terrain, Inst. of Mining & Metallurgy, London, 213-224 (1984).

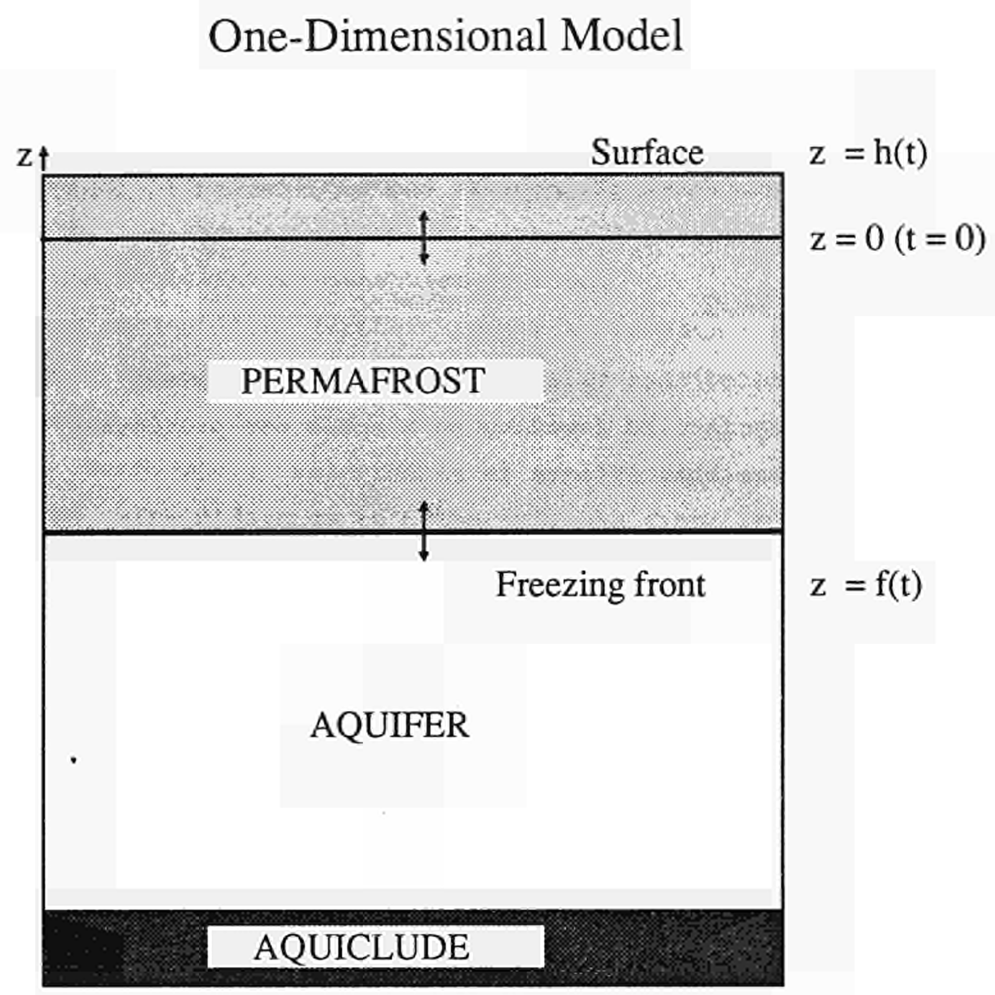


FIGURE I

ILLUSTRATION OF THE PRINCIPAL FEATURES OF THE ONE-DIMENSIONAL MODEL OF PERMAFROST GROWTH

HUMAN INTRUSION INTO UNDERGROUND REPOSITORIES FOR RADIOACTIVE WASTE

Contractor:	Associated Nuclear Services Limited, UK
Contract No.:	FI1W/0170-UK (H)
Duration of contract:	October 1987 to March 1989
Period covered:	January 1988 to December 1988
Project Leader:	D.J. Nancarrow

A. OBJECTIVES AND SCOPE

The objectives of the work are:

- to establish a methodology for the assessment of risks associated with human intrusion events applicable to the types of underground repository studied in the PAGIS and PACOMA projects;
- to delineate the application of the methodology to the alternative disposal concepts, taking into account the characteristics of the host rocks (clay, granite, salt and sub-seabed sediments) and repository designs;
- to produce an authoritative source of reference (database and methodology) for use in safety assessment of HLW and ILW disposal facilities.

B. WORK PROGRAMME

B.1 The various human activities which might result in an intrusion into a repository, or disruption of the host geology leading to enhanced radionuclide migration, will be identified. Relevant information necessary to define and quantify each mode will be collated.

B.2 The radiological consequences, to potential intruders and to others, from defined intrusion modes will be reviewed and appropriate calculation schemes defined.

B.3 The probabilities of intrusive events, identified as significant, will be estimated for reference circumstances. Factors affecting these probabilities will be identified and quantified as far as possible.

B.4 From the analysis made above, a general philosophy for the assessment of risk from human intrusions will be developed. The application of the methodology will be outlined for the particular host geologies and repositories considered in PAGIS and PACOMA, with the aid of examples.

B.5 The capacity for reduction of risks from intrusion by measures to reduce the probability or consequences of intrusion will be examined. This will include an examination of possible anti-intrusion measures.

C. PROGRESS OF WORK AND OBTAINED RESULTS

Summary

A study of groundwater abstraction from sedimentary and crystalline rocks has been undertaken in the context of intrusion into a repository or its environs. A parallel study of methods and techniques employed for exploitation and extraction of physical resources, such as minerals, has been carried out under subcontract. Approaches that may be adopted to assess human intrusion into deep underground repositories for radioactive wastes have been reviewed. A review has also been taken of the capacity for reduction of the risk from intrusion.

The status of progress is as follows: B.1 and B.5 are complete, B.3 and B.4 are in progress, B.2 is commencing.

Progress and results

1. Preliminary review (B.1)

A review of previous assessments of human intrusion into deep underground repositories was reported in the annual progress report for 1987 /1/. This review formed the basis for the definition of information required relating to human activities that could lead to intrusion.

2. Characterisation of human activities (B.1,B.3)

2.1 Groundwater abstraction

Of the host strata of interest, abstraction directly from granite is possible; however, clay and salt formations are not normally used for water abstraction on account of the low yield of the former and the low water and high salt content of the latter. Groundwater could be abstracted from sedimentary formations vertically adjacent to salt or clay formations, such as limestone, chalk and sandstone. Such abstraction may lead to penetration of the repository and/or contaminated aquifers.

The study has included consideration of the following:

- groundwater as a resource in present-day Europe;
- groundwater investigation techniques and development processes;
- likely long-term changes in abstraction;
- characterising borehole depths, yields, life expectancies and frequencies for both granite and sedimentary lithologies under a range of climatic conditions.

Borehole depths, yields and life expectancies are all a function of complex lithological and economic factors which vary considerably on both a global and national scale. The depth of a borehole in sedimentary strata depends primarily upon the depth of suitable aquifers below the ground surface. Such aquifers may be at depths of more than 500 m, but, for sandstone in the UK, are typically 45-60 m. The weathering and fracturing of granite, resulting in increased permeability, is usually limited to less than 100 m and so boreholes typically have a maximum depth of 45-75 m. In general, yields are significantly greater from sedimentary aquifers ($>10\ ls^{-1}$) than from granite aquifers ($<5\ ls^{-1}$) and so the latter are rarely used for the public water supply. In granite regions, the poor yield results in a low occurrence rate of boreholes, these being mainly used for private supplies. The decline in yield with depth, shown in Figure 1, restricts the depth of these boreholes and so the frequency of boreholes below 100 m is even smaller. The greater yields of sedimentary aquifers mean that they are more heavily exploited for both public and private water supplies than granite aquifers. In this case, the frequency of boreholes does not necessarily decrease with borehole depth, although costs often restrict the depth.

2.2 Deep drilling and mining

A study has been undertaken by Mott, Hay and Anderson, under subcontract to ANS, to characterise current methods and techniques employed for resource exploitation and extraction. The study begins with a discussion of the nature of the physical resources and the geological processes that give rise to them. This is followed by a discussion of the general investigation process, the legal framework, and the current investigation techniques including remote sensing, geophysical and geochemical methods. Consideration is given to whether unusual or ambiguous data from the investigation techniques might lead to the identification of the repository. For example, seismic surveys may detect cavities from changes in sound velocity associated with the presence of backfilling clays.

After preliminary investigations, the resource evaluation process will involve drilling. Figure 2 indicates both the range of depths and the more usual depths to which wells and shafts are sunk. If drilling intercepts a radioactive waste repository then, for high level waste, vitrified material may be included in the core. Such material might initially be mistaken for bright coal, or possibly dark flint; further examination for hardness texture and density would then place the material within the igneous range of rock types. In all cases, special interest would be aroused.

The extraction of a resource has been considered for evaporites, coal, bulk aggregates, metallic ores, water, oil and gas, seabed minerals, and geothermal heat. The methods of extraction have been illustrated by example.

3. Approaches to assessment (B.4)

Differing approaches to the assessment of intrusion have been reviewed. These have included: impact criteria; the degree of pessimism; the type of analysis (deterministic or probabilistic).

Criteria applied may relate to dose, conditional risk (i.e. the risk conditional on the intrusion event occurring) or overall risk. Overall risk calculations generally require account to be taken of the annual probability of intrusive event. In view of the difficulty in justifying such probabilities for events far into the future, and hence their subjective nature, it is recommended that overall risks should not be quoted alone but that assumptions regarding the annual probabilities should also be stated clearly.

4. Capacity for risk reduction (B.5)

A review has been undertaken of the potential for the reduction of risk associated with human intrusion. Risk may be reduced by reduction of the likelihood of intrusion and reduction of the consequences. The likelihood of inadvertent intrusion may be reduced by appropriate siting of the repository and by employing the best means available to communicate its existence and nature to future generations. The consequences of inadvertent intrusion may be minimised by packaging wastes in inert matrixes, by carefully considering the spatial arrangement of waste containers and the geometry of the repository, and by use of anti-intrusion shields.

References

/1/ COMMISSION OF THE EUROPEAN COMMUNITIES. The Community's research and development programme on radioactive waste management and storage. Annual Progress Report 1987. To be published as EUR 11482 EN.

/2/ DAVIS, S.N. and TURK, L.J. Optimum depth of wells in crystalline rocks. Groundwater, Vol. 2, Part 2, p. 6-11, 1964.

/3/ KOZLOVSKY, YEA. The world's deepest well. Scientific American Volume 251 Number 6, p. 106-112, 1984.

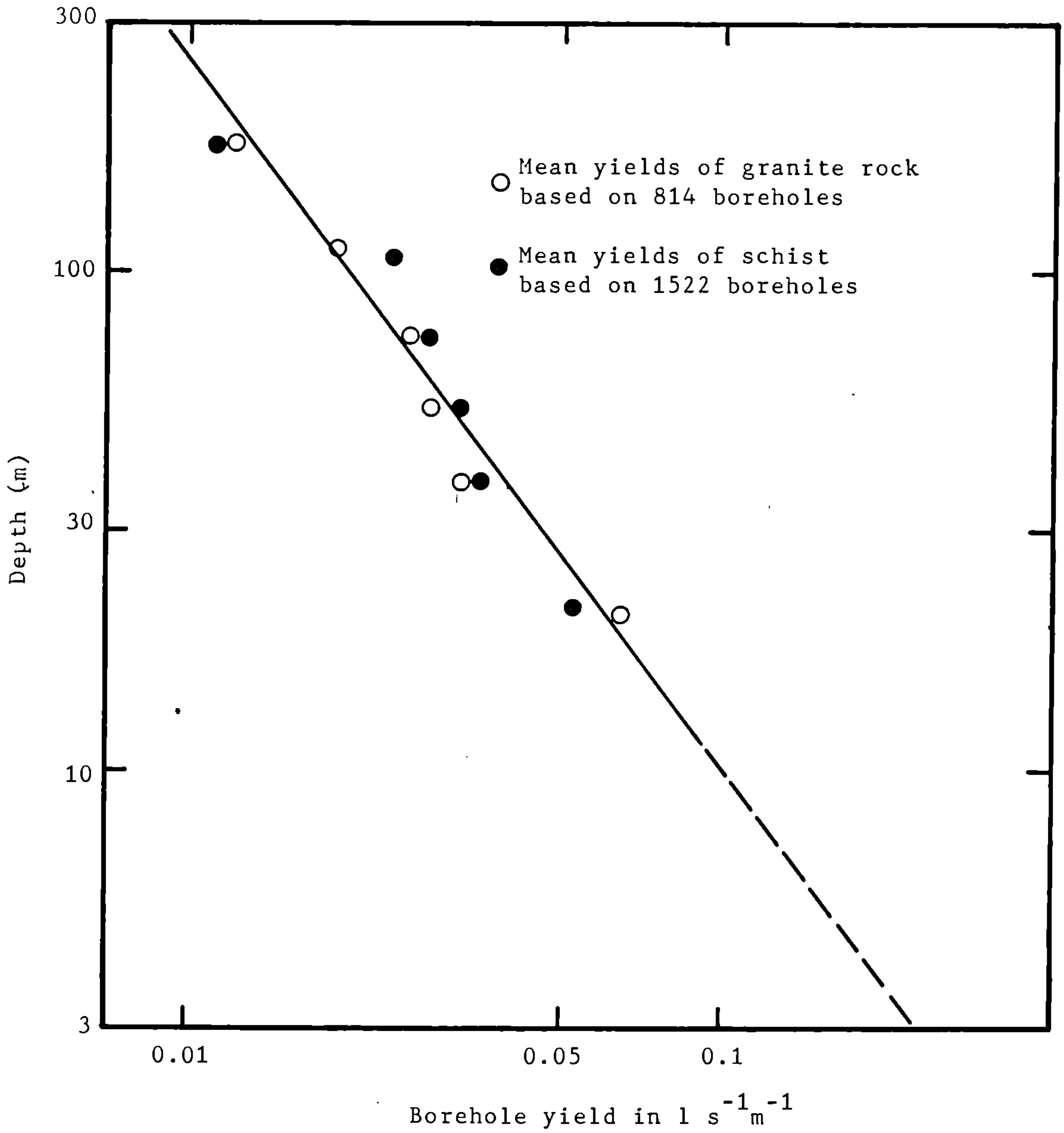
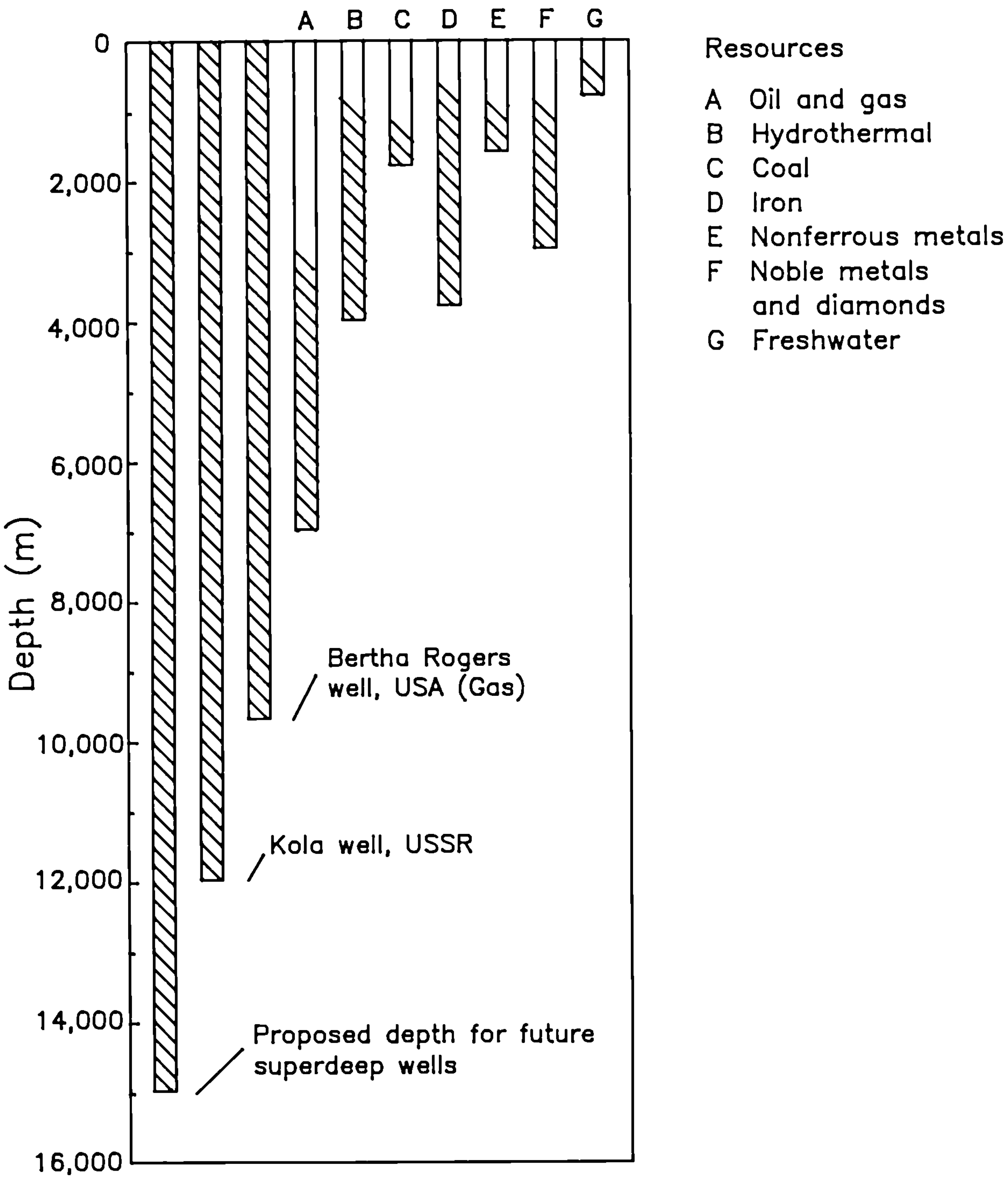


FIGURE 1 YIELDS PER METRE OF BOREHOLE VERSUS DEPTH IN CRYSTALLINE ROCK OF EASTERN UNITED STATES (AFTER DAVIS AND TURK /2/)



Note: The white area on the bar graphs represents the more usual depths to which shafts and wells are sunk

FIGURE 2 TYPICAL DEPTHS AND RANGES FOR WELLS AND MINE SHAFTS (AFTER KOZLOVSKY /3/)

TASK No 6

Joint elaboration of radioactive waste management

POLICIES

TASK No. 6: JOINT ELABORATION OF RADIOACTIVE WASTE MANAGEMENT POLICIES

A. Objective

Joint elaboration of waste management and disposal criteria.

Evaluation of possible approaches, at Community scale, for waste disposal.

B. Research topics dealt with under the 1980-1984 programme

These research topics were not included in the 1980-1984 programme.

C. 1985-1989 programme

The following activities are foreseen:

- Development and harmonization of acceptance criteria for radioactive waste conditioning with respect to their handling and final disposal;
- Development of radiological criteria for disposal, especially for the periods of time involved in geological disposal;
- Elaboration of recommendations concerning the satisfactory execution, taking into account the safety and environmental protection standards, of the various operations involved in the management and disposal of radioactive waste;
- Study of "de minimis" criteria with regard to alpha/non-alpha and radioactive/non-radioactive waste;
- Multi-national dimensions of waste management; influences on its optimization; regional disposal.

D. Programme implementation

A working group of national experts has been set up to deal with Task 6 activities. Topics of first priority have been identified as items (b) and (d) of paragraph C above. Work on radiological criteria for disposal has been performed through cooperation in an ad-hoc working group of national experts and Commission staff. The group produced the draft of a report on "Objectives, standards and criteria for radioactive waste disposal in the European Community", which is near its final form.

In the field of the development of rules for exemption of specific radioactive waste streams from regulatory control, the group has selected radioactive waste not linked to the nuclear fuel cycle (hospital, research and other radioactive waste), as the first waste stream to analyse. Four contracts were concluded in order to produce input for the group's work.

THE MANAGEMENT OF RADIOACTIVE MATERIALS IN NON-NUCLEAR SITES IN THE UNITED KINGDOM.

Contractor : WS Atkins Engineering Sciences. Warrington UK.
Contract number : FI1W-0236
Duration of contract : from; 1st Sept. 1988 to; 1st Sept. 1989
Period covered : up to 20th February 1989
Project leader : Dr B J Tymons

A. OBJECTIVES AND SCOPE.

To study the use, storage, production and disposal of radioactive material at non-nuclear sites.

UK regulatory procedures for nuclear licensed sites and those operated by the UK Atomic Energy Authority (UKAEA) comprehensively monitor the use, storage and disposal of radioactive materials at these sites. This study covers non-nuclear sites, ie unlicensed (non-UKAEA) installations such as hospitals, laboratories or industrial premises, where radioactive substances are routinely handled as authorised.

A sample of sites will be selected, representing categories 2-9 as defined by the Radiochemicals Inspectorate in their 1986/87 report. A detailed study will be made into the use and storage as well as the production and disposal of radioactive materials at the sites. The study will use data gathered on site and from organisations involved in supply, disposal and regulation of radioactive materials at such sites. Computerised inventory methods will also be examined.

B. WORK PROGRAM

3.1 Gather information from organisations associated with the supply, regulation and disposal of radioactive material from non-nuclear sites and report on procedures in the UK.

3.2 Visit representative non-nuclear sites and produce a report presenting our findings and assessing the success of the data gathering.

3.3 Review of the data from 3.1 and 3.2 identifying any problems, including evaluation of computer based data storage methods.

C. PROGRESS OF WORK.

State of Advancement.

Currently, we are engaged in part 3, phase 1 as specified in the technical annex of the contract (3.1 of the work program) ie establishing initial contact with the organisations responsible for monitoring the use, supply and disposal of radioactive materials in the UK. Due to a heavy workload, some delays have so far been experienced establishing meetings with Her Majesty's Inspectorate of Pollution (HMIP) (formerly The Radiochemicals Inspectorate) but this is unavoidable as it is considered essential to obtain the cooperation of HMIP in the study. Research into regulatory requirements dealing with monitoring of the use of radioactive materials at non-nuclear sites is continuing, again with the cooperation of HMIP. The estimated three man-weeks allocated to phase 1 is expected to be met but will be over a longer time period than envisaged.

Progress and results.

Visits have been made to inspectors of the HMIP in London and Lancaster to explain the background to the study and to seek further guidance regarding the best method to achieve the objectives of the study.

It has been decided, as the HMIP centre at Lancaster is nearest to Warrington, that Inspectors based there should form the main basis of our future contacts. Having established a link with HMIP we are seeking their cooperation in arranging visits to non-nuclear sites. The participation of the HMIP in the project from the outset is considered essential as they are the nationally responsible regulatory body. It is considered advantageous to be accompanied by an HMIP Inspector on a visit to benefit from their familiarity with the site visits and to minimise disturbance at the premises visited.

Determination of activity levels and recommendations for the exemption of radioactive wastes from installations non including basic nuclear installations

Contractor : Commissariat à l'Energie Atomique, Fontenay-aux-Roses, FRANCE
Contract n°: FI1W-0237
Working Period : September 1988 - December 1988
Project Leader : A.M. CHAPUIS, J.M. ASSELINEAU

The first step of this work, a basic bibliography, was constituted from recent papers pursued on concepts of exclusion and exemption of substances or materials, also called "de minimis" principles, on the determination principles of derived limits, and on the already realized applications of this methodology (e.g. recycling of materials from the dismantling of nuclear installations, disposal of very low level radioactive wastes).

A survey of the present situation in other countries, especially in Europe, and their approach of this problem was investigated.

Concerning France, we have examined the present reglementary aspects in regard to radioactive waste and non radioactive waste, or standards waste, in collecting the main legislative texts.

The situation with regard to annual production and its development during recent years was analysed in inquiring beside ANDRA* in order to :

- characterize radioactive waste producers (also called, in France, "small producers"),
- characterize produced wastes (chemical and physical form, radioactivity...),
- be acquainted with the different possible ways of waste treatment.

Finally, an investigation was realized in an installation of waste producer in way to have a best approach, in situ, of the reality : What is the producer making ? What are his radioactive wastes becoming ? What are his other wastes becoming ? What are the difficulties encountered ?

* Agence Nationale pour la Gestion des Déchets Radioactifs : National Agency for Radioactive waste Management

PRACTICE OF TREATMENT OF RADIOACTIVE WASTES ARISING OUTSIDE THE NUCLEAR FUEL CYCLE IN THE FEDERAL REPUBLIC OF GERMANY

Contractor:	GRS, Köln, Federal Republic of Germany
Contract No:	FI1W-0238
Duration of contract:	December 1988 - May 1990
Period covered:	December 1988
Project leader:	W. Müller

A. OBJECTIVES AND SCOPE

It is intended to provide an overview on the classification, treatment and, where appropriate, the exemption from regulatory control of radioactive wastes arising outside the nuclear fuel cycle. The investigations is based on an exemplary evaluation of the current situation.

B. WORK PROGRAM

1. Analysis of waste streams for main sources, activities and amounts

2. Investigation of the organisation of waste collection and treatment, especially in the areas

   - criteria for classification
   - licensing situation
   - kind of measurement
   - organisation of responsibilities
   - available conditioning procedures in place

3. Evaluation of results and possibilities for improvements

EVALUATION OF THE MANAGEMENT PRACTICES IN BELGIUM FOR RADIOACTIVE WASTE NOT LINKED TO THE NUCLEAR FUEL CYCLE

Contractor: Belgatom S.A., Brussels, Belgium
Contract N°: FI1W-0239
Working Period: November 1988 - March 1989
Period Covered: November 1988 - December 1988
Project Leader: P. Dardenne

A. Objectives and Scope

The objective is the investigation of the situation of the management of radioactive waste outside the nuclear fuel cycle, which is called AMIRI in Belgium (Applications médicales et industrielles des radio-isotopes).

The study is limited to the Belgian territory. The aim is to set up a "state-of-the-art" report based on information supplied by the agency responsible for the management of this type of waste (ONDRAF/NIRAS) and by a number of waste producers providing voluntarily the requested data. The practice for storage, treatment and transport of this matter will be compared to repository procedures, set up in particular by the national agency. Received information is to be presented in a statistical (anonymous) manner.

B. Work Programme

2.1. Evaluation of types and quantities of radioactive waste based on a questionnaire to be sent to about 60 producers.

2.2. Analysis of the existing practice in Belgium, concerning treatment, storage and transport of this type of radioactive waste in Belgium, and comparison to national regulations.

C. Progress of Work and Obtained Results

64 questionnaires were sent to producers, out of which 19 filled in forms were returned.

PART B

CONSTRUCTION AND/OR OPERATION OF UNDERGROUND EXPERIMENTAL FACILITIES OPEN TO COMMUNITY JOINT ACTIVITIES

The HAW Project

THE HAW PROJECT: DEMONSTRATION FACILITY FOR HIGH-LEVEL RADIOACTIVE WASTE DISPOSAL IN THE ASSE SALT MINE

Contractor: GSF-IfT, Braunschweig, Federal Republic of Germany
Contract No.: FI1W/0003/D
Duration of Contract: from January 1985 to December 1989
Period covered: January 1988 - December 1988
Project Leader: T. Rothfuchs

A. OBJECTIVES AND SCOPE

Since 1968 the GSF has been carrying out research and development programs for the final disposal of high-level radioactive waste (HAW) in salt formations. The heat producing waste has been simulated so far by means of electrical heaters and also cobalt-60-sources. In order to improve the final concept for HAW disposal in salt formations the complete technical system of an underground repository is to be tested in a one-to-one scale test facility.

To satisfy the test objectives thirty high radioactive canisters containing the radionuclides Cs-137 and Sr-90 will be emplaced in six boreholes located in two test galleries (Figure 1) at the 800 m-level in the Asse Salt Mine. The duration of testing will be approximately five years.

For handling of the radioactive canisters and their emplacement into the boreholes a system consisting of transportation casks, transportation vehicle, disposal machine, and borehole slider will be developed and tested. The actual scientific investigation program is based on the estimation and observation of the interaction between the radioactive canisters and the rock salt. This program includes measurement of thermally and radiolytically induced water and gas release from the rock salt and the radiolytical decompositon of salt minerals. Also the thermally induced stress and deformation fields in the surrounding rock mass will be investigated carefully.

The project is funded by the BMFT and the CEC and carried out in close cooperation with the Netherlands Research Foundation (ECN). Since 1988 the French Agence Nationale pour la Gestion des Déchets Radioactifs (ANDRA) is participating in the field of dose and doserate measurements and of laboratory irradiation experiments and in situ inclinometer measurements. Also the Spanish Empresa Nacional de Residuos Radioactivos (ENRESA) is participating in the salt irradiation programme.

B. WORK PROGRAM

B.1. Elaboration of the test plan and the supporting documents for the licensing procedure.

B.2. Development and procurement of the technical components for handling and emplacement of the radioactive canisters.

B.3. Procurement and installation of the data collection system.

B.4. Mining of the test field, drilling of the boreholes, installation of the measuring equipment and preparation for the emplacement of the HAW canisters.

B.5. Test disposal including operation of electrical tests for comparison and assessment of the technical components.

B.6. In situ-measurements of released water and gas from the salt, of thermally induced stress and deformation in the rock mass, and performance of seismic and ultrasonic measurements.

B.7. Accompanying and complementary laboratory investigations to ensure the transferability of the results to other sites.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The test disposal of HAW canisters in the Asse salt mine will be performed with a view to the planning, design and licensing procedure for a national repository in the FRG. In 1988 most emphasis has been given to receiving and installation of the technical components. In late 1988 the two preceeding electrical reference tests in boreholes A1 and B1 were started. Due to a delay in the licensing procedure the transportation and emplacement of the radioactive canisters could not be performed and has been postponed to 1989.

Progress and results

1. Elaboration of the test plan

The test plan was prepared in 1984/1985 and the final version was issued in December 1985/1/. It contains a detailed description of the scope, issues, and objectives and also of the test programme.

All the technical papers for the canister handling system, including the design calculations, were submitted to the responsible licensing authority (Bergamt Goslar) or its consultant (TÜV Hannover). Most of the components are meanwhile approved except for the transportation casks which are to be licensed by the Physikalisch Technische Bundesanstalt (PTB).

2. Development and procurement of the technical components

The fabrication of the technical components of the handling system for the radioactive canisters was terminated in 1988. All the components were delivered to the Asse mine and most of the equipment was mounted in place and is now being tested.

The construction of both the canister guiding system (CGS) as well as the gap monitoring system (GMS) have been successfully completed. The heater A1 together with the special equipment and the heater B1 with a GMS have been installed in the boreholes and are after approval of Bergamt and TÜV successfully in operation since November 8, 1988.

3. Procurement and installation of the data collection system

The Data Collection System (DCS) hardware components together with the software have been installed in the Asse mine. The system was approved by the authorities and is successfully in operation since November 1988.

4. Mining and preparation of the test field

The underground test field, consisting of two parallel galleries, each 60 m long, 10 m wide, and 8 m high, was completely mined in 1985. Until the end of October 1988 the liners in the boreholes A1 and B1 were installed together with the necessary measuring equipment. The remaining six liners will be installed in 1989.

5. Test Disposal

Due to the delayed fabrication of the borehole liners because of QA-requirements the time schedule of the project is again delayed. After start up of the preceeding electrical tests in boreholes A1 and B1 on November 8, 1988 the emplacement of the radioactive canisters is now foreseen for the fall of 1989.

6. In situ measurements

Already at the end of 1987 most of the instrumentation for monitoring stresses, stress changes, rock deformation, temperatures and gas release was installed. The first set of instruments at the emplacement boreholes A1 and B1 has been connected to the automatic DCS. During the isothermal pretest phase which started in 1985 with the start of the excavation of the test galleries a nearly constant secondary stress state developed in the pillar between the galleries A and B. A vertical principal stress component of 20.7 MPa and a correspondent horizontal stress component of only 4 MPa was measured at the end of 1988. The creep rates in the test field were nearly constant in the years 1987 and 1988. After start up of heating at the test sites A1 and B1 these deformation rates increased as expected. The horizontal closure rate at Test Site B1, for instance, increased from 24.5 mm per year in the pretest phase to an average rate of 35.5 mm per year.

The special instrumentation developed to obtain the salt pressure from the liner deformation has been installed together with the A1 heater. Figure 2 shows the salt pressure for the period from start-up of the heaters until December 31, 1988. The value of about 260 bar agrees well with the calculated predictions.

The preliminary acoustic measurements for crack detection near the borehole B1 and in the pillar were completed. The newly developed transient recorders have been installed and are now in operation.

The installation of packers in the 46 boreholes for determination of the generated and liberated gas in the whole test field have been finished. Measuring the gases already present in the rock salt and libe rated at natural mine temperature have been continued. No significant changes to the results measured before have been found.

Since November 1988 the determination of gases generated and liberated at elevated temperatures at the test sites A1 and B1 have been started. These measurements indicated a significant increasing of the components CO_2 and CH_4. In the gap between the borehole liner and the salt small amounts of H_2 have been analyzed.

The amount of these components is still increasing. H_2S is not yet detectable at test site A1 and B1 neither before nor during heating but in other areas of the test field it was analyzed as it was already liberated at natural mine temperature.

Additionally to this gas determination programme it is planned to irradiate salt samples in special canisters (dummy canisters) installed on the top of the radioactive canister stack. Within this programme it is planned to investigate the generation and liberation of gases as a function of the

- mineralogical composition
- dose rate
- dose
- temperature, and
- time

This programme has been set up and the sample ampoules have been designed. The different salt samples are now in preparation.

7. Accompanying laboratory investigations

The investigation of salt samples taken from the pilot cores of the emplacement boreholes on the gas content and the thermal liberation behaviour has been continued. It indicated that significant amounts of CO_2, CH_4, and H_2S have been liberated at room temperature. Heating of the samples stepwise to 200 °C will start soon.

At the Rheinisch-Westfälische Technische Hochschule in Aachen a computer code for calculation of the diffusion and migration of the generated and liberated gases is in development. Significant parameters for this code are the porosity and the permeability of the rock salt. These parameters are currently investigated on salt cores taken from the predrillings of the emplacement boreholes. They are investigated as a function of:

- confining pressure up to 20 MPa
- gas presssure up to 10 MPa
- temperature up to 200°C

The permeability of these samples is in the range of 10^{-17} to 10^{-19} m^2.

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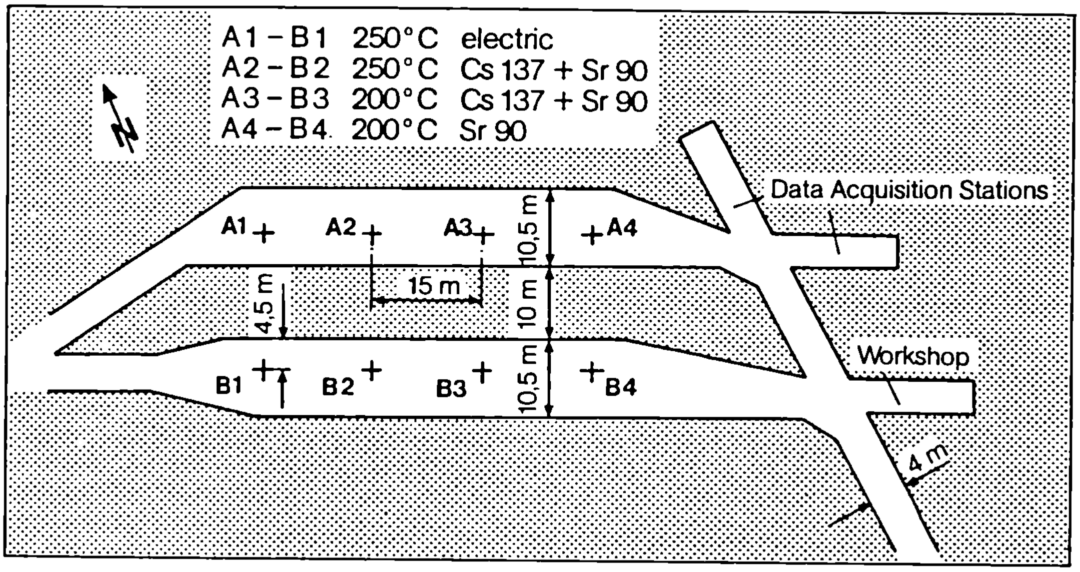

Figure 1: HAW Test Field at the Asse Salt Mine

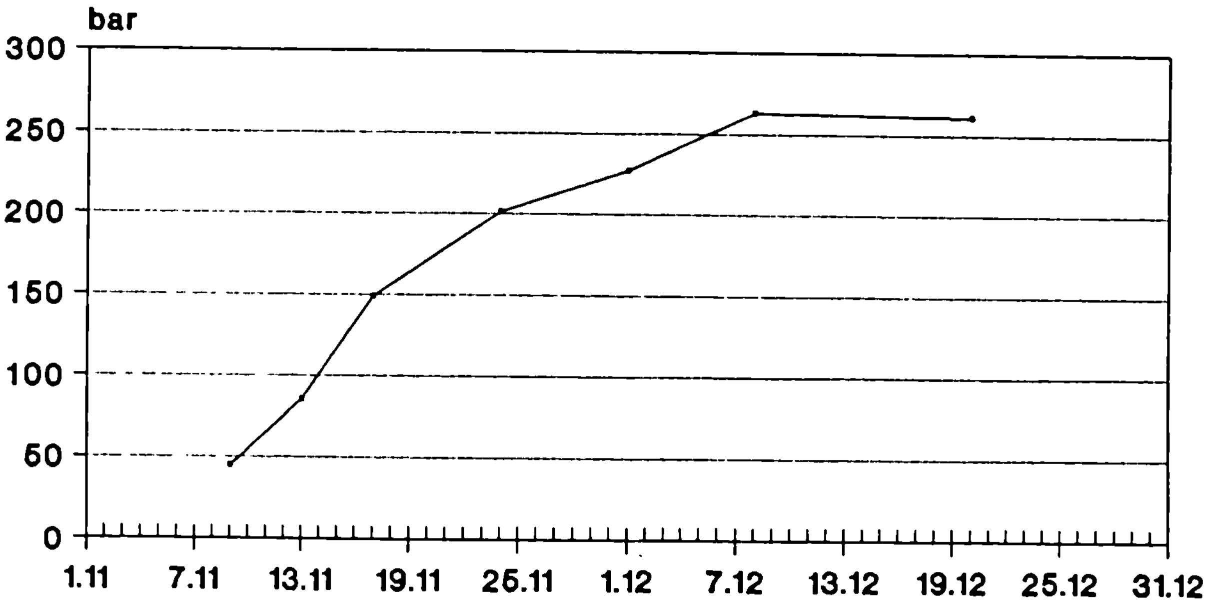

Figure 2: Development of the Salt Pressure at Borehole Liner A1

IRRADIATION EFFECTS ON ROCKSALT

Contractor : ANDRA
Contract n° : FI1W/0199
Duration of contract : from Jan 88 to Dec 89
Period covered : from Jan 88 to Dec 88
Project leader : ANDRE JEHAN

A - OBJECTIVES AND SCOPES

ANDRA is involved in the Research and Development programme for the final disposal of radioactive wastes (HAW project) carried out by GSF-IfT in the ASSE salt mine near Braunschweig (FRG). ECN (Netherlands) and ENRESA (Spain) are also part of this project which receive funds from the CEC. The ANDRA's participation consists of several topics :

- gamma-ray dose assessment (prediction and measurement),
- study of the radiolysis of salt,
- study of heat effects on the rockmass.

These experiments include:
- design and construction of special equipment adapted to the very demanding test conditions,
- in-situ measurements (dosimetry, inclinometry),
- laboratory investigations (irradiations, analysis, calculations).

ANDRA's subcontractors are CEA (Commissariat à l'Energie Atomique), IPG (Institut de Physique du Globe), LMS (Laboratoire de Mécanique des Solides de l'Ecole Polytechnique).

B - WORK PROGRAMME

1. Gamma-ray measurements

Gamma doses and dose rates are recorded in various location of the HAW test field or handling equipment using both solid-state dosimeters and ionization chambers systems.

2. Calculation of dose distribution

Theoretical calculations of gamma flux in the rocksalt surrounding the sources are performed using a three dimension computer model which take into account all mechanical, chemical and isotopic data of the test.

3. Parametric study of the radiolysis of salt

ASSE salt samples are irradiated in a french facility at different dose rates, integrated doses and temperatures. The gases released are then analyzed.

4. Inclinometry measurements

High sensitivity measurements are installed around the test galleries, in a way to assess the displacements due to heating and know the long-term mechanical behaviour of the rockmass.

C - PROGRESS WORK AND OBTAINED RESULTS

State of advancement

The recording systems for ionization chamber measurements are completed and will soon be put under test for calibration. Radio-thermoluminescent products for solid-state dosimetry are selected and calibrated, and the complete measuring procedure is now determined. The prototypes of the dosemeters are constructed and now under test. The calculation of gamma dose distribution didn't start yet but all the data required for the computer model are now described in the very detail and the calculation can start soon. Concerning the analysis of radiolytic gases, the complete procedure is now determined, the irradiation programme started for ambient temperature, and firts results are now available. The inclinometry measurement system, consisting of 3 groups of 4 tiltmeters + 1 temperature recorder, was installed long time before the heating start in holes A.1 and B.1, in July 1988. It is operating with a stand-alone Data Colection System, but will be connected to the main DCS as soon as it will be ready.

Progress and results

1. Gamma ray dosemetry
1.1. Ionization chamber measurements

Such measurements are performed at the surface level in transfer machine and at the test site (- 800 m level) in the guide tube and boreholes, so two equipments were required.

- Surface level (transfert machine)

This equipment aims to record 2 profiles of activity for each glass canister when it is transferred from the transport cask to the mine cask. The ionization chambers are CRGE 10, a specially design, xenon-filled model (size : 7 m diam, 150 mm length). The data are stored on a 3" ½ floppy disk.

- 800 m level (test site)

The recording equipment and the winch are mounted on a trolley. Three different ionization chambers are used, each with a separate cable and spool for the winch : in the guide tube both CRGF 10 (radioactivity) and CRGF 0 (background) are used with a Teleflex type cable which allows to push the chambers into the tubes. In the boreholes a CRGC 10 chamber is used with an ordinary twin-lead shielded cable;

1.2 Solid state dosimetry

Those dosemeters use the properties of certains crystals of emitting light after having been irradiated : the radio-thermoluminescence (RTL). Selected products are Al_2O_3 (corrundum) and $CaSO_4$(Dy), they could be pre-dosed, in order to estimate the influence of temperature during irradiation. These dosemeters will be used :

- in the dummy canister above the sources,
- in the guide tubes and boreholes (mounted on a chain),
- in the transfer machine to measure the total activity of each source.

2. Calculation of dose distribution

The calculation itself didn't start yet but all data are collected and will be soon entered in the model. This model is TRIPOLI II and uses a Monte-Carlo transport method; gamma interactions taken into account are compton diffusion, pair effect, photoelectric effect. The energy deposited in salt is calculated for 1 Curie of each isotope from each source and then summed according to the real activity.

3. Radiolysis of salt

Begining of 1988 was devoted to test and qualify experimental procedures and to determine the irradiation programme (see table I). Samples consist of 200 g of salt sealed under controlled atmosphere in glass ampoules and irradiated by spent fuel elements. Gas analysis procedure includes :

- mass spectrometry (H_2, O_2, N_2),

- gas chromatography (CO, CO_2, N_2O, CH_4),

- ionic chromatography (Cl, SO_4, NO_2, NO_3)

- FTIR (Fourier Transform Infra-Red) only for some samples.

Irradiations started in October but high temperature irradiator is not yet operating due to licensing problems. At that time very few results are available ; no effects are noticeable for doses lower than 10^4 Grays

4. Inclinometry

This part of the contract, signed later in August 1988, aims to measure the movements of the deep rocksalt submitted to heating, using high resolution tiltmeters (10^{-8} rads). Twelve of such tiltmeters were dispached in three locations surrounding the test field (see fig. 1). They were installed three months prior to start the heaters in holes A.1 and B.1, in a way to observe the behaviour of the rockmass without external stress. The very preliminary results show an increase of the creep when the heating started (see fig. 2).

TABLE I	HAW PROJECT RADIOLYSIS OF SALT PLANNING OF IRRADIATIONS				
PARAMETER	SAMPLE N	DOSE RATE (Gy/h)	INTEGR. DOSE (10^6 Gy)	DURATION (Hours)	TEMPERATURE °C
CRUSHED SALT	12	1000	1	1000	Ambient
A/V ROCKSALT	9 6 6 6 6 6	1000 1000 1000 1000 1000 1000	1 0.001 0.01 0.1 1 10	1000 1 10 100 1000 10000	
DOSE RATE	3 3 3 3	100 1000 10000 100000	1 1 1 0	10000 1000 100 10	Ambient
DOSE	3 3 3 3 3 3 3 3 3 3 3	1000 1000 1000 1000 1000 1000 10000 10000 10000 10000 10000	0.0001 0.001 0.01 0.1 1 10 0.001 0.01 0.1 1 10	0.1 1 10 100 1000 10000 0.1 1 10 100 1000	Ambient
TEMPERATURE	3 3 3 3 3	10000 10000 10000 10000 10000	1 1 1 1 1	1 week 1 week 1 week 1 week 1 week	50 to 100
MINERALS	21	10000 1000	1 1	100 1000	Ambient
CONTAINERS	12	1	1	100	Ambient
GASES (N_2, O_2, Ar, He)	12	1 or 0.1	1	100 or 1000	Ambient

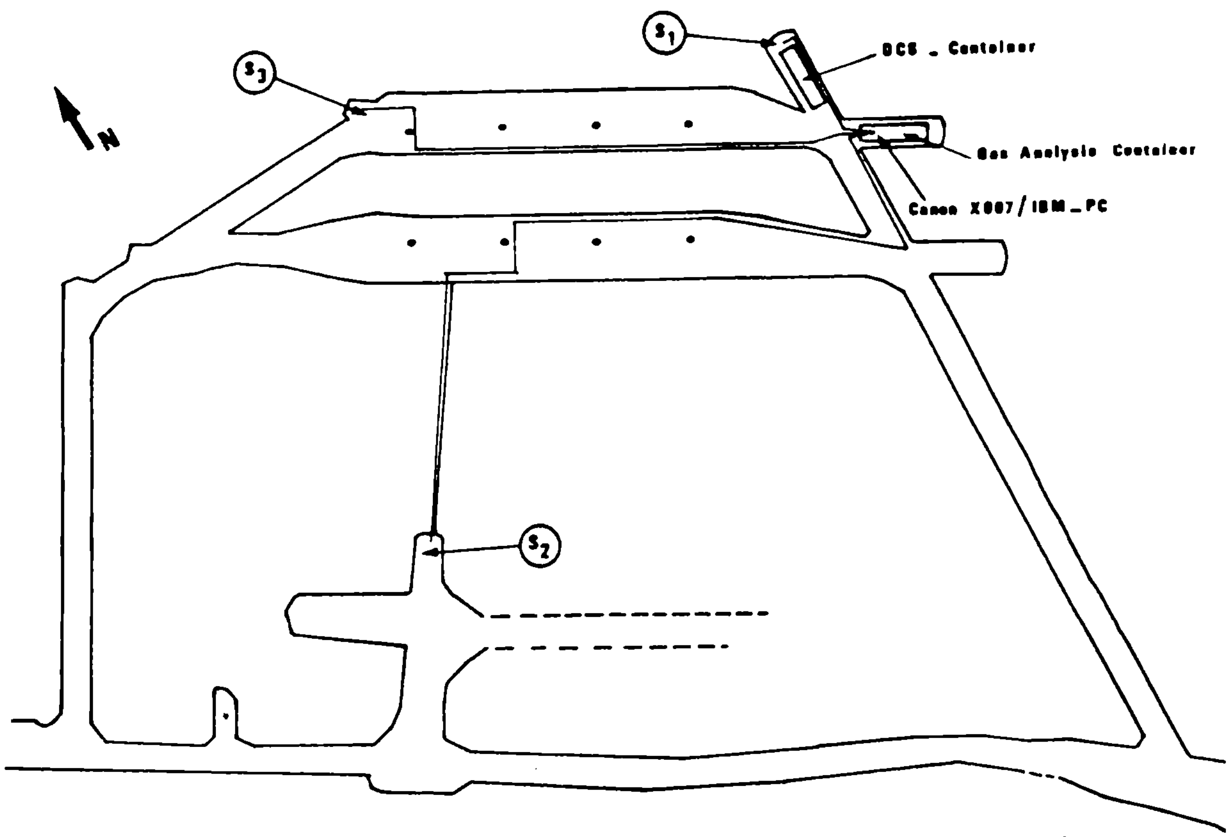

Fig. 1 : LOCATION OF TILMETER STATIONS (S1 -S2 - S3)

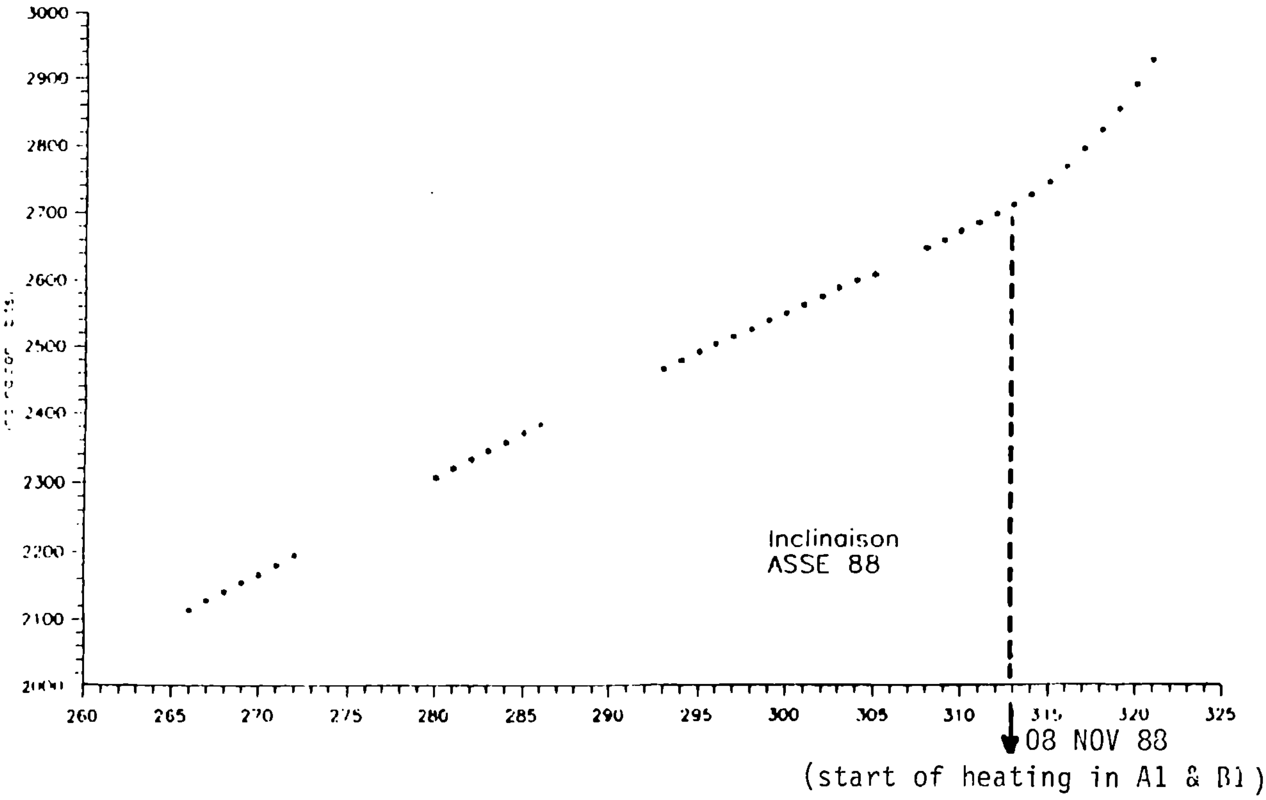

Fig. 2 : INCLINOMETRY MEASUREMENT IN S3

TEXTURAL AND FLUID PHASE ANALYSIS OF ROCK SALT SUBJECTED TO THE COMBINED EFFECTS OF PRESSURE, HEAT AND GAMMA RADIATION

Contractor: ENRESA, Spain
Contract No.: FI1W-0235-E
Duration of contract: November 88-June 90
Project leader: F. Huertas

A. OBJECTIVES AND SCOPE

Previous studies have shown that radiolytic processes induced by irradiation of salt create the migration of fluid phases and the formation of defects in the crystal lattice, i.e.: detachment of sodium ions which cluster together forming colloidal sodium. The formation of colloidal sodium gives rise to stored energy, which can be released by recombination of the colloids with the trapped chlorine. The temperature rises creating, according to this release, the main concern of irradiation damage with respect to the safety aspects of nuclear waste burial.

Due to differences in potential energy between neighbouring crystals, grain boundaries tend to move at the cost of the most energetic crystals; this recrystallization process, enhanced by the presence of brines, clears the crystal from irradiation damage.

The main goal of the project is to calculate the above-mentioned phenomenon, using the Jain-Lidiard theory, in heterogeneous and impure material, such as rock salt, when exposed to different dose-rate, temperature and time of irradiation.

The research work is being done in cooperation with the Netherlands Energy Research Foundation (ECN), the University of Utrecht and the University of Barcelona. The irradiation will be conducted at the HAW test field at Asse (FRG).

B. WORK PROGRAMME

1. Sampling of salt specimens from spanish sites and the HAW test field; preparation of synthetic polycrystalline and Harshaw samples.
2. Chemical, petrological and mineralogical characterization of the various salt samples. Solid and fluid phase determination, chemical evolution from step heating and microstructural analysis.
3. Textural analysis of undisturbed samples.
4. Preparation of samples and pressurized containers for the irradiation phase.
5. Irradiation of 180 samples in the HAW test field at Asse. Samples will be irradiated from 6 months to 5 years at different dose-rate and temperature, in pressurized containers with different brine contents.
6. Irradiation of some salt specimens at the High Flux Reactor (HFR) (Petten)
7. Post-irradiation analysis to investigate the amount of stored energy and the variation in chemical composition, water content and microstructure.
8. Theoretical calculations, on the basis of the Jain-Lidiard model, of the damage which could be expected by irradiation and heating.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Salt samples from the Sallent mine (Spain) have been obtained by airflushed rotary drilling.

Machining of Sallent samples, to be placed in the pressurized containers for the irradiation phase, is in progress although with some difficulties due to failures of samples, because of the textural characteristics of the salt rock under study. The texture of Asse samples does not create that problem.

Preparation of synthetic polycrystalline samples is also being difficult, so is the acquisition of gold foil for wrapping the samples. The possibility of replacing the gold by silver is being considered.

Prototype pressurized containers are being tested at ECN.

Drilling fines from Sallent have been analyzed by X-ray diffraction for a first mineralogical characterization of the salt. 40 core samples have been analyzed by thermogravimetry.

Irradiation at the HFR has been initiated. The system is being modified to ensure a better control on the temperature exposure of the samples.

Progress and results

40 samples of the Sallent mine have been machined down to the appropriate diameter. Their exact lenght has yet to be defined since it is being considered to place a pill of polycrystalline shynthetic salt, for reference, on top of each sample. This new approach tries to account for the lack of homogeneity observed between samples. The timing for having the 72 samples ready for loading the canisters by April is tight due to machining difficulties and the preparation of polycrystalline shynthetic salt samples.

Irradiation of Sallent samples at the HFR has been carried out. It was not possible to retrieve the thermal isolation of the system, consequently during the first cycle the temperature reached 130ºC, during the rest of the irradiation cycle took place at 100ºC. It has been decided to construct a new narrower pipe, to hold the canisters inside and so allow for more heat to dissipate towards the pull. The already irradiated samples will be used on recrystallization experiments to compare this behaviour with that of the Asse samples SP-800 from previous research A new GIF facility to irradiate under pressure is being designed at ECN.

The preliminary results of the lithogeochemistry analysis performed by XRD with the drilling fines of Sallent show the presence of halite and anhydrite as major components, small amounts of quartz and almost negligible contents of polyhalite. The results are summarized in Table I.

Chemical composition has been evaluated by AAS and volumetric methods. The results are presented in Table II. 40 core samples, crushed to a diameter between 2 and 3 mm, have been analyzed by thermogravimetry; data of the water loss up to 450ºC is shown in Table II.

TABLE I : MINERALOGY

	HALITE (%)	ANHYDRITE (%)	OTHER (%)
MEAN VAL.	95.55	2.76	1.52
ST. DEV.	2.45	1.88	1.09

TABLE II : GEOCHEMICAL COMPOSITION AND WATER CONTENT

	Cl (%)	SO4 (%)	Ca (%)	Br (ppm)	K (ppm)	Mg (ppm)	Sr (ppm)	Li (ppb)	Rb (ppb)	Water (%)
MEAN VAL.	57'95	1'96	0'96	60	112	428	230	720	< 250	0'34
ST. DEV	1'51	1'32	0'60	5	32	99	162	280	--	0'17

The HADES Prcject

The HADES Project : a pilot facility in the argillaceous layer beneath the nuclear site at Mol

Contractor : SCK/CEN, Mol, Belgium
Contract No. : FI1W/004/B
Duration of contract : from January 1985 to December 1989
Period covered : January 1988 - December 1988
Project leader : A.A. Bonne

A. OBJECTIVE AND SCOPE

In 1974 SCK/CEN launched a R&D-programme concerning the possibilities for disposal of high level solidified and alpha-bearing radioactive wastes in a continental stratiform clay formation (Boom clay) situated below its own site. Site investigations, safety studies, repository design, conceptualisations and in situ research confirm progressively the favourable characteristics of the host rock and the site for disposal of radioactive wastes.

Many particular areas require further studies and technological tests on a larger scale and in situ demonstrations under realistic conditions. These technological tests, studies and demonstrations will contribute to increase the confidence in the technical practicability, the economical feasibility and the safety of the disposal option in deep clay.

The direct demonstrations deal with the constructibility of real scale galleries without particular conditioning of the rock, the choice and dimensioning of a realistic lining and support system, the interaction between the underground structures and the immediate geological environment (e.g. the influence of heat and radiation), the handling of hot and radioactive canisters, the backfilling and its behaviour in time, the performance of various system components during the operational phase and of monitoring systems.

Within the HADES project a technological test related to a gallery lining technique according to the convergence-confinement principle is performed by ANDRA (France) (see contract FI1W/0112).

B. WORK PROGRAMME

The demonstration/pilot phase of the HADES project is scheduled in two phases, which are complementary to each other and may be developed in parallel.

B.1. Phase I : the construction and operation of a test drift with tests related to :

B.1.1. Mining technology (digging, lining, extrados backfilling, rheology).

B.1.2. Radioactive waste disposal (experimental emplacement, backfilling, degradation of waste matrices and migration of radionuclides, in situ irradiation of clay, thermo-mechanical behaviour of clay and gallery structures, monitoring and auscultation systems).

B.2. Phase II :

B.2.1. The construction of a pilot facility with a new shaft and extended gallery, connecting chamber and utility structures.

B.2.2 Tests and observations on handling, emplacement, backfilling and retrieval of dummies and finally of acutal radioactive wastes.

(The performance of B.2. is scheduled beyond the present contract period).

C. STATE OF PROGRESS AND RESULTS OBTAINED

State of progress

After having finished the construction of the Test Drift on December 17th of 1987, the Test Drift was completed by installing several service and safety items. The general auscultation of the time dependent behaviour of the clay host rock around the Test Drift and of the TD lining were continued and confirmed the excellent geotechnical behaviour of the whole structure.

Validation exercises of rheological models for simulating the geotechnical behaviour of the Boom clay during the construction of the TD have been launched. The test unit for the Cerberus test (combined radiation/heater test) has been installed in the TD.

Progress and results

1. Mine-by-test around the stiff section of the TD (B.1.1)

The geotechnical observations and survey undertaken in the TD and in the clay mass around it are briefly described hereafter.

2. Convergence measurements and pressure build-up on the lining

The diametrical convergence of the lining reaches, after 12 to 15 months, about 1 % of the excavated diameter with an actual convergence rate of about 0.5 mm/month. The total pressure acting upon the lining has not shown any important change recently. By comparing the evolution of convergence and confinement with time, the contribution of the intercalary wooden plates (between the concrete segments) in the overall convergence is well enhanced.

3. Deformations in the clay mass

The displacements in the clay body are recorded in the roof of the TD at different distances from the TD. By extrapolation of these measurements, a convergence of 8 % may be inferred at the excavated wall. Measurements are sensitive to the excavation progress, they started with a tunnel heading at a distance of 2 times the diameter, reach about 50 % of the total deformation when the tunnel face is directly beneath the tunnel face and continue to increase until the heading reaches 4 to 5 times the radius further.

4. Pore pressure measurements

After the important initial pore water pressure drop, due to the excavation and face advance, the data recorded show a slow but continuous pressure increase. A zone of influence of about 4 times the excavation radius, as well ahead as behind the front face is observed again.

5. Clay front auscultation

From the displacements measured at several positions behind the front, it may be inferred that a volume up to 4 m in the clay is influenced by the front movement, which reached 65 mm in November 1988.

6. Geotechnical auscultation of the clay (B.1.1)

Self Boring Pressuremeter (SBP) tests have been carried out by the company PMIT (Pressuremeter In situ Techniques (Cambridge) in April 1988. A complete interpretation of all data recorded is not yet available, but the tendency confirms laboratory triaxial tests performed at the University of Liège on clay sampled during the excavation works : the mechanical characteristics (E modulus, shear strength) are significantly lower at 223 m than at greater depth. An undrained shear strength of 0.8 MPa, should be taken into account, instead of 1.0 to 1.5 MPa, as expected earlier.

7. Cerberus test (B.1.2)

The purpose of the Cerberus test (Cerberus = Control Experiment with Radiation of the BElgian Repository for Underground Storage) is the in situ study, by simulation, of the heat and radiation impact a HLW-source upon its immediate clay environment. The simulation source consists in a Co-60 source of 450 TBq and two heating elements. The test set-up is shown on figure 1.

This year, the test device was manufactured and placed into the clay. The authorization to use for this purpose a maximum 555 TBq of Co-60 was granted in April 1988.

For handling the container with the Co-60 source in the shaft and in the TD a loader has been designed and built with a teleguided boom extension (figure 2).

The safety report for the Cerberus-test was submitted to the authorities and actually no major objection was formulated. The container for handling the Co-source was also approved by the authorities.

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Excavation response studies at the Mol facility
OECD/NEA Workshop on excavation response in deep radioactive waste repositories, Winnipeg, Canada, April 1988

NEERDAEL B., BONNE A.
Ongoing research on geological disposal of radioactive waste in the Boom clay layer - last in situ developments
Third Meeting of ISAG, Washington DC, 2-4 May, 1988

NEERDAEL B.
Geotechnical investigation in the Hades Project
ISRM Symposium Rock Mechanics and Power Plants, ENRESA Workshop on Rock mechanics and radioactive wastes, Madrid, September 1988.

NEERDAEL B., DE BRUYN D.
Geomechanical research in the test drift of the Hades underground research facility at Mol
CEC Contractor's Meeting on Geomechanics of clays for radioactive waste disposal, Brussels, 1-2 December 1988

DE BRUYN D. and al.
Construction of test drift in a deep tertiary clay formation
Tunnel, 1988, 4 (in print)

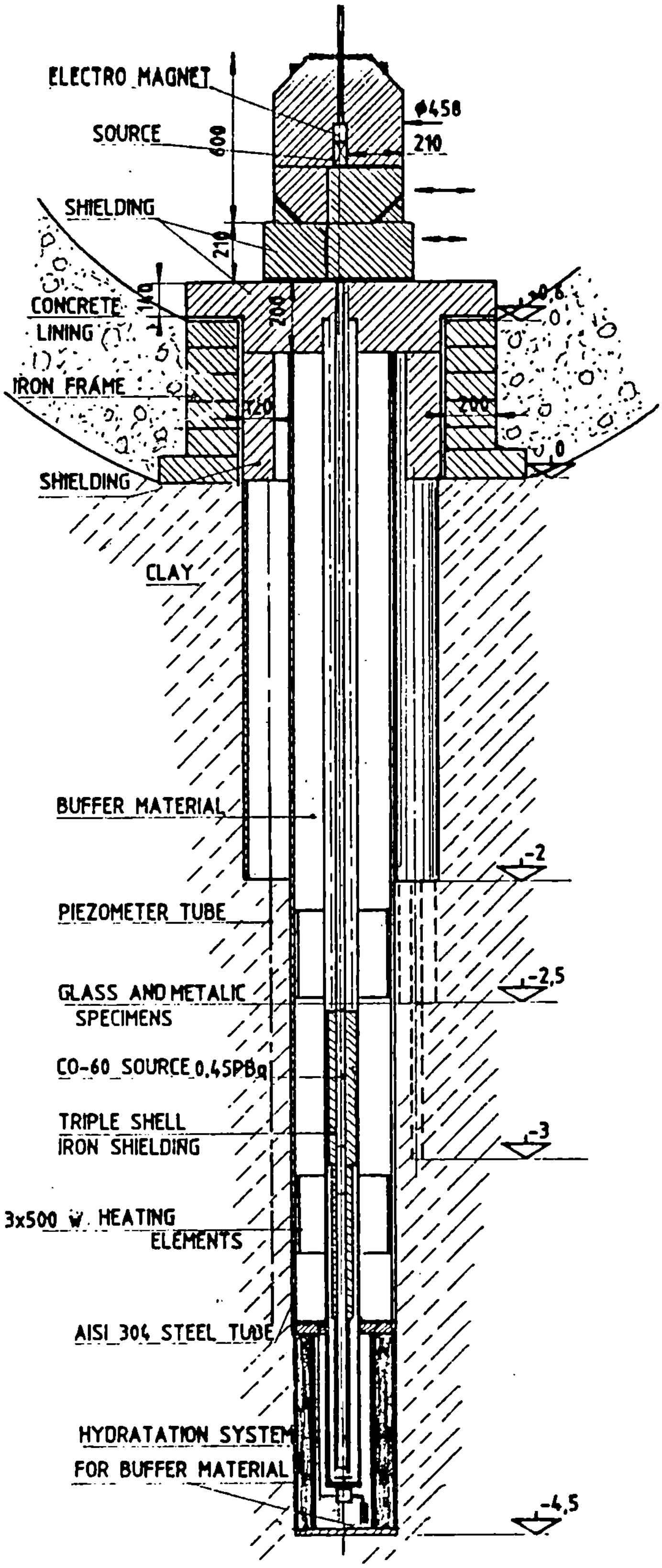
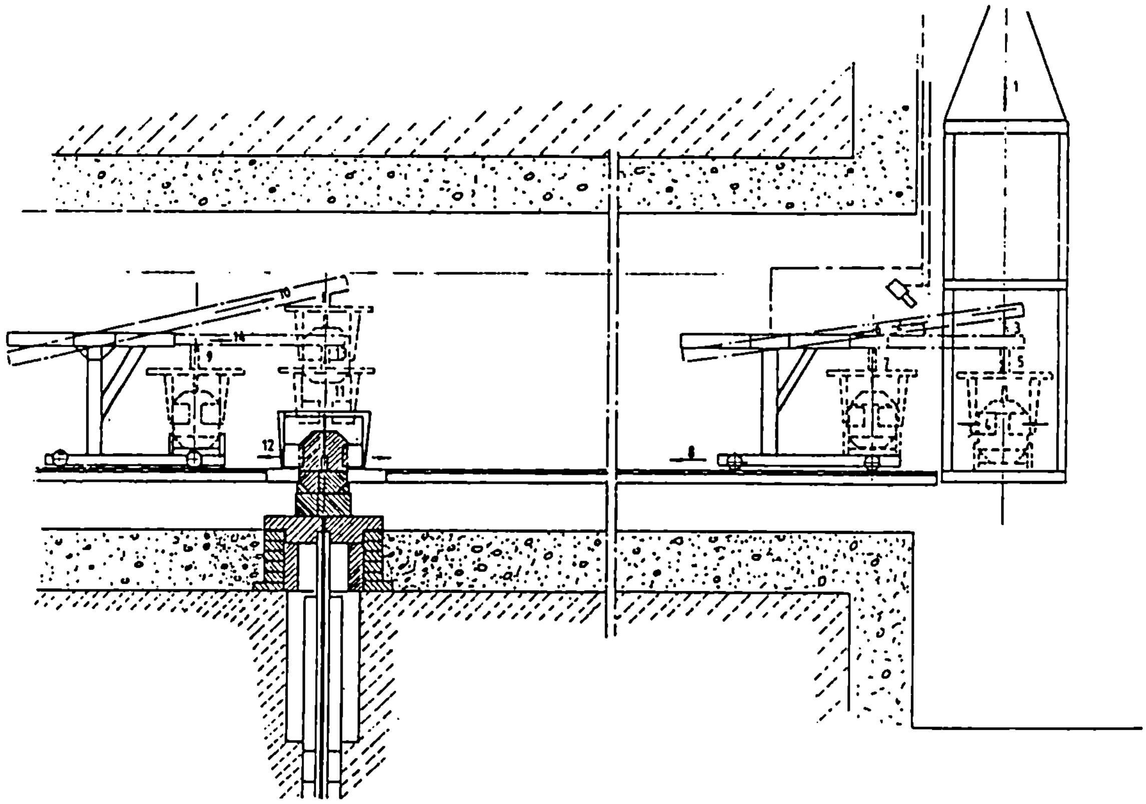


Figure 1. Lay-out of the Cerberus test



1. Transport of the container with Co-60 source from surface to URL
2. Sliding out of the HBE (Hydraulic Boom Extension)
3. Lowering of the HBE
4. Jamming the container with the clips
5. Lifting up of the container
6. Sliding back of the HBE
7. Putting down the container
8. Transport to the Cerberus experiment

9.-12. idem in reverse

Figure 2. Handling machine and loader for the Co-60 source (Cerberus test)

DIMENSIONING OF LINING OF GALLERIES EXCAVATED IN DEEP CLAY FORMATIONS

Contractor : ANDRA, PARIS, FRANCE
Contract n° : F I1W/0112
Duration of contract: January 87 - December 89
Period covered : January 88 - December 88
Project leader : R. ANDRE JEHAN

A - OBJECTIVES AND SCOPE

The dimensioning of lining of galleries in deep clay formations depends directly on the long term stress undergone by the support.

The mechanical characteristics of the clay formation could be used to optimize the lining. It seems possible to minimize the pressure in the support, by allowing the excavation wall to converge enough with time. Different factors can be considered to reach this objective, as the time lag for the rock to come to contact with the support or the flexibility of the lining.

The objective of this work is to implement a lining wich answers this criterium. It is made of steel ribs showing a significant stiffness with sliding devices adapted at the joint elements, allowing the convergence. This configuration will test the closure - confinement concept : the convergence of the wall prevents the confinement to rise above a certain threshold. Moreover this thin lining presents practical advantages i.e. the handiness and ease of transport, the rapidity for building and the small volume of material to dig and dispose.

The conception of the project and the study of the rockmass-lining behaviour are conducted by LMS (Laboratoire de Mécanique des Solides de l'Ecole Polytechnique). Engineering department of SIMECSOL was responsible for the instrumentation and measurements during construction, collaboration of CEN/SCK for surveying was since requested.

B - WORK PROGRAMME

1. Build up of a 12.5 m long test gallery in the continuation of a concrete drift made by the CEN/SCK. A transition zone of 2 m with concrete archstones prevent any perturbation of the CEN gallery.

2. Measurements of :

   - closure of implemented rings,
   - sliding of the ribs,
   - rib strain,
   - pressure on the outer face lining,
   - displacement inside the clay formation,

   during and after excavation, for 24 months at least.

3. Interpretation of results

C - PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The excavation, lining and instrumentation of the gallery were performed from 10/20/87 to 12/03/87. A buffer zone was set up at the end of the drift. It consists of a portion of a circular gallery 2 m long lined with concrete archstones. Its end is completed by a hemispheric front stabilized with gunite (figure 1). The measurement started during the digging and is still underway. The interpretation of results obtained for a year as been presented to the Commission in December 1988.

Progress and results

2. Measurements

The measurements was kept on for 24 months at least. The frequency was daily during the excavation phase, weekly during the first month after the completion of the work, bi-monthly during the 6 next months (1st semester 1988), then monthly. It will eventually be quarterly during the second year. Some measures are already automatically taken.

. Closure of complemented rings : a little ovalization in the vertical direction can be observed in the four ribs.

. Sliding of the ribs : in a first time after digging (3 months) slidings are importants, then the phenomena's evolution is quite moderate.
The sliding of the ribs are depending on their distance of the stabilized front (figure 2). Nine months after the gallery's construction the average convergence reaches 1,8 %.

. Rib strain : KOVARI's theory is used to estimate the normal load and the bendings moments, related to the strains by an elastoplastic model of steel behaviour. The critic normal force for the first sliding has a value of 500 kN the same that predicted value.
The normal force is about 980 kN and the bendings moments about 25 kN. The maximum values don't reach steel plastic limit.

. Pressure on the outer face lining : it is measured by Gloetzl cells disposed on the interface clay-backfilling. The pressure increases quickly as soon as the rib has been set and during one month, then its average value is quite constant and less than 1 MPa.

. Displacement inside the clay formation with extensometer : they increase in accordance with pressure measured by Gloetzl cells. They are the smaller than the excavation is away (at 10 meters there is no displacement).

3. Interpretation of results

- The lithostatic stress can be considered almost isotropic.

- The average confinement pressure is about 1.4 MPa after nine months. The experimental curve of confinement (figure 3) pointed out that pressure and closure become in a long run less and less important, but the stabilization is not yet reached.

- The Gloetzl cells don't give exactly this confinement pressure (the measures are 66 % of the value calculed by ribs strains).

The process permits rock mass movements without appearence of too large plastic strain (the lenght of the plastic zone reaches 5 meters from gallery's wall).

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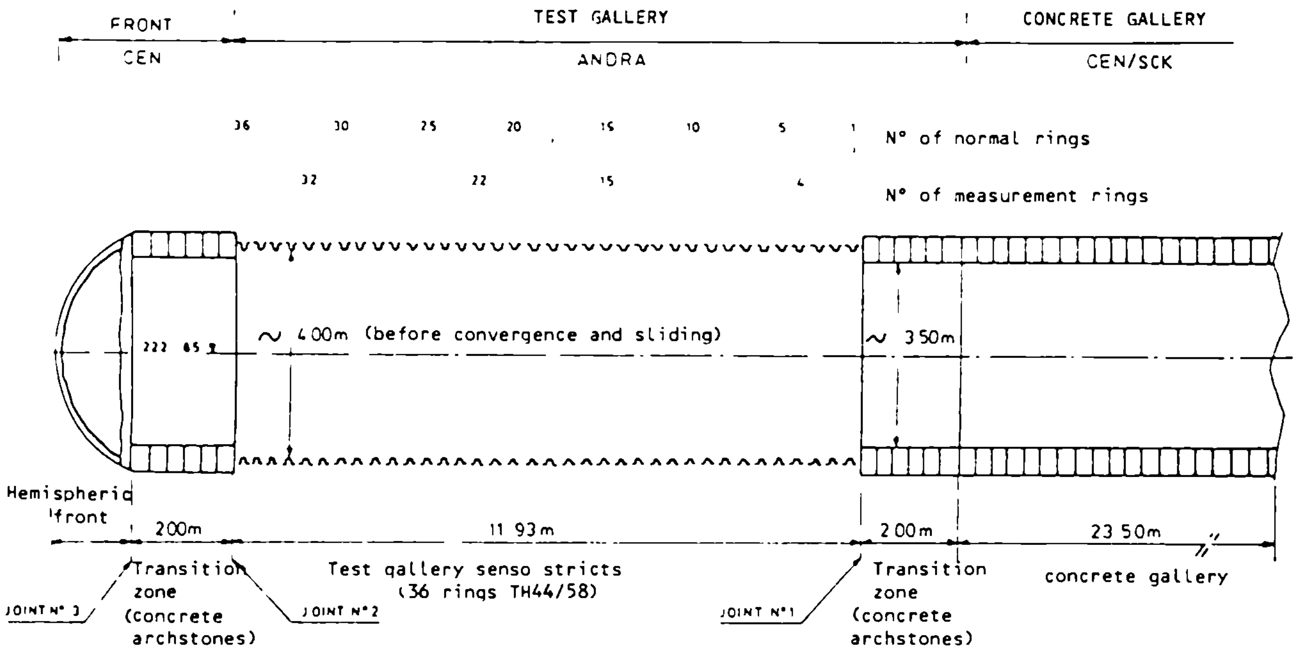


Fig. 1 : ANDRA TEST GALLERY

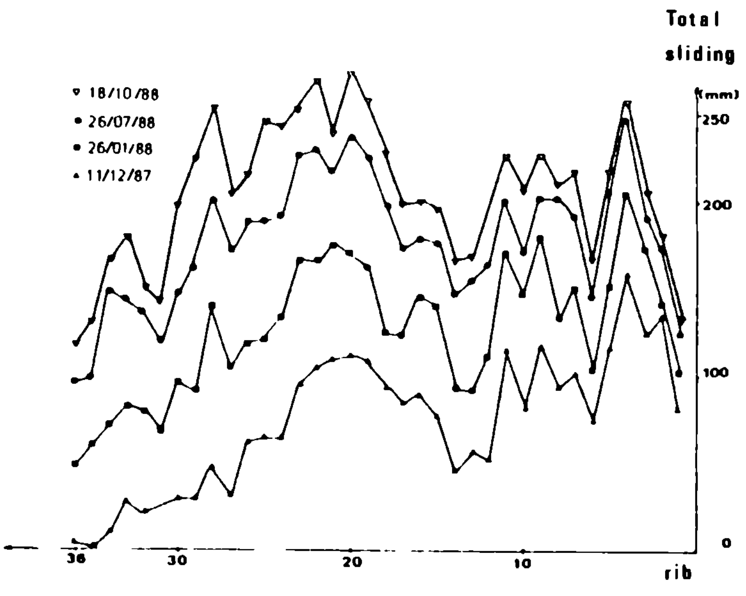


Fig. 2 : AVERAGE SLIDING

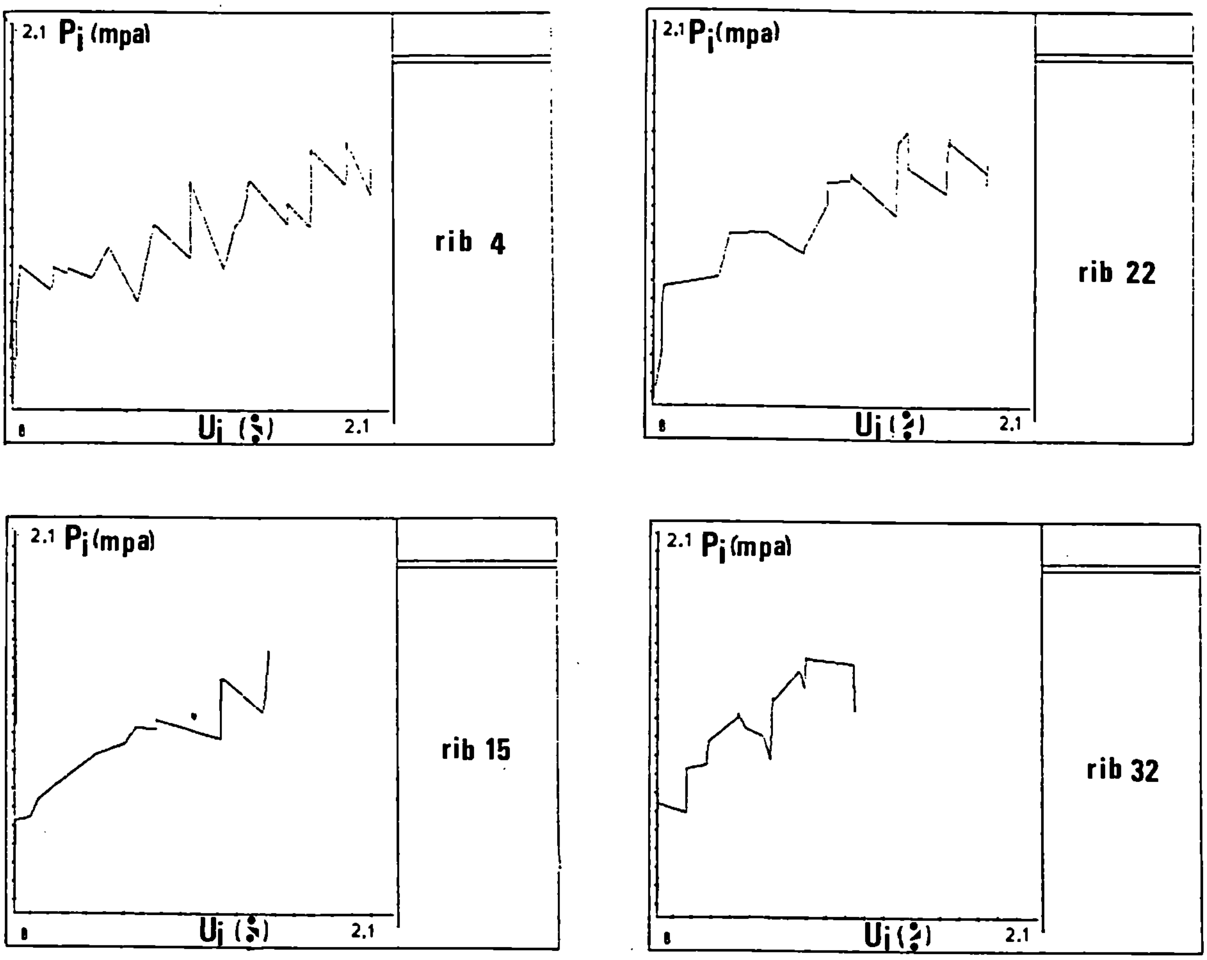


Fig. 3 : EXPERIMENTAL CONFINEMENT CURVES

REPOSITORY TUNNEL CONSTRUCTION IN DEEP CLAY FORMATIONS

Contractor : Geotechnical Consulting Group, London, UK
Contract No. : FI1W/0200
Duration of contract : February 1988 - January 1991
Period covered : February 1988 - December 1988
Project leader : Dr R J Mair

A. OBJECTIVES AND SCOPE

At Mol, Belgium, the feasibility of constructing a repository at large depth in a clay formation is being investigated (HADES project). The most recent construction has been a test drift of a length of 40 m and an internal diameter of 3.5 m: 60 cm thick concrete block segments were used for linings. As an extension to the test drift, a 12 m long gallery has been constructed using sliding steel ribs as linings. Extensive measurements of lining loads, pore-pressures and ground movements have been made in the test drift.

The objectives of this project are as follows:

(i) Analyse and interpret available measurements made at the HADES project at Mol in respect of tunnel construction.

(ii) Carry out laboratory stress path testing on samples of Boom clay taken during the construction of the test drift at Mol.

(iii) Research and develop a self-boring retracting pressuremeter (SBRP) as an in-situ testing device for use in hard clays, with specific reference to tunnel lining design.

B. WORK PROGRAMME

The work programme consists of the following activities:

1. Data Analysis and Interpretation: (a) synthesis of data (b) plasticity calculations (c) finite element analyses (d) interpretation and generalization of data in respect of possible deep repository construction in clay formations.

2. Laboratory testing: (a) development of a high pressure computer contolled stress path triaxial testing apparatus (b) testing of samples of Boom clay.

3. In-situ Testing:

3.1 Develop a prototype self-boring retracting pressuremeter.

3.2 Carry out field tests in the UK with the SBRP in appropriate geological materials such as mudstones.

3.3 Depending on results, carry out tests from the gallery or test drift at Mol.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The available measurements made at Mol on lining loads, pore-pressure and ground movements have been synthesized. Plasticity calculations and finite element analysis have been undertaken of an advancing tunnel heading at great depth in a clay formation corresponding to the test drift construction at Mol. Good agreement has been obtained between the plasticity solutions and finite element analysis. The measured immediate build-up of stress on the linings in the test drift is well predicted. Further finite element analyses will be undertaken with effective stress soil models to investigate the effects of different soil behaviour and the long-term build-up of radial stress on the linings. The high pressure computer controlled stress path triaxial testing apparatus has been designed and assembled, and pilot tests on Boom clay samples undertaken. The SBRP has been designed, working drawings produced and the first prototype instrument manufactured. Field tests will be undertaken in appropriate geological materials in the UK, such as mudstones, but not at Mol as originally considered.

PROGRESS AND RESULTS

1. All the available measurements for the small experimental shaft, the experimental gallery (2.1 m OD) and the test drift have been assembled and examined. Emphasis has been placed on the measurements made for the test drift.

The analyses have aimed at interpreting and understanding the behaviour of an advancing tunnel heading in clay, as shown in Figure 1. It is assumed that the rate of advancement of the tunnel is sufficiently fast that the clay behaviour around the heading is undrained. There is then a build-up of pressure on the lining under undrained conditions from zero when it is installed to a maximum value at some distance back from the face. Plasticity calculations and finite element (FE) analysis have been performed assuming elastic-perfectly plastic soil behaviour in terms of total stresses (i.e. using Young's modulus, E_u, and undrained shear strength, c_u). The plasticity calculations have been based on the simplifying assumption that a tunnel heading can be approximated as one half of a thick sphere; cylindrical symmetry is used for conditions remote from the tunnel face. For the FE analysis, axisymmetric conditions were assumed and the actual test drift construction sequence was modelled as closely as possible. The soil parameters adopted were based on an assessment of the currently available laboratory test results on Boom clay.

The predicted build-up of lining pressure with time from the FE analysis is shown on Figure 2, together with the pressure inferred from load cell measurements. The predicted pressures from plasticity calculations A and B are also shown, and they are also in reasonable agreement with the measurements. Calculation A assumed the same input parameters as the FE analysis; calculation B assumed a lining stiffness reduced by a factor of 2.35. The reduction in assumed lining stiffness by a factor of 2.35 leads to a change in predicted lining pressure of only 7%.

Lining convergence and ground movements can also be reasonably predicted if the appropriate stiffness of the lining is used in the calculations. The stiffness of the test drift lining at Mol is dominated by the wood packing between the concrete blocks, and it appears that its stiffness may be reducing with time.

2. A high pressure computer controlled stress path triaxial testing apparatus has been designed and assembled at The City University. Tests will be performed on samples of Boom clay recovered from the test drift excavation at Mol. Appropriate levels of effective stress are up to about 2.5 MPa and the triaxial cell has been designed for pressure up to 5 MPa. The apparatus is capable of following compression and extension stress paths.

The triaxial cell has been mounted in a 5 tonne loading frame fitted with a stepless strain controller. Pore water pressure and volume transducers have been fitted. A double acting hydraulic piston is used to apply the stress controlled axial loading. Cell and piston pressures are achieved using hydraulic pressure multipliers, which operate from a low air pressure compressed air supply. A system of metering valves and accumulators ensures a smooth control and application of hydraulic pressures. Various commissioning tests have been conducted and a number of pilot tests on Boom clay samples have been successfully completed.

3. The design and construction of the first prototype SBRP has been completed at the Univesity of Newcastle upon Tyne. Its purpose is to measure in-situ stiffness and ground response of soil and weak rock when they are unloaded from in-situ stress conditions, with specific reference to shaft or tunnel lining design. To achieve this the instrument will be installed with minimum disturbance to the surrounding ground by a self-boring process; it then must be capable of being retracted under controlled conditions so that measurements can be taken of the change in stress and associated displacement.

A mechanically supported membrane system has been developed operating on a principle of longitudinal wedges to ensure that the instrument remains rigid during installation. The pressure from the soil is measured at several points in the retracting section of the instrument. The length to diameter ratio of the retracting system is 5:1 to ensure plane strain conditions over a significant length of the membrane. The diameter is 84 mm. The self-boring drilling system has been developed from previous work at the University of Newcastle upon Tyne with an expanding self-boring pressuremeter (SBP), which has been successfully used in hard clays and weak rocks.

The first prototype, which has been manufactured, is made of brass and is designed to operate at low stresses in soil. Its purpose is to test the principle, assess the tolerances and method of operation. A second instrument, made out of stainless steel and designed to operate at stresses in excess of 6 MPa, will incorporate any design changes made as a result of experiences with the first.

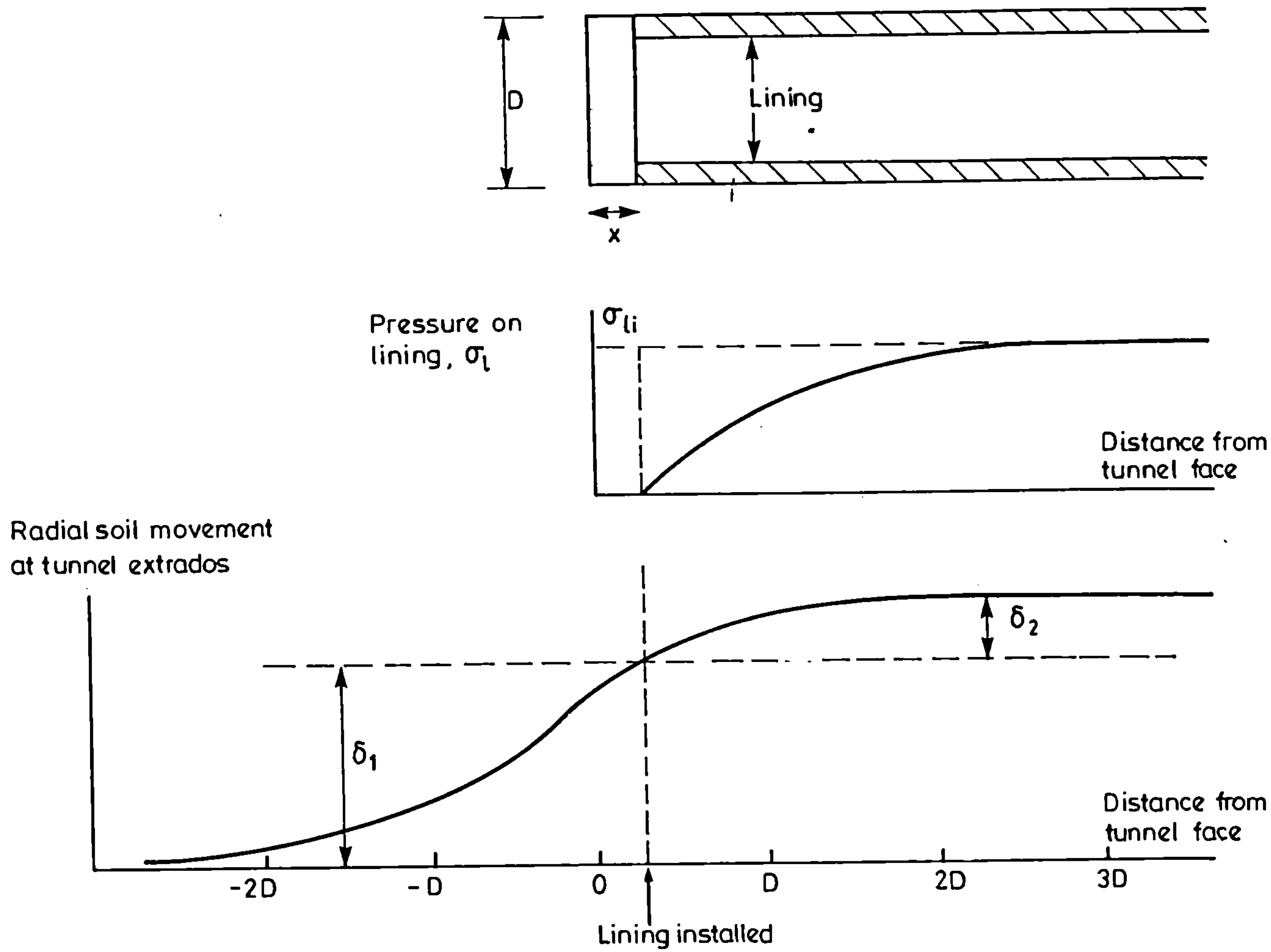
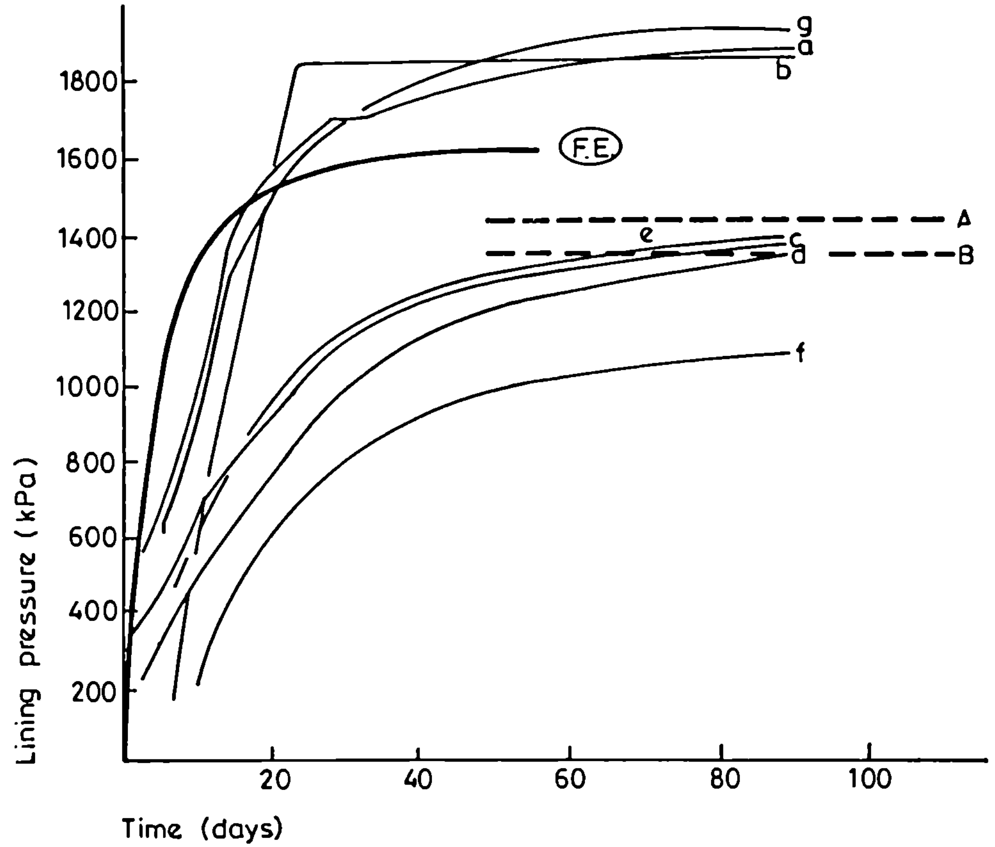


FIGURE 1 - Behaviour of an advancing tunnel heading in clay



	Ring	Load cell
a b	Ring 15	15 h 31 v
c d	Ring 43	15h 24
e f g	Ring 105	16h 48h 64v

Plasticity Calculations
- A - using same input parameters as F.E.
- B - using reduced stiffness for lining

FIGURE 2 - Predicted build-up of lining pressure for the test drift at Mol compared with measurements

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