Technical University of Denmark

Nuclear Safety Research and Facilities Department annual report 1996

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Publication date:
1997

Document Version
Publisher's PDF, also known as Version of record

Risø-R-984(EN)

Nuclear Safety Research and Facilities Department Annual Report 1996

Edited by B. Majborn, K. Brodersen, A. Damkjær, H. Floto, K. Heydorn and P.L. Ølgaard

Risø National Laboratory, Roskilde
April 1997

Abstract The report presents a summary of the work of the Nuclear Safety Research and Facilities Department in 1996. The department's research and development activities are organized in three research programmes: Radiation Protection, Reactor Safety, and Radioanalytical Chemistry. The nuclear facilities operated by the department include the Research Reactor DR3, the Isotope Laboratory, the Waste Treatment Plant, and the Educational Reactor DR1. Lists of staff and publications are included together with a summary of the staff 's participation in national and international committees.

Frontpage illustration:

Research Reactor DR 3 seen from Roskilde Fiord
(Photo: P. Winstrøm)

ISBN 87-550-2303-7
ISSN 0106-2840
ISSN 1396-3619

Information Service Department, Risø, 1997

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1 Introduction

The Nuclear Saftety Research and Facilities Department is engaged in research and in the operation of the nuclear facilities at Risø National Laboratory.

In 1996 the department's research and development activities were organized in three research programmes: Radiation Protection, Reactor Safety, and Radioanalytical Chemistry. On January 1, 1997, the research programme "Radioecology" were transferred to the department from the Environmental Science and Technology Department. Hence closer relations has been established between the research programmes at Risø covering nuclear safety, radiation protection, radioecology, and the use of nuclear methods. The nuclear facilities operated by the department include the Research Reactor DR3, the Isotope Laboratory, the Waste Treatment Plant, and the Educational Reactor DR1.

The research and development work of the department is carried out in close cooperation with Danish and foreign universities and research institutes and also with the Danish nuclear and radiation protection authorities: The Emergency Management Agency and the National Institute of Radiation Hygiene. The department participates in national and international research programmes including some European Commission programmes and the Nordic Nuclear Safety Research Programme.

This report describes the work of the department in 1996 with an emphasis on the results of the research and development activities. Lists of staff and publications are included together with a summary of the staff's participation in national and international committees

2 Radiation Protection

2.1 Dosimetry

Personnel Dosimetry

Risø's personnel dosimetry service covers the individual monitoring of the personnel at Risø and the Niels Bohr Institute Tandem Accelerator. Only persons actually involved in radiation work are equipped with a personal dosemeter. In areas where the use of personal dosemeters are not required, the radiation levels are controlled through an extensive area-monitoring programme using thermoluminescence (TL) dosemeters.

The main statistics of the dosimetry service for 1996 are shown in Table 2.1 and Figure 2.1.

Table 2.1. Statistics for monitoring of Risø Personnel in 1996.

No. of persons monitored	728
No. of persons receiving external doses above 0.2 mSv (the registration level)	163
No. of persons receiving internal doses from intake of tritiated water	58
Total collective equivalent dose to the monitored personnel: External doses Internal doses Total	 225 mSv 28 mSv 253 mSv

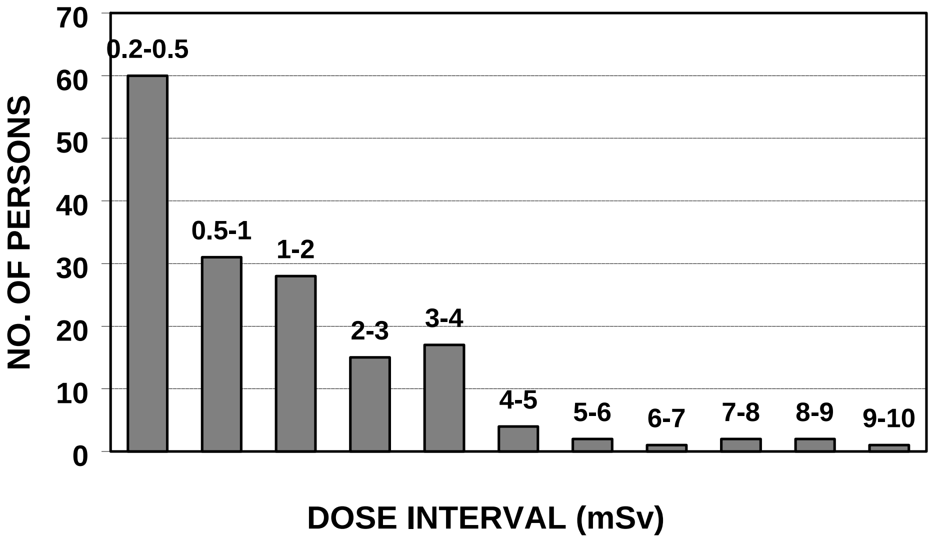

Figure 2.1 Distribution of whole-body doses (effective doses) in 1996.

Initiatives were taken to introduce TL glow-curve analyses in the routine personnel monitoring programme. The method is useful for identification of false background dose readings caused by abnormal glow curve structures. A software package (TL-Data Saver) developed by the Rados Company for dosimetry applications using the Alnor reader was used to create a database with TL glow curves, representative for routine monitoring. The database will be tested in a glow curve analysis program developed by Dosimetry Services Company, USA, who will also adapt the software for routine dosimetry applications based on the Alnor reader.

Dosimetry of Beta and Low-Energy Photon Radiation

Work has continued in developing techniques for dosimetry of low-penetrating radiation. The group participates in a new three-year EU research project covering various aspects of dosimetry of weakly penetrating radiation, i.e. measurement of personal photon dose equivalents, beta dose measurements based on beta spectroscopy, standard dosimetry based on the extrapolation chamber measurement method, and determination of dose rates around highly radioactive particles (hot-particle dosimetry). In collaboration with CEA, Fontenay-aux-Roses, France, the responses of TL dosemeters to exposures from low-energy photon radiation have been studied. For dosimetry studies of beta radiation fields it is important to know the dose rate distribution over the area of interest. Mappings of dose rate distributions have been made using a phosphor imaging system recently acquired by the Solid State Physics Department, Risø.

Beta Spectroscopy

Within the joint European research project on dosimetry of weakly penetrating radiation, Risø has the main responsibility for developing a beta spectrometer device capable of measuring skin doses in mixed beta/photon radiation fields. In addition to dose determination the device will also provide information on the beta spectrum to decide on adequate shielding or to identify the radionuclide. The beta spectrometer has been designed and is presently being produced in the Engineering and Computer Department at Risø.

The detector unit contains three silicon detectors of different thicknesses arranged in a telescope unit. By using two thin front detectors and a thick back detector the photon-induced signals will be rejected on coincidence requirements for all three detectors thus enabling beta dose determinations to be made in mixed beta/photon radiation fields.

The portable beta spectrometer consists of 3 distinct hardware units: Detector assembly (DA), processing and support unit (PSU) and laptop computer. Figure 2.2 shows a graphical view of the set-up. The laptop computer executes a control, monitoring, and displaying program. The detector assembly contains the detector unit together with thermo-electric cooling elements, bias supplies, preamplifiers and charge generation for calibration.

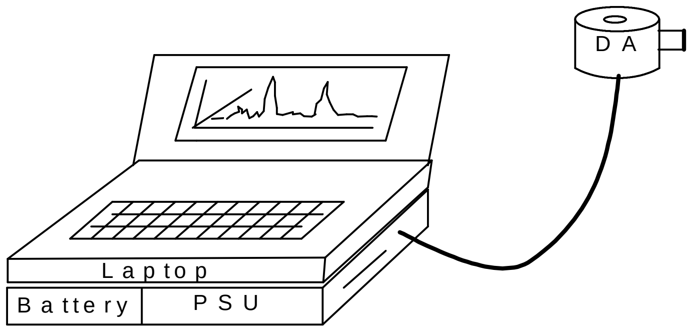

Figure 2.2. View of portable beta spectrometer.

During a preliminary study, a number of specific parameters connected to beta spectroscopy with Si detectors were investigated. The results of the study provided important information on the influence of backscattering from the Si detector on the spectrum and on various factors influencing the noise level of the detector. The results further emphasised the importance of determining the spectra of beta radiation fields used for calibration purposes.

Monte Carlo (MC) calculations have been applied to study the energy spectra obtained by the three Si detectors of the beta telescope spectrometer. The results have been used for studying specific design parameters for the detector unit and will at a later stage be compared with experimental data. MC calculations using the EGS4 code with the PRESTA algorithm were performed using the model of the detector shown in Figure 2.3. Monoenergetic parallel electron beams incident from the front (source radius same as detector radius) with energies ranging from 25 keV to 3.5 MeV were simulated. The energy losses of each incoming electron in each of the three Si detector layers were calculated using the user code DOSRZ. From these energy losses the energy and angle of incidence of the electron will be calculated in order to estimate the dose at a given depth. Figure 2.4 presents examples of energy spectra obtained from the MC calculations.

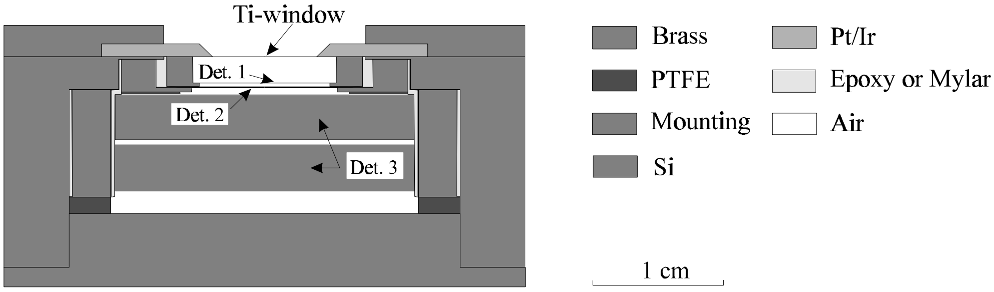

Figure 2.3. Detector model used in MC simulations. The three Si detectors have tichness 50 μm, 150 μm, and 2x3500 μm, and areaes 50 mm^2, 100 mm^2 and 300 mm^2, respectively.

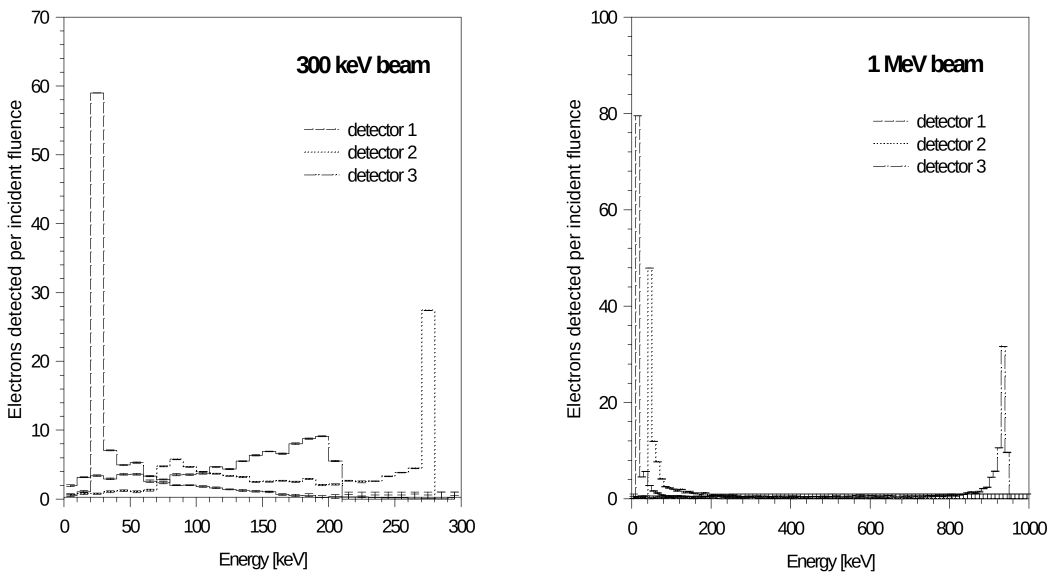

Figure 2.4. Energy-loss distributions in the three Si detectors calculated for300 keV and 1 MeV electron beams

Distribution of Dose rate in Beta Radiation Fields

The Molecular Dynamics Model 400 Series PhosphorImager^{TM} laser scanning device recently acquired by the Solid State Physics Department was used for measurements of the dose rate distribution of the beta radiation field from a

^{147}Pm source. The storage phosphor screen (imaging plate IP) is a position sensitive detector with a dimension of 20 x 25 cm^2. The resolution of the scanned image is 88 x 88 µm^2 , and the thickness of the phosphor layer is 40 mg cm^{-2}.

Two IPs have been tested, one having a protective coating of 1.1 mg cm^{-2} and the other without any coating. It is concluded that the coating is of minor importance compared to the thickness of the phosphor layer (see Figure 2.5). Hence, the IP with coating and high sensitivity will be used for further studies. The dose range found for this IP is from 40 nGy to 650 mGy. The preliminary results obtained so far indicate that the imaging system presents a promising tool for determination of dose homogeneity of extended beta radiation fields.

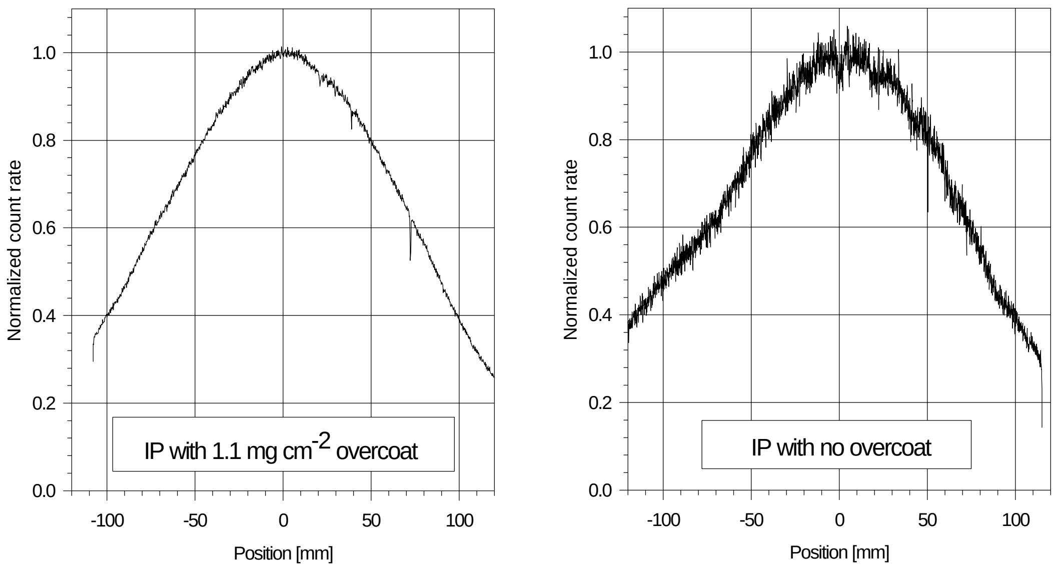

Figure 2.5. Dose distribution measured with the IPs with and without protective coating. The radiation field was from a ^{147}Pm source at 20 cm distance. Maximum dose for the plate was 52 mGy with coating and 348 mGy without coating.

Responses of TL detectors to Low-Energy Photon Radiation

In recent years low-energy photon radiation from synchrotron radiation facilities has been applied in various research fields, e.g. physics, biology, chemistry, medicine and technology. The rapidly growing number of persons engaged in experiments with low-energy photon radiation increases the need for accurate dosimetry in this field. In the present study the responses for a number of thin TL detectors for skin dose ($H_P(0.07)$) from exposure to low-energy photon radiation were measured. The irradiation was carried out at the Danish National Institute of Radiation Hygiene (25, 33 and 48 keV, ISO Narrow Spectrum Series), at CEA Fontenay-aux-Roses France (6.5, 8.5 and 12.2 keV from a low-energy X-ray facility) and at Risø (6 keV from a ^{55}Fe beam). As seen from Figure 2.6, detectors prepared from LiF:Mg,Cu,P significantly underestimate $H_P(0.07)$ doses from photons with energies below 10 keV, in contrast to LiF TLD-700 and MgB_4O_7:Dy detectors which overestimate the dose in this energy range.

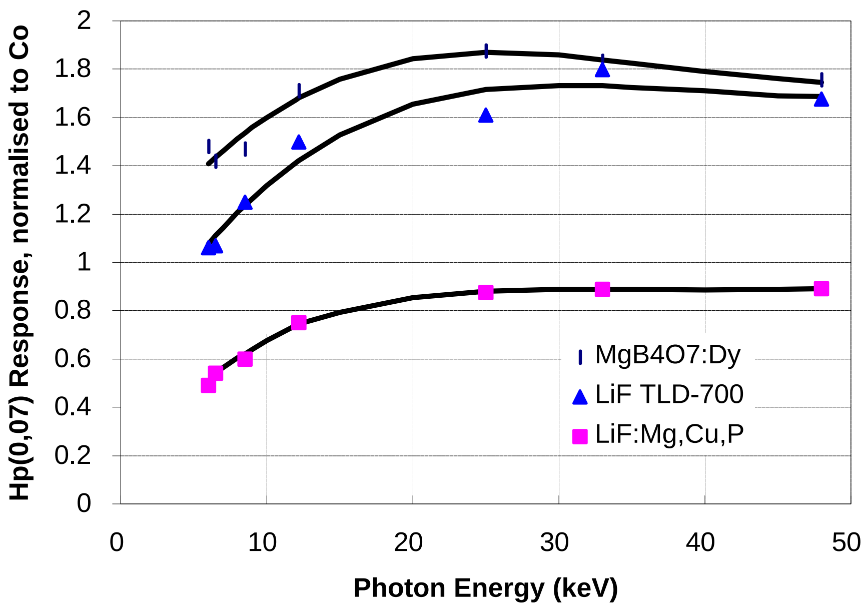

Figure 2.6. Response of three types of thin TL detectors covered with a 6 mg.cm^{-2} Mylar filter to exposures from low-energy photon radiation. Responses are expressed in terms of $H_P(0.07)$ response relative to that from ^{60}Co radiation. Detector thicknesses are: MgB_4O_7:Dy : 2-3 mg.cm^{-2}, LiF TLD-700 : 20 mg.cm^{-2} and LiF:Mg,Cu,P : 5 mg.cm^{-2}.

Calibration of Gamma Cells

Two gamma Cells established at Odense Sygehus, each containing about 1500 Ci ^{137}Cs were re-calibrated using LiF TLD-700 TL detectors. The dose rate distribution for irradiation of blood in the irradiation container was determined by irradiating the LiF detectors in a polystyrene phantom as illustrated in Figure 2.7. The results showed deviations from the previous calibrations of the

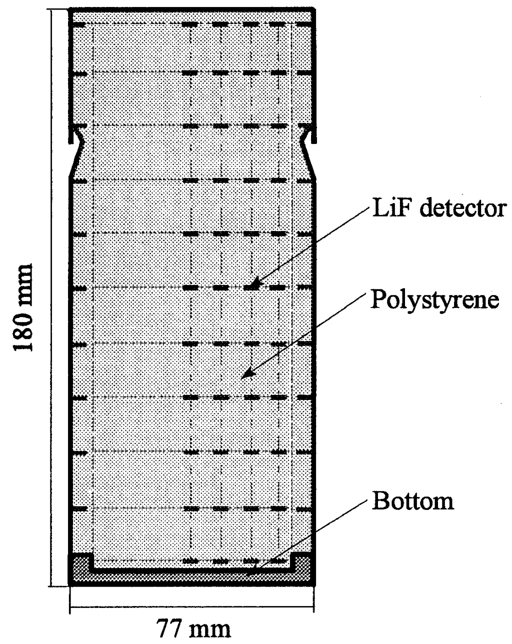

Figure 2.7.Sketch of irradiation container illustrating the positions of the LiF TL detectors in a polystyrene phantom simulating irradiation of blood samples in the container

two gamma cells of less than 1% and 4%, respectively.

Control of Irradiated Spices

The dosimetry group assisted the FDB company with TL measurements of three selected samples of spices for irradiation control.

2.2 Development of Instruments and Methods for Optically Stimulated Luminescence (OSL)

The intensive programme on the development of OSL instruments and methods has continued. The programme has involved the implementation of new optical systems, improved mechanical components, a more efficient irradiation unit, and new computer hardware and software for control and operation of the automated TL/OSL reader. The new improved OSL instrument configuration has been used for investigations of OSL characteristics of Al_2O_3:C and quarts materials and for retrospective dose determination using ceramic materials collected in Chernobyl.

The Development of New Luminescence Detection Systems.

Two new OSL detection units, using ellipsoidal mirrors as reflector for the emitted light, were developed and tested. The first one, depicted in Figure 2.8, is referred to as the hybrid system, since this uses many features that are common to the existing OSL excitation unit. This system uses only a partial reflector. The second one, the full reflector system, is shown in Figure 2.9. It uses a full ellipsoidal mirror with the OSL excitation light being introduced from an external source through light guides.

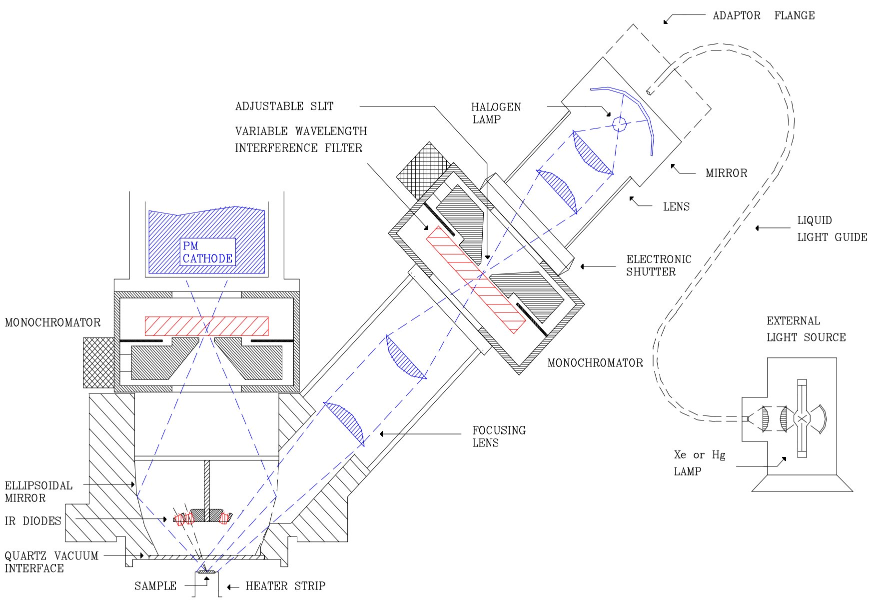

Figure 2.8. Schematic diagram of the hybrid system with optional excitation and emission monochromators. A small section of the ellipsoidal mirror has been cut away to allow for optical stimulation using a halogen lamp.

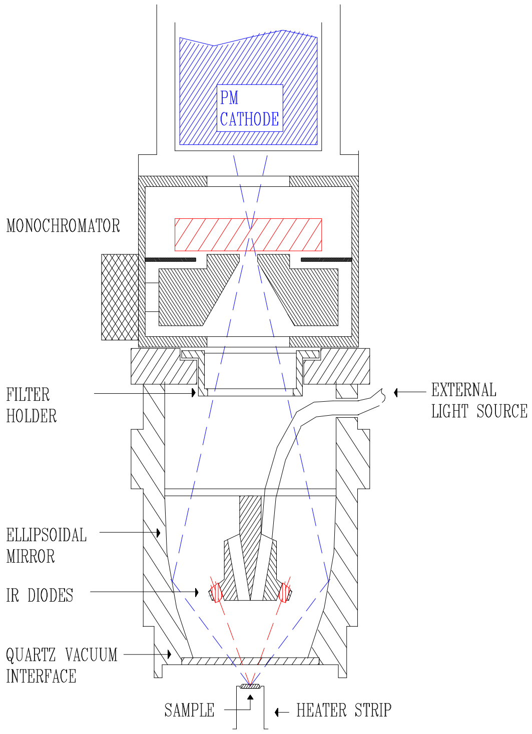

Figure 2.9. Schematic diagram of the full reflector system. In this system visible excitation is introduced by up to four optional light guides.

The light sources and excitation efficiencies of the two systems are described below. Both designs have the sample placed at one focal point of the mirror, while the second focal point directly corresponds to the position of either the PM cathode or, if installed, the entrance slit of the Risø designed compact monochromator. In this way, no secondary optics are required.

The Hybrid System

In this system a module with an array of 23 LEDs can be placed above the sample (Figure 2.8). This will allow IRSL measurements to be made in addition to the OSL measurements with green/blue light. The solid angle occupied by the LED module is small and decreases the effective sensitivity by only 15%.

In the hybrid system, the excitation source (75W tungsten-halogen lamp) and geometry are the same as in the standard Risø OSL unit. The efficiency of excitation is thus unchanged, and the compact monochromators of the standard unit can be attached for measurement of luminescence excitation spectra. The standard OSL system also incorporates an array of 13 diodes, clustered around the sample.

The Full Reflector System

This system (Figure 2.9), has a collection efficiency comparable to that of the hybrid system. The central light guide holder contains an array of 15 IR LEDs. At first sight this would not appear optimal, but the IR diodes are closer to the sample thus compensating for the fewer LEDs as compared to the hybrid system. The advantage of the system is the greater flexibility. Provision is made to accommodate up to four 3 mm diameter light guides, each of which can be coupled to different excitation sources.

The liquid light guide system
As a less expensive alternative to the ellipsoidal mirror systems a new compact combined infra red /green light stimulation light (IRSL/GLSL) unit with a much improved sample-to-PM tube distance has been developed. A significantly enhanced GLSL sensitivity is achieved using an eight mm diameter liquid light guide system with high transmission (98% over 380 - 550 nm) for illumination of the sample. Filtered wavelength bands are provided using either a 150 W tungsten halogen lamp (lifetime: 2000 hours) or a 150 W xenon lamp mounted in a remote lamp house equipped with electronic shutter and exchangeable filter pack. The liquid light guide OSL unit uses quartz lenses for defocusing the stimulation light to ensure that it falls uniformly on the sample. IRSL is performed using IR LEDs in a close position to the sample. The unit focuses the emitted luminescence onto the photo cathode using a quartz lens with short focal length. The liquid light guide system is shown schematically in Figure 2.10.

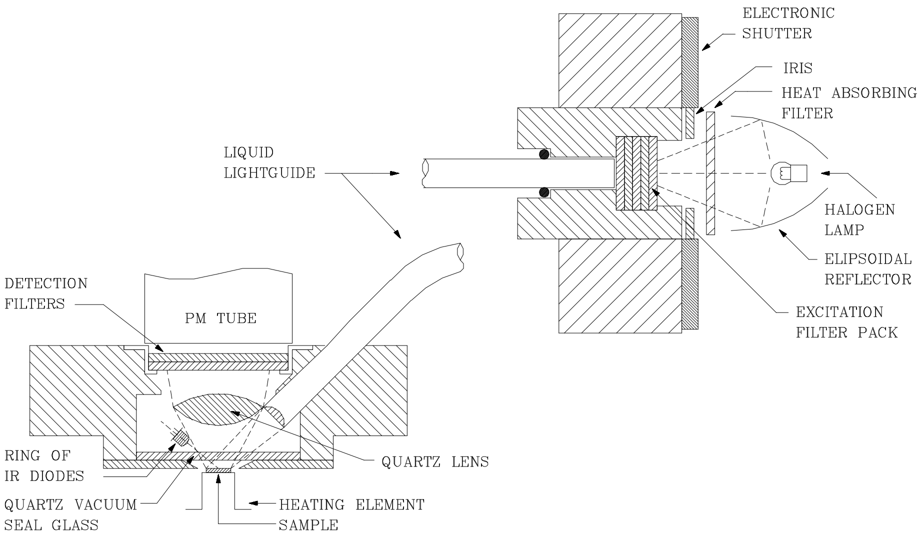

Figure 2.10. Schematic diagram of the liquid light guide OSL unit

Measurements and results
Several measurements were performed to determine the increased collection efficiency of the new systems as well as the usefulness of the new external excitation sources. All measurements were made using an EMI 9635Q photomultiplier tube in combination with 6 mm of Hoya U-340 filter. The results show that the hybrid and full reflector systems (without the IR diode assembly) have collection efficiencies, relative to the standard system, of 306% and 345%, respectively. With the IR diode assembly and two light guides inserted in the full reflector system there is still a relative signal of 241%. The results for the liquid light guide system showed a factor of 4 to 5 in improved sensitivity as compared to the standard Risø OSL system.

Sample Turntable

A two speed turntable has been added to the system in order to reduce processing time. If the turntable is moved from one sample position to the adjacent position, the turntable rotates with normal speed. If, however, the turntable advances several positions, it is accelerated to a higher speed. Investigations have confirmed that the acceleration and deceleration are sufficiently gentle to leave small grains undisturbed. It is possible, though, to disable the high speed feature.

In addition, the sample capacity has been increased from 24 samples to either 36 or 48 samples. All three turntables are fully interchangeable so the new, high capacity turntables may also be used in older systems.

Detachable Irradiation Unit with Sr/Y-90 Beta Source

A completely new Sr/Y-90 beta irradiator attachment was developed in which the source is mounted in a rotating, stainless steel wheel as shown in Figure 2.11. This design allows the source to be positioned closer to the sample than previously, thus increasing the dose rate by more than a factor of two. Also, the external radiation level has been reduced. In addition, the dose received by samples under the closed irradiator has been reduced dramatically.

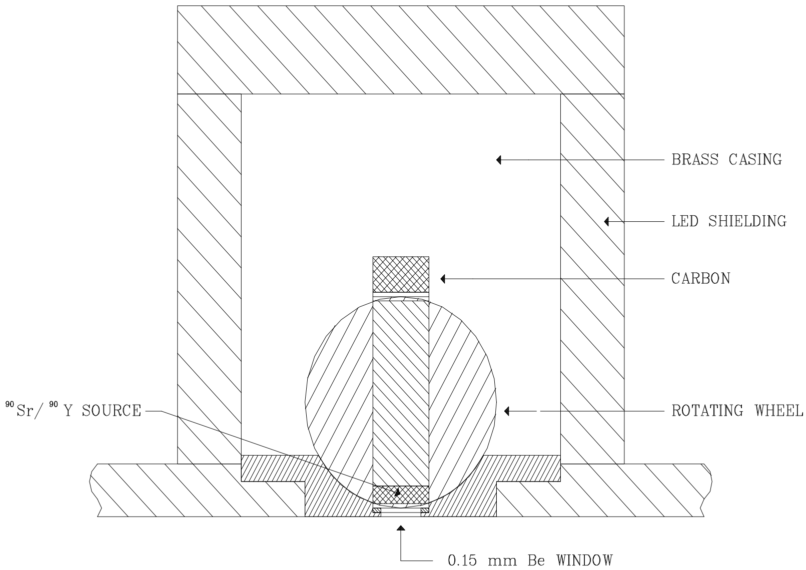

Figure 2.11. Cross section of the new irradiator design. The source is placed in a rotating stainless steel wheel as shown. When not irradiating, the wheel is rotated 180 degrees.

Hardware

The control of timing, sample positioning, data acquisition, error checking etc. of the new automated TL/OSL reader has been incorporated into a "Mini-Sys" computer which is a 386 based PC unit with 2MB flash disk and 2MB RAM. The Mini-Sys connects to the host computer via an RS-232 serial cable thus eliminating the need for an expansion board in the host PC. The Mini-Sys computer is equipped with a two line digital text display showing the system status and the command which is currently being executed (e.g. "Recording OSL..."). This display also reports failure messages.

An expansion board may be added to the Mini-Sys providing access to 16 digital input channels and 8 output relays. By sending appropriate commands to the Mini-Sys it can be instructed to activate one or more relays or wait for a specific input before proceeding. This feature adds the ability to interface the reader with other laboratory equipment such as lasers and flash lamps for illumination. Also, it is possible to use external equipment to trigger the Mini-Sys to start data acquisition.

Software
The response received from a large number of questionnaires sent to users of the Risø TL/OSL system, has resulted in a completely re-designed software package for the Mini-Sys PC. Control software for creating, editing and executing TL/OSL sequences will be available for both DOS and Windows. The Windows version has been expanded to include several common methods of analysis applicable to dosimetry and dating including TL glow curve and OSL decay curve analysis and plotting and fitting facilities.

The central feature of the Mini-Sys software is a high level command language interpreter. Approximately 40 commands allow full control of system hardware and data acquisition methods. The system allows users to write their own programs in PASCAL, BASIC, C, etc., to perform unlimited variations of the standard experiments included in the user application software.

Other features and capabilities of the new software include 1) monitoring real sample temperature, 2) monitoring room temperature, 3) the ability to perform flash heating in which the heater temperature is set before the sample is raised, 4) substantial enhancement of features for creating measurement sequences and 5) significantly enhanced flexibility in data file handling and the accommodation of data file information.

Two new and powerful data acquisition methods have been added to the standard set of TL and OSL commands: Thermo-Optically stimulated Luminescence (TOL) in which the illumination is pulsed during a TL measurement and Pulsed Optically Stimulated Luminescence (POSL) in which the OSL decay is measured following an optical pulse.

OSL Characterisation of Materials

Al_2O_3:C as a Sensitive OSL Environmental Dosemeter
The introduction of Al_2O_3:C as a material for TL dosimetry has opened the possibility of several promising applications for ultra-high sensitivity dosimetry, particularly for short-term exposure in personal, environmental and extremity dosimetry. The indications for this material are that it possesses a TL sensitivity some 40-60 times greater than that of LiF TLD-100 making it a strong candidate for low dose applications. However, thermal quenching processes in the material cause the TL sensitivity to depend strongly on the heating rate used, with less sensitivity obtained at higher heating rates. This feature can be a disadvantage in routine TL dosimetry where fast heating automatic readers are applied. Another potential limitation in the use of this material is its sensitivity to light. Light sensitivity manifests itself in three ways: (i) a light-induced fading of the TL signal; (ii) the generation of a TL signal in unirradiated samples, and (iii) the phototransfer of charge from deep states to shallower states giving rise to a phototransferred TL (PTTL) signal.

The light-induced fading of the TL signal from Al_2O_3:C and the subsequent phototransferred TL (PTTL) into shallow traps indicate the potential of this material as a sensitive OSL dosemeter. An even higher sensitivity, i.e. emitted luminescence photons per unit radiation dose , is predicted for OSL as compared to TL. Given that Al_2O_3:C is already one of the most sensitive TL materials, the OSL sensitivity promises to be exceptionally high. The stimulation spectrum for Al_2O_3:C shown in figure 2.12 was obtained using the Risø developed monochromator in the wavelength range 400 - 700 nm.

Typical OSL decay curves for Al_2O_3:C exposed to ^{60}Co radiation and stimulated with a wavelength band 420-550 nm are shown in Figure 2.13. Preliminary investigations of the OSL sensitivity of Al_2O_3:C have shown that a dose of 1 micro Sv (10 hours of exposure to the natural background radiation) can easily be detected. The dose response of either continuos wave or pulsed OSL reveals a linear relationship up to about 50 Gy. Examinations of the OSL signal over an interval of 80 days following irradiation have revealed no measurable fading of the signal over that period.

Comparison of OSL from Quartz Using Different Stimulation Wavelengths
The OSL characterisation of quarts has continued. An experiment was designed to measure the extent to which the OSL signal, as measured using a variety of stimulation wavelengths, is bleached using light at 514 ± 25 nm. This bleaching wavelength was chosen to match that from an argon ion laser, and the experiment aimed to see whether the bleaching decay rate for the luminescence signal varied as a function of the stimulation wavelength.

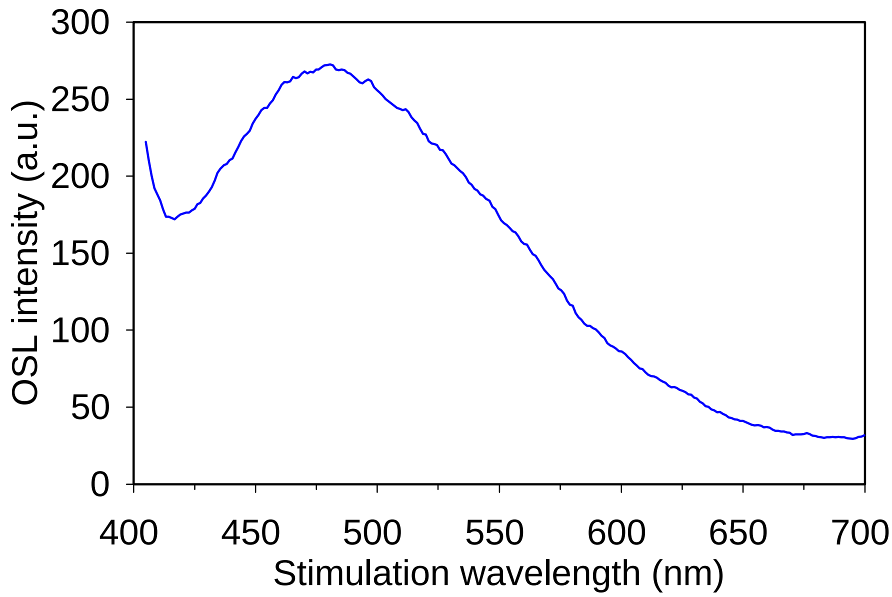

Figure 2.12. Stimulation spectrum for Al_2O_3:C obtained with the Risø developed monochromator.

After heating to 450 deg.C, irradiating to a beta dose of 10.8 Gy and preheating at 220 deg.C for 300 s, the quartz sample was exposed to 514 ± 25 nm stimulation light for time periods of 0 to 3200 s (power at the sample was 0.4 mW cm^{-2}). A stimulation spectrum from 400 to 650 nm was then measured (Fig. 2.14). The same sample aliquot was used for all measurements, and the signal level corresponding to no irradiation was subtracted from all measurements. At wavelengths below 425 nm the data are affected by cross talk between the stimulation and detection wavelengths. It should be noted that the stimulation spectra in Fig. 2.14 have not been corrected for the variation in stimulation power since the aim of the measurements was to compare the loss of signal at various wavelengths. Figure 2.15 shows the data from Fig. 2.14 as bleaching curves, at wavelength intervals of 25 nm. This shows that the OSL signal from this quartz sample is equally reduced by exposure to 514 nm light over the stimulation range from 450 to 610 nm. The similarity in these bleaching decay curves suggests that over the range from 450 to 610 nm the OSL measurements must be probing a similar group of traps. The change in behaviour seen at shorter stimulation wavelengths (<450 nm) is thought to be due to some degree of cross talk between the stimulation and detection wavelengths.

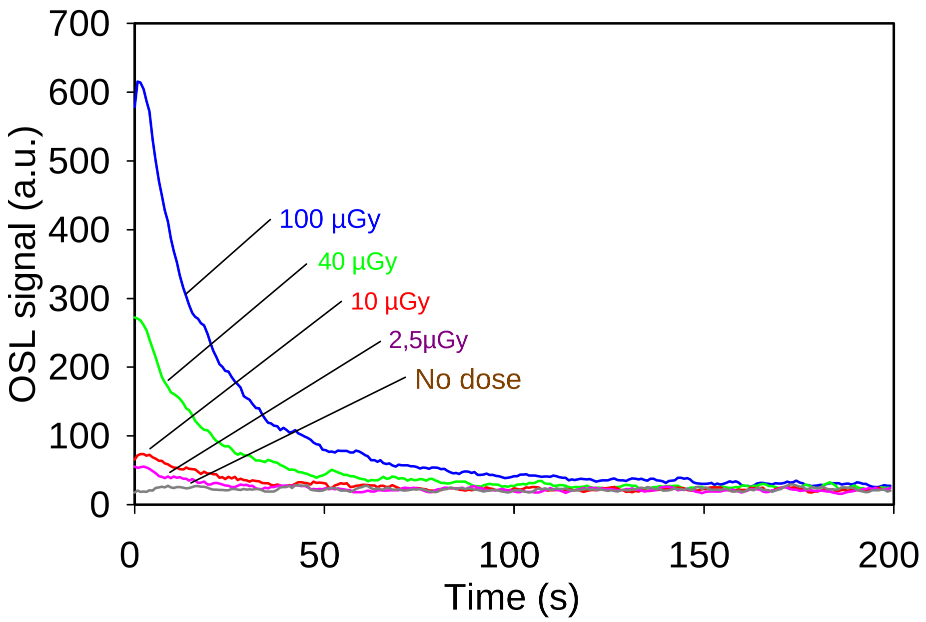

Figure 2.13. OSL decay curves for Al_2O_3:C exposed to ^{60}Co radiation and stimulated with a wavelength band 420 - 550 nm.

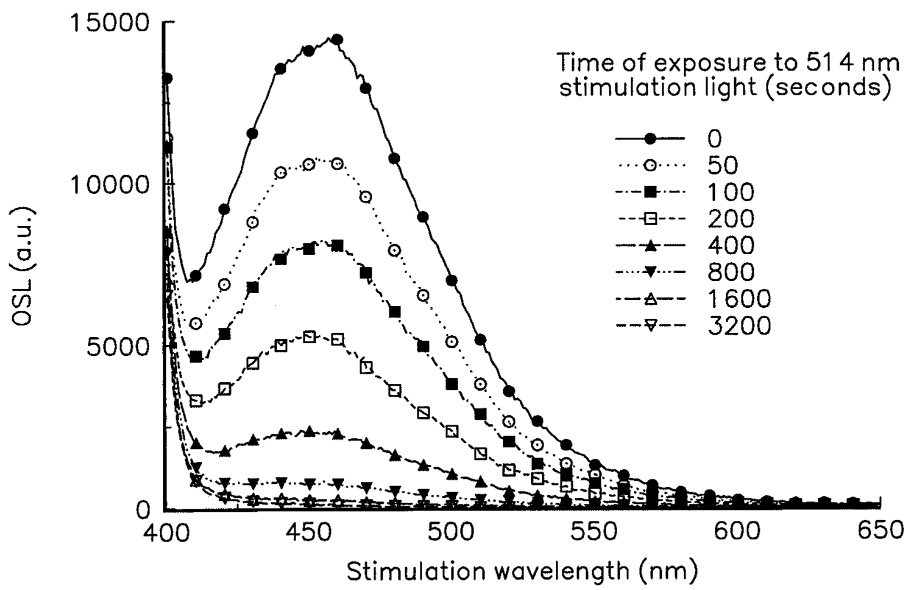

Figure 2.14. Stimulation spectra of a quartz sample, which has been heated to 450 deg.C, irradiated to a beta dose of 10.8 Gy and preheated at 220 deg.C for 300 seconds. The sample was exposed to 514 nm stimulation light for various periods of time, after which the stimulation spectra were measured.

Retrospective dosimetry

The general development of methods to retrospectively determine accident doses in populated areas has continued. As part of an EU project dosimetric assessment using new techniques based on OSL on porcelain and bricks were developed and tested at Risø. The work has been concentrated on the optimisation of the OSL response of brick and porcelain materials to low doses as part of an intercomparison project.

Experiments were carried out to investigate the possibility of enhancing the OSL sensitivity of porcelain. Thermal activation procedures showed that the OSL signal in many cases followed the same sensitisation pattern as the 110 deg.C TL peak in porcelain which leads to the use of OSL in the so called predose techniques.

A low-dose intercomparison based on measuring luminescence signals from brick and porcelain samples collected in Chernobyl was carried out within the EU luminescence group. In order to accurately determine the acquired doses a beta source of 5 mCi was attached to the automated Risø TL/OSL reader and calibrated. The aim of the intercomparison was to compare measurements using different luminescence methods on ceramic samples that was known to have received doses in the range of 100 mGy. Risø entirely used OSL with green light stimulation (420 - 550 nm) for both porcelain and quartz samples. The OSL results obtained at Risø were in very good agreement with known values and the mean values obtained by the other participants of the intercomparison.

Figure 2.16 shows a typical OSL growth response curve for a toilet top tank sample collected in Chernobyl and figure 2.17 shows the depth dose profiles (dose versus depth) for three brick samples also collected in Chernobyl.

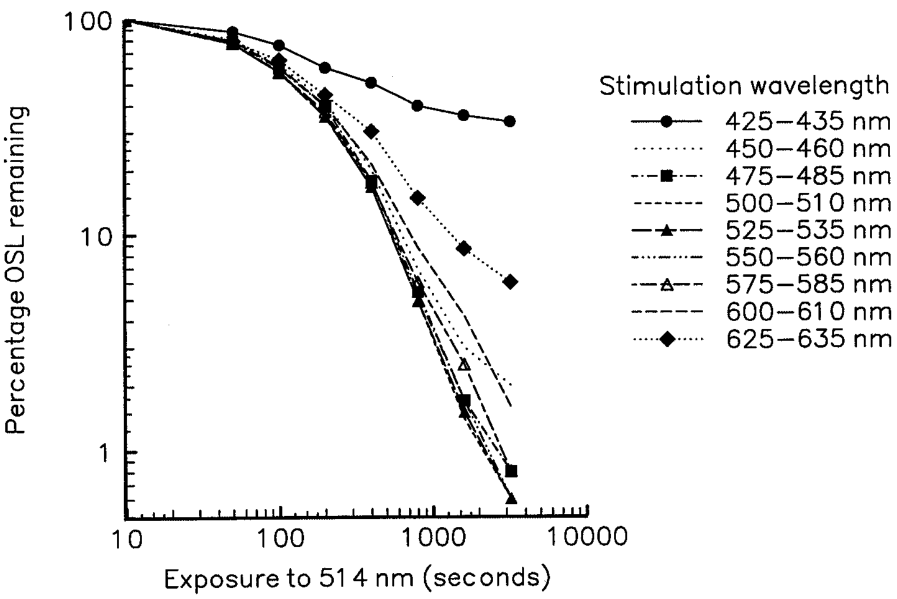

Figure 2.15. Data from Figure 2.14 plotted as bleaching curves at wavelength intervals of 25 nm.

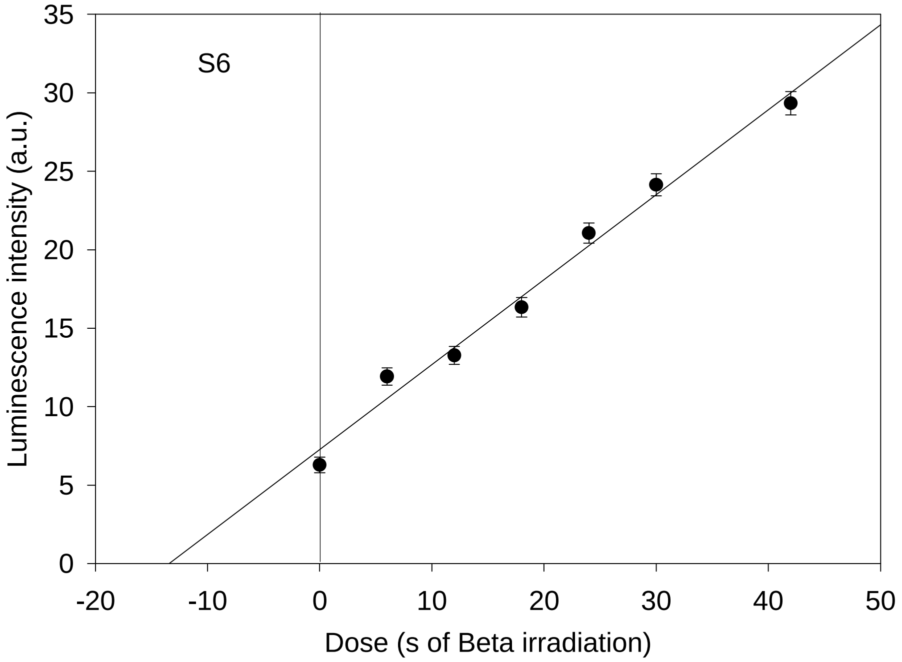

Figure 2.16. OSL growth response curve for a porcelain sample from a toilet top tank collected in Chernobyl.

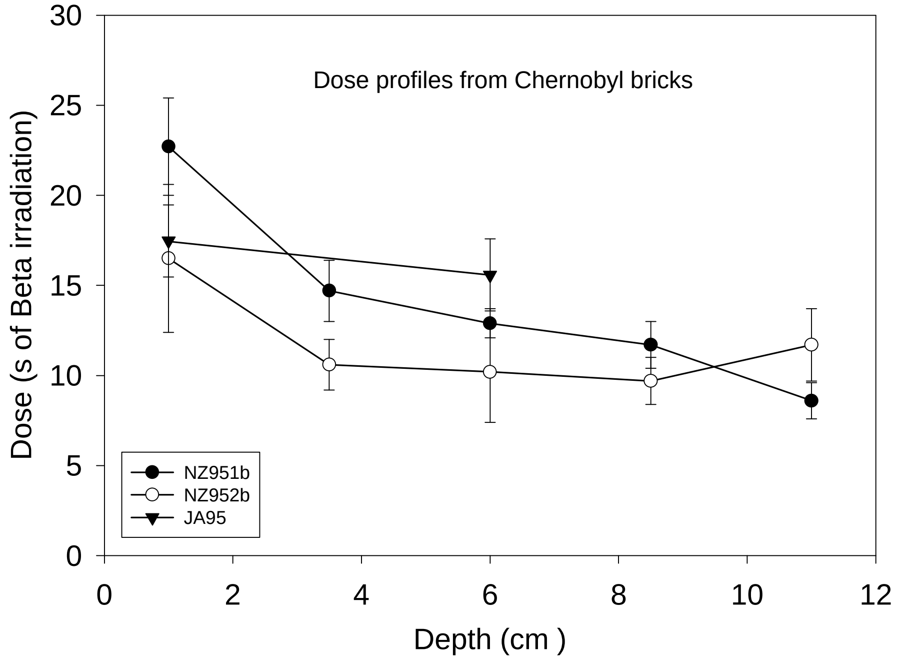

Figure 2.17 Depth dose profiles for three brick samples collected in Chernobyl.

2.3 Radon

Reduction of radon in Danish houses

In 1996, the first major Danish research project on radon-reduction methods in existing houses was completed. The project was initiated in 1994 as a collaboration between Risø and four other Danish institutions: the Danish Building Research Institute, the National Institute of Radiation Hygiene, the Geological Survey of Denmark and Greenland, and COWI Consulting Engineers and Planners AS. The project was financed by The National Building and Housing Agency and the National Institute of Radiation Hygiene.

Since radon-reduction methods for existing houses are relevant mainly for houses with high radon levels, it was necessary first to identify such houses for evaluation of the methods. Based on previous findings, it was known that geology is the prime factor controlling indoor radon, and that granite, clayey till, and limestone are geologies with a high indoor-radon potential. Three regions representing these types of geologies were therefore identified, and a total of 270 houses were surveyed for indoor radon in the winter 1994-95. From these results, 24 houses with annual radon levels estimated to be above 200 Bq/m^3 were invited to participate in the testing of the radon-reduction methods. 21 house owners accepted the offer. Furthermore, 42 houses with estimated annual radon levels in the range 100 to 200 Bq/m^3 were selected as controls. In the fall of 1995, radon-reduction methods were then installed in the reduction houses, while nothing was done in the control houses. In the winter of 1995-96, a final set of radon measurements were carried out. The reduction efficiencies of the

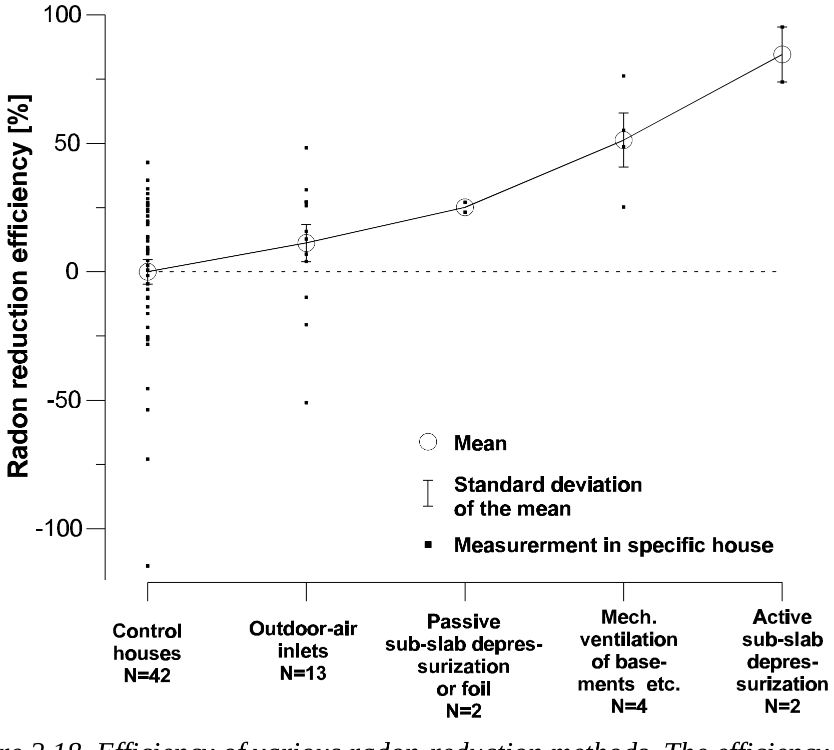

Figure 2.18. Efficiency of various radon-reduction methods. The efficiency gives the reduction in radon level relative to the pre-mitigation level. For comparison, the graph also shows the results for the control houses. The scatter of points reflects the natural variation of indoor-radon levels from one winter to another.

methods were assessed based on the winter measurements conducted in the reduction houses before and after remediation. Furthermore, observations in the 42 control houses were used to correct for natural variation of the winter concentrations.

The following reduction methods were tested: active sub-slab depressurization (2 houses), passive sub-slab depressurization (1 house), mechanical ventilation of basements (4 houses), floor sealing by use of polyethylene foil above the concrete slab (1 house), and installation of air vents (13 houses).

The main findings of the study are shown in Figure 2.18. In agreement with studies in other countries, the active sub-slab depressurization technique was found to be highly efficient. This type of system works by reversing the pressure difference across the concrete slab of the house such that air flows from the house towards the soil. The reduction efficiency estimated from the two winter measurements were confirmed by short-term measurements based on continuous monitoring with ion chambers (see Figure 2.19). Before the remediation, the house shown in Figure 2.19 had an annual indoor-radon level of 1000 Bq/m^3. After the remediation, the annual level was estimated to be just 50 Bq/m^3.

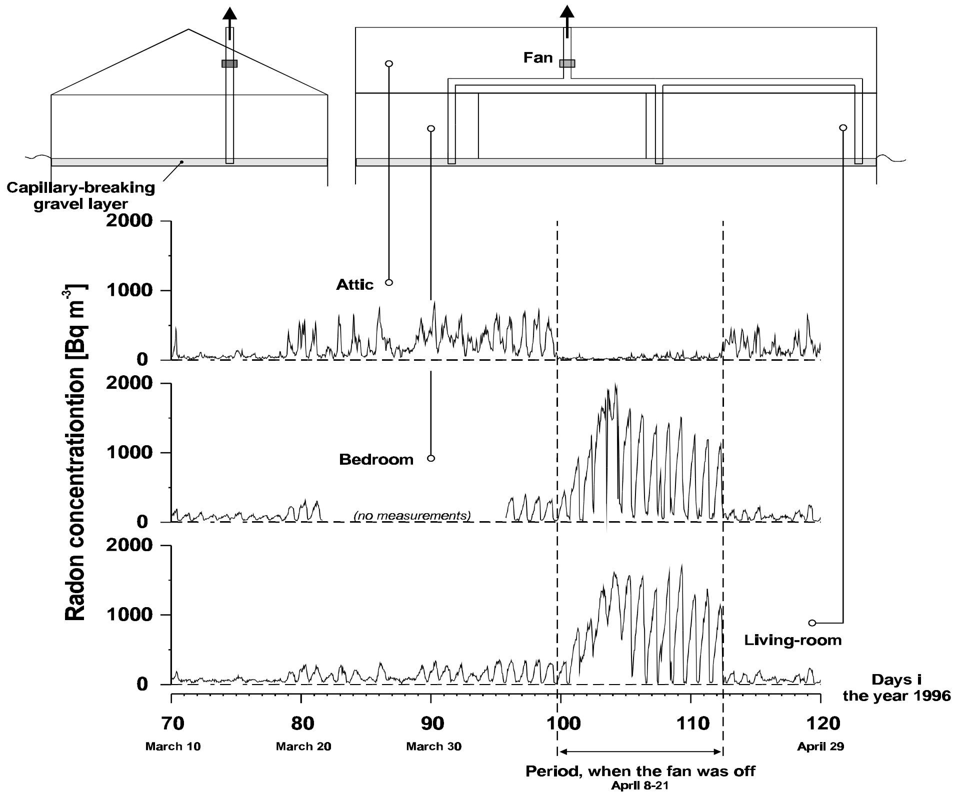

Figure 2.19. Active sub-slab depressurization is a highly efficient radon-reduction method as demonstrated by the continuous measurements shown in this figure. The sub-slab suction was on for the entire period shown in the figure except for the part indicated by the dashed lines. It is seen that the radon concentration in the house increased markedly when the system was switched off, and that it decreased again when the fan was turned back on. The radon level in the attic followed a reversed pattern probably because of leakage from the ventilation channels.

Mechanical ventilation of basements was also found to be a very efficient reduction method. In comparison, the passive methods (air vents, cover-up of the floor with foil etc.) were found to be less effective than the active systems mentioned above. Because of the (natural) winter-to-winter variability of indoor-radon levels these results were not found to be significantly different from the measurements in the control houses.

The study demonstrates, that efficient radon-reduction methods are available for Danish houses. However, more work is needed to obtain reasonable reduction efficiencies with passive systems such as improved natural ventilation.

Sampling of soil-gas radon

In 1995, Risø participated in an international workshop on state-of-the art of soil-gas measurements methologies held at the Environmental Measurements Laboratory in New York. The workshop demonstrated, that the main source of uncertainty in soil-gas radon determinations is the soil-gas sampling procedure. Although the method applied by Risø gave good results at the workshop, it was found desirable to study the method in further detail. In 1996, such a study was undertaken at Risø in collaboration with a visiting Ph.D. student from the Kernfysisch Versneller Instituut in the Netherlands. The study comprised both experimental investigations at a clayey-till field site and numerical modelling.

The specific purpose was to determine the optimum volume of soil gas that should be extracted from the soil to obtain an accurate determination of the radon concentration at 1 m depth, and to assess the sensitivity of the sampling technique to air leakage along the probe.

The probe is sketched in Figure 2.20 (a): A 12 mm hollow PVC pipe is hammered 90 cm into the ground and a 20 cm cavity is drilled by hand. Air is then sampled from approximately 1 m of depth. The radon concentration of the sampled air is determined with scintillation cells. During normal measurements only few litres of air are drawn from the probe. The study confirmed that this is a good measurement protocol and that the sampled air in this case indeed comes from the soil centred at 1 meter of depth.

To investigate the sensitivity of the method to air leakage along the probe, experiments were carried out where a radon concentration much higher than that of the soil was established around the probe at the soil surface. This was accomplished with a small chamber and an external source of radon. In other words, air with a high radon concentration was used to trace fast-flow paths from the surface to the sampling point in the soil. Air was then withdrawn continuously from the probe at a rate of 1 litre per minute. Figure 2.20 (b) shows the results of one set of measurements and numerical-simulation results conducted with a time-dependent radon-transport model. The model assumes that a crack exists along the probe, and that this crack extends from the soil surface and 40 cm downwards. It is seen from the figure, that the break-through of the high-radon pulse reaches the sampling cavity after approximately 30 litres of sampling. The good agreement between the measurements and the modelling results (and the visual inspection of the probe) suggests that such a 40 cm crack probably exists along the probe, but that such a crack will not significantly influence the measurements in so far as the standard sampling procedure is followed.

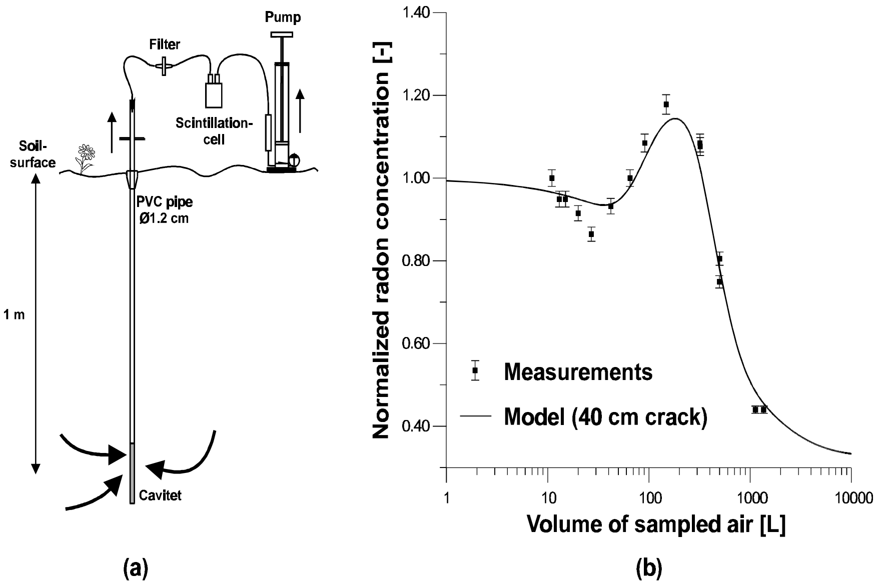

Figure 2.20 (a) Sketch of sampling equipment used for soil-gas radon measurements. (b) Field test of the sensitivity of the sampling technique to leakage along the probe based on the use of high-radon levels as a tracer (see text). The points are experimental results and the solid line is a numerical model simulation based on the assumption that a 40 cm crack exists along the probe.

Radon transport in fractured media.

Clearly, macropores or fractures in soils such as clayey tills can enhance the ability of soil to conduct flows of gasses, but little is known regarding the influence of macropores on the availability of radon for advective transport. The problem is that radon is generated mainly in the matrix of the soil. Hence, if the gas flow is confined to macropores there may be less interaction between the source of radon and the gas flow in comparison to the situation where the flow is distributed more uniformly over the soil. An improved understanding of these issues may have implications for the development of techniques to assess entry of radon or other soil gasses to houses.

In 1996, the experimental part of the investigations was focused on transport through an intact sample of clayey till positioned in an arrangement of the type shown in Figure 2.21. The experiments took place at the Geotechnical Institute, that has developed the techniques needed for the handling of such large intact soil samples. The main set of experiments concerned the mapping of breakthrough curves of radon (radon-222) and thoron (radon-220).

The experimental data are currently being analysed, and different types of transport models are used to interpret the results. The main model has been developed at Risø. It assumes that the soil consists of a matrix of soil transversed by small pinholes. These pinholes simulate the macropores observed visually in the sample when the sample was dismantled after the experiments. The study will be concluded in 1997.

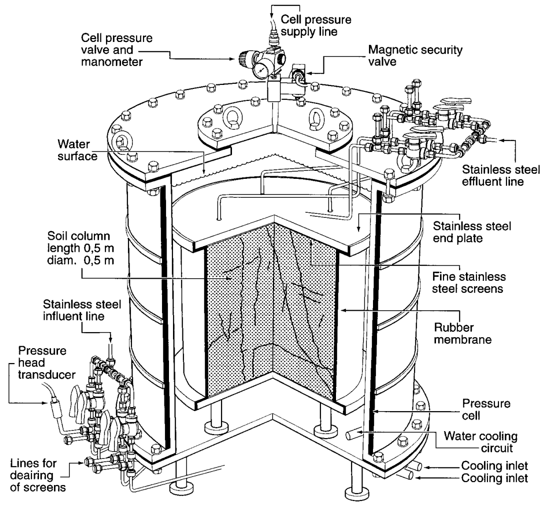

Figure 2.21.The Geotechnical Institute has designed a technique for handling of intact soil samples of 50 cm in diameter. The soil sample is located in a water-filled pressure cell which subjects the samples to mechanical stress as in the natural environment. In 1996, Risø studied soil-gas and radon transport through one sample. [By courtesy of the Geotechnical Institute, Lyngby, Denmark].

2.4 Emergency Preparedness

Action Levels in the Event of a Nuclear Accident

Following a nuclear accident with a release of radioactivity to the atmosphere, measurements carried out as part of a nuclear emergency preparedness programme may provide early indications of the severity and scale of the accident, thereby contributing to an assessment of the off site radiological consequences of the accident.

The measurements may also facilitate a rapid emergency response by invoking pre-determined action levels as site- and accident-specific observable thresholds for the implementation of dose reducing protective actions.

In a study carried out as a part of the Nordic Nuclear Safety Research (NKS) programme EKO-3.3, dose rate measurements are analysed as being early indicators of the forecasted radiation doses to the public in the event of a severe accident at a nuclear reactor. The accident is assumed to include core-melt, loss of coolant and a breach of containment, followed by a release of activity to the atmosphere. The extent, to which dose rates and doses averted by means of protection are correlated, is evaluated in an Monte Carlo calculation. The dose rates and the avertable doses are obtained with the help of a simplified atmospheric dispersion model using two years of meteorological data collected at Sturup airport near the Barsebäck nuclear power plant. The study shows that dose rate measurements carried out in the early phases after an accident may provide a valuable tool for decision support, and that action levels of dose rates may be defined for the purpose of assisting a rapid emergency response.

A CsI(Tl) Based Spectrometer as a Field Instrument for Contamination Analysis

A portable gamma ray spectrometer/dose rate detector based on a CsI(Tl) scintillator and combined with a GPS position device has been assembled. The aim is to test the latest available technology for field use by nuclear emergency measurement teams.

The field instrument has the following main components:

- CsI(Tl)-scintillation unit 40x40 mm (A standard 3"x3" Na(Tl) unit is optional).
- GPS position device (12 channel parallel detector unit).
- Computer - single board processor with LCD screen and solid state disk.
- Cellular phone or radio modem link to a central receiver.
- Power supply from 12 V battery

The instrument is able to display the actual position on maps, and to transfer data to a central receiver.

Exercise

On November 11, 1996, Risø together with the Emergency Management Agency (EMA) took part in exercise INEX-2-CH. The exercise dealt with an accident at the Swiss Nuclear Power Plant Leibstadt. 29 countries in Europe, America and Asia participated in the exercise, the main purpose of which was to test the exchange of information.

International exchange of data from early warning stations

Data from Denmark's 11 early warning stations are once a week sent to the EU Joint Research Centre at Ispra, Italy. Similarly, 13 other European countries are sending data to Ispra. The 14 participating countries all use different data formats. However, data from the 533 early warning stations in the 14 countries are transformed to a common format at Ispra and returned to all participants. In case of an nuclear accident the data exchange rate could be increased to several times a day.

2.5 Publications

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Unpublished. Abstract available

3 Reactor Safety

The work of the Reactor Safety Programme was in 1996 concentrated on reactor physics and severe accident investigations as well as maintaining the necessary knowledge for nuclear preparedness activities.

3.1 Reactor Physics

3.1.1 Calculations of Neutron Doses to Reactor Components

Calculations have been performed of the neutron fluxes in power reactor components outside the reactor core under contract with two Swedish nuclear power plants. For the Ringhals 1 reactor (a boiling water reactor of Swedish design) the neutron doses received by the stainless steel moderator tank were calculated, and for the Forsmark 1 reactor (also a Swedish BWR) both neutron doses and neutron induced activations of the moderator tank, the fuel support plate, the fuel top guide, the steam separator, and the pressure tank were calculated.

To verify these calculations, which were performed by use of standard diffusion theory codes, it is planned to perform Monte Carlo calculations as well.

3.1.2 Pu-Benchmark Calculations

A NEA benchmark problem of multiple Pu-recycling in PWRs has been calculated with the newly developed version of the neutronic code CCCMO_N. This code is able to perform burn-up calculations on 32 heavy nuclides (from ^{232}Th to ^{252}Cf) as compared to the 12 nuclides in the old version. The outcome of the exercise demonstrated the need for further improvements in the CCCMO_N code. The most important shortcoming of the code was the resonance treatment: A much more sophisticated resonance treatment has to be introduced in order to cope with the very high Pu-concentrations which are used in Pu-recycling fuel (some 10% Pu contents in depleted U).

3.1.3 Development of an Improved Thermo-Hydraulic Model for Natural Circulation BWRs

A fluid dynamics code DYNOS based on a mechanistic 4-equation drift-flux model for two-phase flow has been developed. The code includes a multiplum of correlations available for the user, and it may work with both ordinary water and refrigerant 12 as a fluid. While the code is especially suited for the simulation of the fluid dynamics of natural circulation, boiling water reactors, it is capable of treating a wide range of general fluid dynamic problems. It will be combined with neutronic calculations.

Validation of the code has been performed in cooperation with the Interfaculty Reactor Institute at Delft University of Technology, the Netherlands. Here an experimental facility, DESIRE, especially designed for the study of gravity driven two-phase flow is available. Most data used in the validation originates from test runs on this facility. A simplified diagram of the DESIRE facility is shown in Figure 3.1. The static part of the code was validated against void fraction, pressure drop and natural circulation experimental data with excellent results (Figure 3.2). A comparison with results obtained with the Norwegian MONA code was also performed.

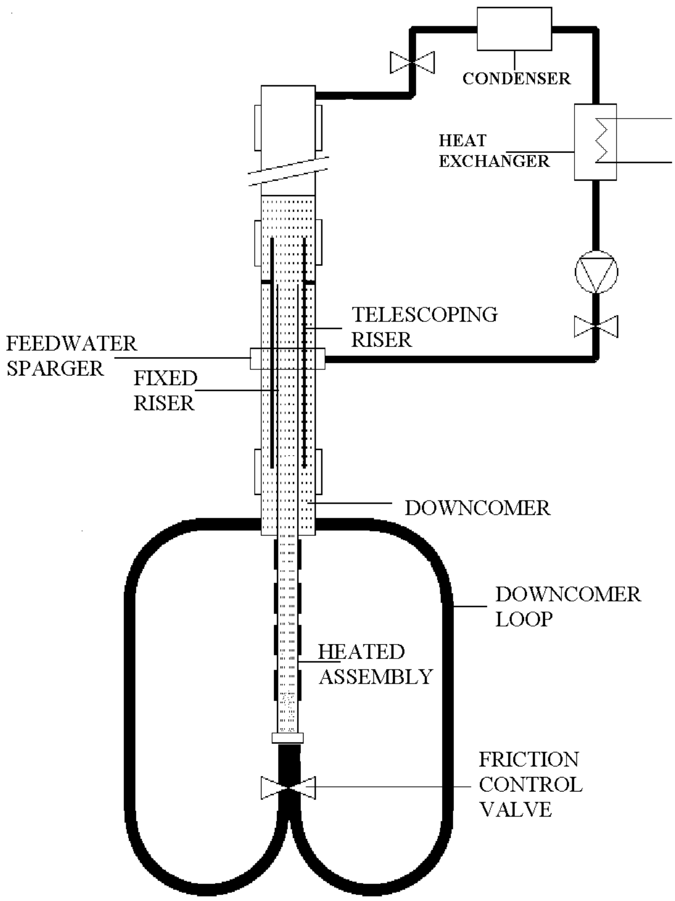

Figure 3.1. The DESIRE facility at the Interfaculty Reactor Institute at Delft University of Technology.

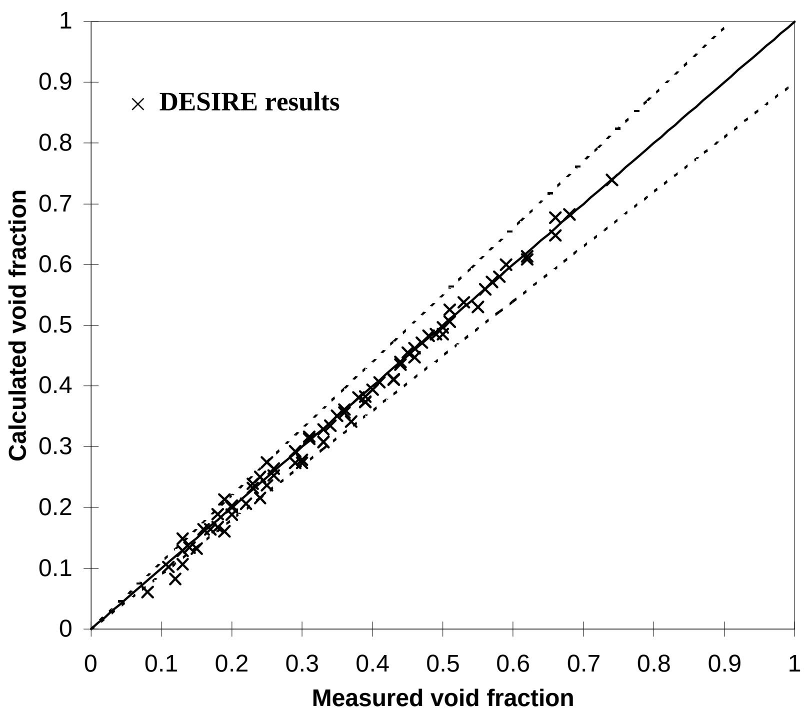

Figure 3.2. Void fraction results.

The validation of the dynamic part consists of step responses in power, pump trips and stability limits under natural circulation. Although not yet finished the results obtained so far have been very promising.

3.2 Severe Accidents

Severe accidents, i.e. accidents which go beyond the design basis accidents, have been an important part of the reactor safety research, internationally as well as nationally in the later years. The 1996-activities in this field of the Reactor Safety Programme have all been part of larger international projects.

In 1996 work was performed on the following projects:

- Recriticality studies (3.2.1)
- Core meltdown and coolability (3.2.2)
- Risks connected to the decommissioning of old nuclear submarines (3.2.3)
- Inventory of radioactive materials in the Ignalina nuclear power plant (3.2.4)
- Aerosol resuspension (3.2.5)

3.2.1 Recriticality Studies

In special hypothetical reactor accidents a situation may occur where the reactor core is left uncooled for a time long enough for the control rods to melt and disappear from the core, but not long enough for the fuel to be damaged. Upon reflooding, uncontrolled criticality will occur, possibly with detrimental consequences for the reactor fuel.

A code RECRIT was written which follows such an accident by use of very simplified models. However, after criticality has been reached, the thermo-hydraulics of the system becomes very complicated and could not be simulated by the code. In collaboration with VTT of Finland, the code has been provided with much more refined thermo-hydraulic models, which should simulate much better the progression of the accident after the criticality. However, the code is not quite operable as yet. This work has been performed as part of a RAK-2.1 project under the Nordic Nuclear Research Programme (NKS). As a result of this work a proposal for a research project was submitted to the EU. The proposal has been accepted and the project will run for the next two years.

3.2.2 Core Meltdown and Coolability

A broad literature study on core meltdown and in-vessel coolability for light water reactors has been performed within the current research program. In addition, Risø participates with a literature study in the Nordic Nuclear Research Programme (NKS) sponsored RAK-2.1 project, in which hypothetical severe accidents are analysed by computer simulations.

The understanding of severe accident phenomena has progressed substantially since the Three Mile Island (TMI) accident in 1979. Detailed information on the final state of the damaged reactor and analyses of the accident progression are found in reports from the TMI-2 Accident Evaluation Program and the subsequent Vessel Investigation Project performed under the auspices of the OECD. International cooperation and research programs have made an important contribution to the understanding of meltdown phenomena.

While the processes in the early stages of core heatup and meltdown are reasonably well understood, the behaviour of debris beds and molten pools as well as the phenomena of melt relocation, blockages, and molten fuel/coolant interactions are much less well understood. These late-phase processes are given special attention in the literature study under the RAK-2.1 project.

3.2.3 Risks Connected to the Decommissioning of old Nuclear Submarines

An international study on "Cross-Border Environmental Problems Emanating from Defence-Related Installations and Activities" is performed under the NATO/NACC Committee of the Challenges of Modern Society. Risø has participated in investigations of the risk involved in the decommissioning of old nuclear submarines. An analysis has been made of the potential sources of cross-border radioactive pollution, such as reactivity accidents, loca's, loss-of-shielding accidents and leaks from radioactive waste storage facilities. Furthermore, an order-of-magnitude approach has been developed for the calculation of the energy developed and the radioactivity produced during a reactivity accident in a submarine reactor during defueling. The probability per defueling of such an accident has been estimated.

3.2.4 Inventory of Radioactive Materials in the Ignalina Nuclear Power Plant

As part of a study of the consequences of a hypothetical accident at the Ignalina nuclear power plant in Lithuania the inventory of radioactive materials in one of the Ignalina reactors was calculated by use of the CCCMO code.

3.2.5 Aerosol Resuspension

In the case of a severe accident in a light water reactor, fission products and structural materials will be released from the damaged reactor core. A significant part of the material will be deposited as particles in the pipes of the reactor coolant system. In a later phase of the accident the particles may be torn loose and become resuspended in the containment atmosphere if the velocity of the steam-gas mixture in the pipes increases. This resuspension can be important for the magnitude of a release of radioactivity to the environment.

A series of experiments on resuspension is being performed at the STORM facility at the European Union's Joint Research Centre at Ispra, Italy. A member of the Reactor Safety group takes part in the experiments and in the analysis.

Until now, the experiments have concentrated on deposition and it is found that deposition by turbulent motion of the gas stream is enhanced more than expected if the pipe wall is significantly colder than the gas. Preliminary results for resuspension suggest that the particles are torn loose as aggregates rather than as single particles.

3.3 Nuclear Knowledge Preparedness

The programme participates in the Danish Nuclear Preparedness Group, the aim of which is to ensure that the necessary knowledge in the field of nuclear safety and technology exists in Denmark so that information in this field, needed by Danish authorities and media is readily available. The members of the group come from Risø National Laboratory, the Technical University of Denmark and the Danish Emergency Management Agency.

3.3.1 Annual Report on the International Status of Nuclear Power

In 1996 the second, annual report on the international status of nuclear power was published in Danish. The report has been distributed to politicians, civil servants and other interested persons as well as to the Danish media.

3.3.2 Reactors in Nordic Surroundings

As part of the Nordic Nuclear Research programme (NKS) the Reactor Safety Programme has been responsible for compiling information on reactors in Nordic surroundings under the RAK-2.3 project. In 1996 the following reports were prepared:

- Description of the Prototype Fast Reactor at Dounreay
- Description of the Advanced Gas Cooled Type of Reactor (Figure 3.3 and Table 3.1)
- Accidents in Nuclear Ships

Figure 3.3. AGR stations in Great Britain.

Table 3.1. AGR stations in operation.

Station unit	MWe	Generation of AGR	Sister plant	Start of operation
Dungeness B	2 x 660	first	a	1983-85
Hinkley Point B	2 x 660	first	a	1976
Hunterston B	2 x 660	first	a	1976-77
Hartlepool	2 x 660	first	b	1983-84
Heysham I	2 x 660	first	b	1983-84
Heysham II	2 x 660	second	c	1988
Torness	2 x 660	second	c	1988
Total number of AGR units : 14 at 6 sites and 7 stations				

Furthermore, also as part of the RAK-2.3 project, a data base containing data of nuclear power plants, situated i countries close to the Nordic region, has been produced.

3.4 Activities at the DR 1 Reactor

Activities at the DR 1 reactor have concentrated on courses on experimental reactor physics for university students and high-school classes.

3.4.1 Education

The reactor has been used almost exclusively for educational purposes. About 40 high school classes have carried out one-day or half-a-day experiments at the reactor. The total number of high school pupils visiting DR 1 in 1996 was about 700.

A number of students from the Technical University of Denmark have carried out experiments at the reactor over a period of three weeks. Some of the experiments are:

- Determination of the reactor's temperature-, power, and bubble-coefficients
- Neutron activation analysis
- Measurements of neutron cross sections
- Neutron radiography
- Health physics experiments
- Core flux distribution measurements.

3.5 Publications

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4 Research Reactor DR 3

DR3 is a heavy-water moderated and cooled nuclear research reactor which has been in operation since 1960. It was originally built as a Materials Testing Reactor, but today it is used as a Multipurpose Research Reactor. The operation cycle is 4 weeks, of which 23½ days is continuous operation and 4½ days is shut down The vertical experimental facilities comprise 13 tubes in the core, 50 mm diameter, and 14 tubes in the D_2O and graphite reflectors with sizes ranging from 10 to 18 cm in diameter. Four tubes, 18 cm diameter. pass horizontally and tangentially to the core. These facilities were intended for loop experiments, but turned out to be excellent beam ports. Two beam ports are supplied with thermal neutrons from a water scatterer, two others are supplied with cold neutrons from a 38 K cold hydrogen neutron source.

Six three-axis spectrometers and a Small Angle Neutron Scatterer (SANS) instrument are supplied by the neutron beams from the four beam ports belonging to the tangential 18 cm diameter tubes. One of the cold source beam ports is connected to a building outside the reactor hall, by means of a neutron guide tube. The monochromatic cold neutron flux at the sample position is: $7 \cdot 10^6$n $cm^{-2}s^{-1}$(E_n=5 meV). Two of the three-axis spectrometers are multi-purpose instruments with a high degree of flexibility, facilitated by five different detector arms which can be attached or detached in less than one hour.

Neutron scattering is an important technique for the study of large molecules such as polymers and biological molecules and for the study of superconductivity and magnetism in high-temperature superconductors. These are examples of recent applications of the beam facilities of DR3. DR3 is appointed as a Large European Beam Facility and the neutron beam instruments are intensively utilized by researchers from Risø and from other EU-countries.

The neutrons from DR3 are also used for activation analysis, isotope production and transmutation doping of silicon. These activities are described in section 5.

In 1996 a major inspection of the reactor installation was carried out during an extended shut down in September-October. The reactor tank was emptied and inspected by periscope and found to be in good condition. The graphite reflector was inspected and found to be intact and undamaged. Maintenance of the cooling circuits was carried out during the period where no fuel was in the reactor.

In 1996 DR3 was kept in operation at 10 MW for 6644 hours, corresponding to 76% of the year.

5 Isotope Laboratory

5.1 Radioanalytical Chemistry

The Isotope Laboratory participated in an intercomparison arranged by the National Institute for Occupational Health prior to participation in an EU project concerning the preparation and certification of human serum and urine. As, Se and Zn were determined by neutron activation analysis.

Co-operation with the John F. Kennedy Institute has continued with the diagnosis and verification of Menkes disease by the determination of Cu in chorionic villi and placenta samples by radiochemical neutron activation analysis. In co-operation also with the University of Helsinki, As, Cu, Mn and Se were determined by radiochemical neutron activation analyses in samples from patients with occipital horn syndrome, which is related to Menkes disease.

A project for determining Pt in various organs of cancer patients was completed. Results were reported at the 2nd International Symposium "Metal Elements in Environment, Biology and Medicine", held in Timisoara, Rumania, October 27-29, 1996. A more detailed publication is under preparation in co-operation with Copenhagen University Hospital.

An improved method was developed for the determination of trace amounts of Pt in airborne dust and soil from Copenhagen. The method includes the digestion of air filter samples and preconcentration of Pt by electrolysis. In co-operation with Ben Gurion University, Beer Sheva in Israel, we attempted to determine Pt in highly polluted air filter samples by instrumental neutron activation analysis.

5.2 Irradiation services

The Basic Irradiation Service section continues to fulfil our commitment to cover all needs for neutron-irradiated materials for technical and scientific purposes in Denmark. It is responsible for the production of radioactive isotopes and other radioactive materials for industry, hospitals and research institutions. An increasing part of the deliveries to domestic as well as foreign customers are unprocessed irradiations from dedicated reactor irradiation facilities. All radioactive materials needed for Risø's own research are made as ready-to-use preparations.

A total of 1096 irradiations were carried out for use by Risø and 30 different external customers in Denmark, Sweden, Germany, Switzerland and the U.K. The number of dispatched batches of NH ^{82}Br is only slightly smaller than the year before in spite of the 6 week shut down period of the DR3 reactor in 1996, and there are an increasing number of scientific customers from abroad using irradiations in the thermal neutron facility for fission track, and argon-argon dating of rocks. Altogether 140 shipments of other radioactive products were made to a variety of institutes, industry and hospitals; approximately 28 % were radiopharmaceuticals.

Contracts with the Danish Isotope Pharmacy and the Department of Environmental Science and Technology regarding control analysis of radiopharmaceuticals were signed in October. In November a quality manual was implemented for the production of radiopharmaceuticals. At the end of the year the production of radiopharmaceuticals was stopped after more than 35 years of continuous production and in the last 18 years with a production licence from the Danish National Board of Health. The demand for our special radiopharmaceuticals had decreased so the sale had dropped to an unprofitable level.

For research applications at Risø 48 irradiations were performed and 120 deliveries of radioisotopes in specially prepared forms were made. For educational purposes 444 solid radioactive sources were supplied to the Nordic countries.

5.3 NTD-Silicon

Neutron transmutation doping of silicon takes place in 7 facilities in the DR 3 reactor. Three facilities are placed in the heavy water in vertical positions and one facility is placed in a horizontal position in the heavy water.

A new horizontal 5" facility has been assembled on a test site outside the reactor and is ready for test according to schedule. As planned it will be installed in DR 3 in September 1997.

Due to the level of documentation of the quality management system the Danish Standards Association has recommended a decrease of the number of annual audits from two to one.

The laboratory has now signed agreements with three internal suppliers.

Chemical doped silicon has been a great competitor to NTD-silicon, and the market for NTD-silicon has dropped world-wide. Risø has still a fair market-share, and we are trying to increase our part.

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5.4.1 Publications in International Journals, Proceedings and Reports

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6 Waste Management

6.1 Practical Waste Management

The Waste Management Plant at Risø is a nuclear facility for the collection, processing and storage of Danish radioactive waste. Chemically toxic waste, ordinary waste water purification and laundry facilities are also taken care of by the staff of the Waste Management Plant.

6.1.1 Radioactive waste

By agreement with the Danish Institute of Radiation Hygiene radioactive wastes from all Danish users of radioisotopes must be treated and stored at the Waste Management Plant at Risø, if it contains activity above specified limits.

The treatment methods for radioactive waste are unchanged from previous years:

A balling press is used for compaction of low-level solid waste. In 1996 5.5 t out of total of 9.2 t solid low-level waste was coming from outside, primarily from hospitals, industry and various laboratories. The waste from outside contained about 1290 GBq radioisotopes with half-live longer than 1 year, but most of this was in form of a few large closed sources.

Through more than 30 years low-level radioactive waste water from the facilities at Risø has been purified by distillation. The release with the distilled water and from various other minor sources to the recipient, Roskilde Fiord, was ~100 MBq of mixed β-emitters. This is 1.0 % of the permitted releases. In addition 3680 GBq of tritiated water were released to the fiord and about 8 GBq ^{14}C-carbon dioxide to the atmosphere.

The evaporator concentrate is bituminized using a method developed at Risø 25 years ago. The purification of the 1770 m^3 waste water gave rise to about 2 t bituminized material containing about 2.4GBq β-activity. As noticed last year the termination of the hot cell work at Risø has reduced the content of long-lived radioisotopes in the bituminized product.

The ordinary radioactive waste treatment procedures resulted in a total of 80 drums containing bituminized evaporator concentrate or compacted low-level solid waste.

During 1996 the last waste units from the old storage area "Betonrørslageret" were transferred to the storage building for low-level waste near the Waste Management Plant. The operation comprised a total of 3289 old 210 drums containing evaporator concentrates, compacted solid waste or contaminated soil collected between the start of the research centre and up to 1989. Many of the older waste units were in unsatisfactory condition and 38% had to be overpacked using an outer 280 L drum. The 3-year operation gave a collective total dose of 70 mSv to the involved personnel, only half of the initial estimate.

The photo Figure 6.1 shows some old waste drums on the way to renovation and storage in the new facility. External radiation from the units were in most cases very low so that they could be handled without special precautions.

At the end of 1996 the new storage building contained 4341 waste units. The drums in addition to the above mentioned older units are new ones produced since 1989 or drums filled with secondary waste produced in connection with the emptying of the old storage facility.

External γ-scanning for ^{137}Cs and ^{60}Co were made on all drums which might contain γ-emitters. A description of the measurement principles are available, see Section 6.2.1.

Although all waste units have been removed from "Betonrørs-lageret" some slightly contaminated concrete constructions remain at the site. Permit for demolition of these concrete parts has been obtained from the authorities. The operation is planned to take place in 1997, leading to eventual complete declassification of the site. As typical for all declassification the central problem is documentation of sufficiently low levels of activity. A large effort in measurement and reporting is needed. Some preliminary studies was carried out in preparation for extension of the facility for storage of waste which is α-contaminated or with high external radiation (Centralvejslageret). Permits from the authorities will be applied for in 1997.

Figure 6.1. Transport of old waste units. The nearest is a badly corroded single layer drum, but in most cases the drums are double: a 100 L drum with the waste material inside a 210 L drum and with the annulus cast with cement mortar.

The storage facilities at Risø are only for temporary use and eventually the radioactive waste must be disposed of permanently.It is expected that this will take place in connection with the future decommissioning of the nuclear facilities at Risø.

6.1.2 Inactive systems

Decontamination of protective clothing, ordinary laundry facilities, collection of chemically toxic waste, and purification of ordinary sewage water from the Risø area are also taken care of at The Waste Management Plant.

More stringent environmental requirements to the releases of organic materials and especially nitrogen components to the recipient, Roskilde Fiord has made a reconstruction of the biological waste water treatment plant necessary. Construction of the new plant was initiated in 1995 and completed in the summer 1996. The plant operates according to the active sludge principle and is quite conventional. It appears to function satisfactorily, but some minor break downs of mechanical equipment combined with cool winter weather has so far prevented smooth operation of the aerobic/anaerobic nitrogen purification. Figure 6.2 gives, however, an indication of the improvement in the removal of organic materials from the waste water.

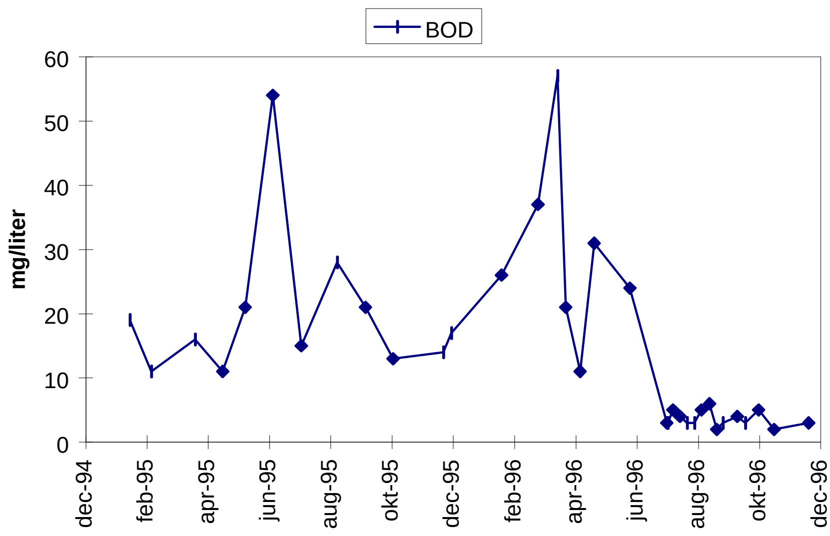

Figure 6.2. Concentration of BOD (organic material expressed as biological oxygen demand) in effluent from the biological purification plant at Risø. The new permissible concentration is 15 mg/L.

Semi-dry sewage sludge from the old waste water purification system has been in temporary storage for about 5-8 years. It is somewhat contaminated with heavy metals including uranium. The sludge was transferred to the controlled tip for inactive waste at Risø after permission from the environmental protection authorities was obtained early in 1996.

6.2 Waste Management Research and Development

Knowledge about isotope contents and general properties of radioactive waste units is needed for safety assessments of long-time storage or disposal of radioactive waste. Interaction of waste units with the surroundings under various conditions is also important.

Although actual disposal of Danish radioactive waste is not expected for a considerable period, it is considered important to maintain some research efforts and participation in projects related to safety assessment of disposal of radioactive waste.

Formally the research projects are part of the Radiation Protection programme, see Chapter 2, but because some of the studies are closely related to practical waste management they are reported here in continuation of the description of the treatment and storage systems.

Staff from the Waste Management Plant is contributing to international research and development within the following areas:

6.2.1 Measurement of isotope inventories etc. in Risø waste

The system used at Risø for γ-scanning of waste drums has been reported. The system uses an iodide crystal as detector with distance and in some cases perforated lead screens to regulate the size of the signal. Each drum is measured from four sides.

Due to the low resolution of the iodide crystal it is in practice only possible to measure contents of ^{137}Cs and ^{60}Co. Interference from short-lived radioisotopes may occur for new drums. Use and calibration of the system is described in detail with special emphasis on uncertainties. The later is mostly dependent on the unknown distribution of γ-emitters inside the waste drums. Figure 6.3 shows the effect on calculated activity of random positioning of a single ^{60}Co source in a standard Risø drum filled with a waste material with density 1 kg/L. An underestimation of the activity with a factor of about 0.7 might occur. The effect is even more pronounced for a ^{137}Cs source. However, a signal from a combination of sources is a more likely situation and similar calculations with random position of many sources reduces the underestimation of the activity.

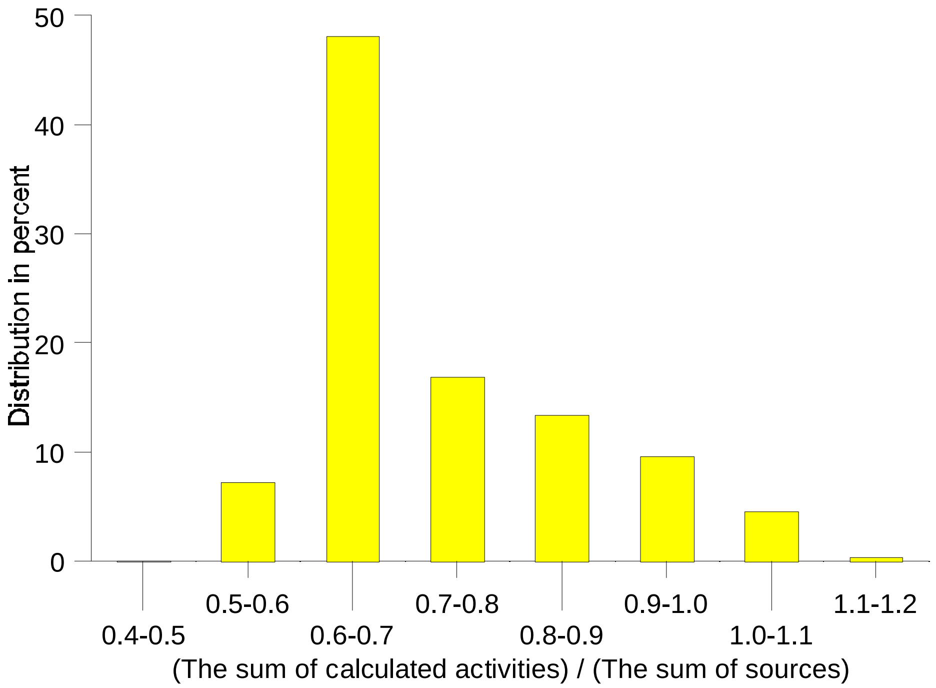

Figure 6.3. The ratio between calculated activity and actual activity for a single ^{60}Co source randomly positioned in a waste drum.

For inventory calculations correction factors of 0.85 for ^{60}Co and 0.8 for ^{137}Cs will therefore be used to compensate for the lack of knowledge about the distribution of activity in actual waste units.

Additional α-analyses of bituminized evaporator concentrates from the Risø plant were performed, but the results have not yet been reported.

6.2.2 Properties of Bituminized Materials

The final report for an EU contract on "Characteristics of Bituminized Waste" was prepared, covering the period March 1992 to March 1996. The experimental work was finalized early 1996 but development of the supporting models was continued during most of the year. A separate report on the modelling will be issued in 1997. A paper summarizing results from more than 10 years Risø-studies of water uptake, swelling and leaching from radioactive bituminized materials containing soluble salts is in preparation.

The studies have given an improved understanding of the mechanisms determining behaviour of water exposed bituminized waste. This is important for formulation of the source terms for activity release needed in safety assessment of disposal of bituminized evaporator concentrates. This type of waste is produced as a α-contaminated product with medium-level activity at some reprocessing plants and as a low-level material at various European research centres, including Risø.

An visiting American PhD student has - among other topics - been working on leaching of plutonium from such bituminized material.

6.2.3 Properties of Cementitious Materials

The final report for an EU contract on "The Performance of Cementitious Barriers in Repositories" mentioned in the previous annual report has been delivered to the CEC, but is still queuing for publication.

In the meanwhile a new EU contract within the same field has been signed. Experimental work is starting in 1997, but a few preparatory experiments have been carried out in 1996 to investigate the retention of radioisotopes in cracked concrete barriers where the internal surfaces are covered by calcite deposits.

A numerical model of deposition of calcite inside such cracks was developed during the previous contract. A description of the model has been written and will be issued in 1997. The model is based on simplified cement chemistry, but it gives results in reasonable agreement with experiments. Time has not yet permitted a planned extension to more realistic conditions including the dissolution behaviour of hydrated calcium silicate.

The above mentioned American PhD student has also looked at Sr-isotope retention in precipitating iron hydroxides from corrosion of steel. This will be combined with the studies on cracked concrete behaviour representing a situation which must be considered typical in disposal facilities for low- and medium-level waste.

6.2.4 Nordic cooperative project

Within the framework of the Nordic Nuclear Safety programme a particular project, NKS-AFA1, is aimed at waste management practices, waste inventories and safety assessment methodologies used in the Scandinavian countries.

Risø is contributing with data about the Danish waste and with experience gained in the studies mentioned in Sections 6.2.1 to 6.2.3.

The project is seen as a valuable opportunity to follow the comprehensive work carried out in Sweden and Finland, and - more in line with Danish requirements - the Norwegian preparations for a repository for low-level waste.

6.3 Soil Chemistry

Considerable work on geochemistry related to transport of radionuclides from buried radioactive waste was previously carried out at Risø under contract with the EU. This particular effort has now practically disappeared due to lack of funding, changes in staff, etc.

However, the relation between soil chemistry and radioecology is of considerable interest for dose evaluation when radioisotopes from buried waste eventually are reaching the biosphere, and might represent a possibility for maintaining a certain expertise within the related fields of soil chemistry and geochemistry.

In 1996 work on a model of the cycling of the major elements in soils and plants in a forest area was finalized. A general description is available together with a first comparison between calculations and measurements at Klosterhede, an experimental forest site in western Jutland.

The model has been developed for the NECO project (Nitrogen Deposition, Turnover and Effects in Coniferous Forests Ecosystems) financed by the Danish Strategic Environmental Research Program. The main emphasis has been on the nitrogen system, but the SAMUS model (single area model of unsaturated soil) is quite general and can be used to study cycling of other elements as well.

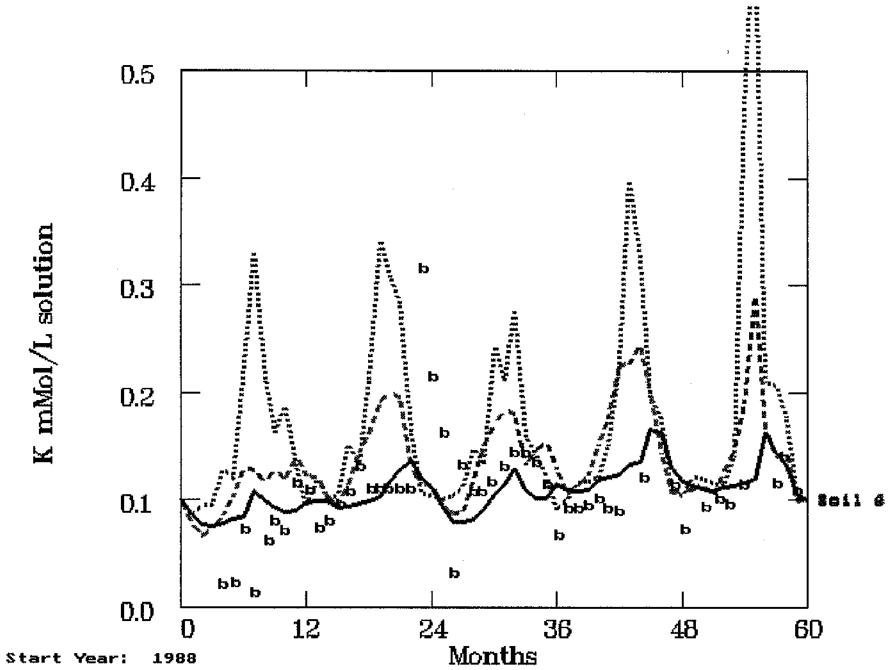

Figure 6.4. Calculated seasonal variation in potassium concentration in soil solution from the Klosterhede site. The upper curves represents solutions at about 5 cm, the middle one at ~15 cm and the bottom one at ~50 cm depth below the surface. The 'b's represents analyses of samples from the bottom layer.

The model might for example be modified to include radioisotopes.

From the large variety of curves obtainable from the model Figure 6.4 shows calculation results for potassium concentrations in the soil pore solution compared with measurements from the Klosterhede site. The annual peaks are due to a combination of increased microbiological decay of litter material and partial dry out in the summer season. This variation should also to some degree be reflected in the downward movement of radionuclides.

6.4 Publications

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Bibliographic Data Sheet **Risø-R-984(EN)**

Title and authour

Nuclear Safety Research and Facilities Department
Annual Report 1996

Edited by B. Majborn, K. Brodersen, A. Damkjær, H. Floto, K. Heydorn and P.L. Ølgaard

ISBN	ISSN
87-550-2303-7	0106-2840 1396-3619

Department	Date
Nuclear Safety Research and Facilities	April 1997

Pages	Tables	Illustrations	References
56	2	28	

Abstract (max. 2000 characters)

The report presents a summary of the work of the Nuclear Safety Research and Facilities Department in 1996. The department's research and development activities are organized in three research programmes: Radiation Protection, Reactor Safety, and Radioanalytical Chemistry. The nuclear facilities operated by the department include the Research Reactor DR3, the Isotope Laboratory, the Waste Treatment Plant, and the Educational Reactor DR1. Lists of staff and publications are included together with a summary of the staff 's participation in national and international committees.

Descriptors INIS/EDB

INTERNATIONAL COOPERATION; PROGRESS REPORT; RADIATION PROTECTION; RADIOACTIVE WASTE MANAGEMENT; REACTOR PHYSICS; REACTOR OPERATION; REACTOR TECHNOLOGY; RESEARCH PROGRAMS; RISOE NATIONAL LABORATORY

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