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ScienceDirect
Procedia Environmental Sciences 2 (2010) 713–719
Procedia Environmental Sciences

International Society for Environmental Information Sciences 2010 Annual Conference (ISEIS)

# Numerical modelling for the interpretation of a laboratory mock-up experiment of bentonite/granite interface

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**Abstract**

Performance assessment of a deep geological repository requires understanding diffusion and determining diffusion parameters under real conditions because diffusion is a key transport mechanism in hosting geological formation. FEBEX (Full-scale Engineered Barrier Experiment) is a demonstration and research project dealing with the bentonite engineered barrier designed for sealing and containment of the high-level radioactive waste repository. To support field investigations of FEBEX in situ test, a large–scale laboratory mock-up experiment (MUE) is being performed at CIEMAT facilities to study tracer migration at the bentonite/granite interface. Numerical models of MUE are presented here for HTO, $^{36}Cl^-$ and $^{137}Cs^+$. Experiments are modeled with 2-D axi-symmetric finite element grids and are solved with CORE2D V4. Model results indicate that numerical solutions with reference parameters reproduce measured data for HTO and $^{36}Cl^-$ but show large discrepancies for $^{137}Cs^+$. Relevant diffusion and retention parameters are identified by sensitivity analysis for tracer concentrations in borehole, bentonite and granite, respectively. Interpretation of $^{137}Cs^+$ data measured in the tracer chamber is perfomed by taking into account the uncertainties in initial activity $C_0$ and initial time $t_0$. Optimum values of $C_0$ and $t_0$ are obtained. The best fit is obtained with De-filter equal to $2.03 \cdot 10^{-10}$ m$^2$/s and Kd-bentonite equal to 5 m$^3$·Kg$^{-1}$.



*Key words*: mock-up experiment; bentonite/granite interface; diffusion, numerical model; sensitivity analysis; tritium; chloride; cesium.

## 1. Introduction

The solution to protect people and the environment against the radiations emitted by radionuclides contained in high-level radioactive wastes (HLW) consists on isolating them in such a way that radionuclides are not released to biosphere along any of the possible paths[1], [2],[3]. Usually, such radioactive high-level wastes (HLW) are proposed to be disposed in deep geological repositories which are currently favoured by many countries such as Belgium, France, Finland, Germany, Japan, Spain, Sweden, Switzerland and United States. Geological disposal is based on the concept of retention of waste by a combination of engineered containment barriers within a geological barrier. This provides a series of multiple engineered barriers comprising the solid conditioned waste-form, the waste container and any waste overpack (collectively referred to as the waste package), a buffer made of a material such as clay, grout or crushed rock that separates the waste package from the host rock together with any tunnel linings and supports. The geological barrier supports the engineered system and provides stability over the long term during which time radioactive decay reduces the levels of radioactivity. Salt deposits, clay and hard rock (granite) formations are considered as potential host rocks for radioactive waste repositories. Many designs for geological disposal facilities envisage the use of bentonite as an 'engineered barrier'. The stability and containment function of the overall geological repository system must be ensured for geological time scales. This highlights the relevance of the rock formation hosting the repository as the

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doi:10.1016/j.proenv.2010.10.081

ultimate barrier retarding the release of radioactivity to the biosphere. Thus, the knowledge and quantification of the processes that govern the migration/retention of radionuclides through the geosphere constitutes a key cornerstone in assessing the long-term future performance of nuclear waste repositories.

Laboratory experiments at small-scale are performed to improve our understanding of diffusion processes and determine key diffusion and retention parameters [4], [5]. Mock-up experiment performed at bentonite/granite interface is such an experiment.

In this paper, a numerical model for the interpretation of a laboratory mock-up experiment at is presented. This research work is part of the FUNMIG (Fundamental Process of Radionuclide Migration) European Integrated Project. This project aims at studying and modelling the fundamental migration process in the far field of a HLW repository. A large–scale laboratory mock-up experiment is carried out by CIEMAT to provide laboratory support for field investigations at FEBEX in situ test gallery. Scoping calculations for the design of the experiment are presented in Samper et al [5], [6].The main objective of this work is to study the mass transfer from the bentonite to granite and to identify the relevant transport parameters for critical radionuclides from laboratory experiments.

## 2. Mock-up experiment

Cylindrical rock blocks were obtained from the FEBEX gallery (Fig. 1) near the location of heater 1 which was dismantled in 2002. Two of such blocks (RB0-1 and RB15-1) are from granite region whereas the third one (RB27-2) was taken from lamprophyre area. The Mock-up experiment was performed on block RB0-1 (see Fig. 1).

A cylindrical block of granite of 38.8 cm of diameter and 30 to 40 cm long was obtained from FEBEX gallery. Tracer migration at bentonite/granite interface was mimicked by placing a small cylindrical plug of compacted FEBEX bentonite at the centre of the granite block. After installing the sampling system, the whole block was immersed in a bath with groundwater to maintain water hydration. A series of small boreholes were drilled for water sampling and monitoring tracer migration.

Tracers were injected in a column filter placed in the centre of the bentonite cylinder. Tracers include: HTO (conservative), 36Cl- (conservative and suffering anion exclusion) and 137Cs+ (sorbing).

Sixteen small boreholes (16 mm) with stainless steel sampling filters were placed perpendicular to bentonite at different radial distances from clay and at different elevations. This configuration allows for 3D monitoring of tracer migration. The scheme of the test with the location of monitoring boreholes drilled in granite block is shown in Fig. 2.

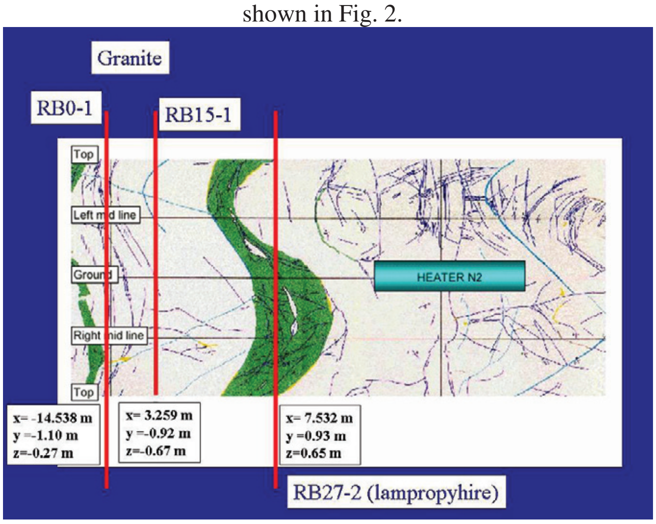



*Fig. 1.* FEBEX gallery with location of rock samples (left) and block RB0-1 used for mock-up experiment (right).

## 3. Numerical model

Mock-up experiment has been simulated with CORE2D V4 [7]. Given the symmetry with respect to sample axis, numerical calculations of MUE are performed with a 2-D axi-symmetric anisotropic model. Figure 3 shows the model geometry and the 2-D finite element grid which considers four material zones: 1) Tracer section where tracers are injected, 2) Filter, 3) Bentonite and 4) Granite. Experiments have been modelled using an approach similar to that used for modelling diffusion experiments in Opalinus clay [8].

Bentonite parameters adopted for scoping calculations were derived from previous CIEMAT experiments on FEBEX bentonite [9]while those of granite were taken from CIEMAT experiments on granite columns [10]. They are listed in Table 1. Initial tracer activities are equal to zero everywhere except in the borehole where activities per unit volume of water were prescribed according to known total tracer activities. The borehole containing the tracer does not represent a constant-concentration boundary condition, but a fixed initial-mass boundary condition. A no-flow condition is used in all model boundaries because tracers cannot leave the granite block.

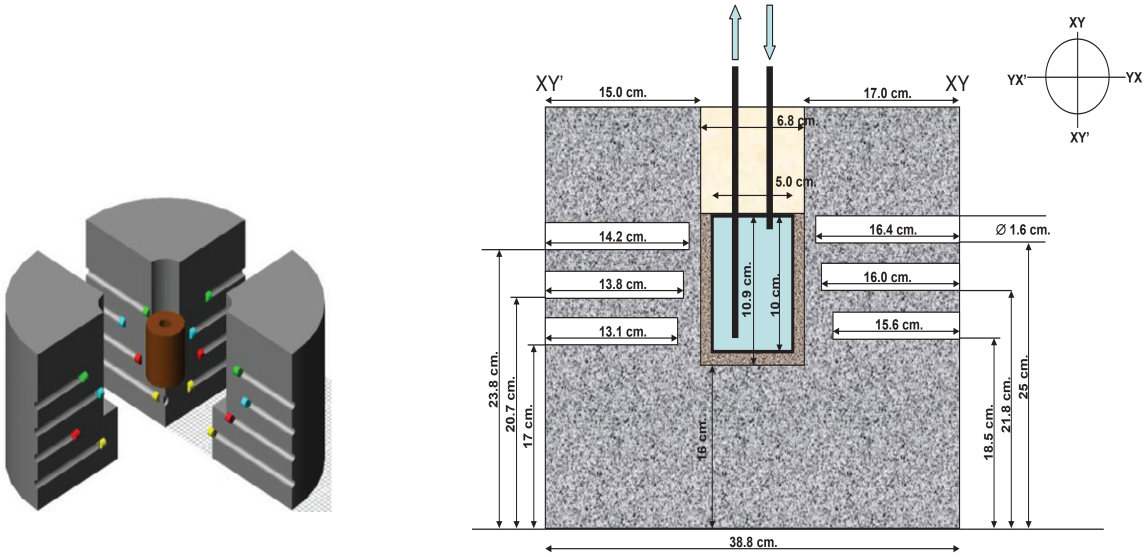



***Fig. 2.*** Scheme of MUE (left) with indication of final geometry (right).

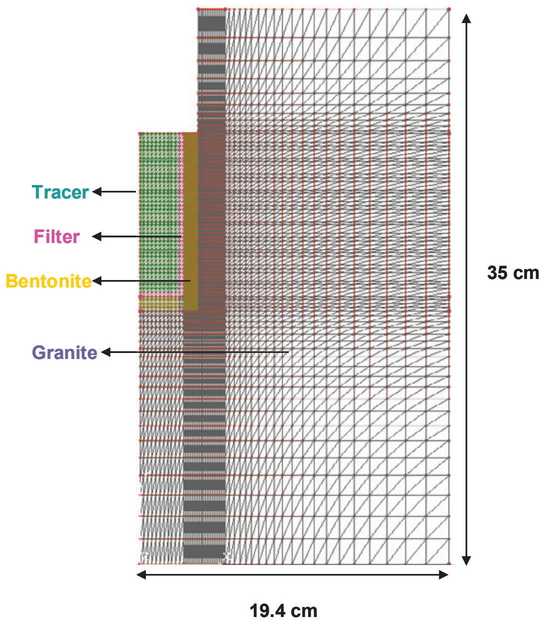



***Fig. 3.*** 2D finite element grid used for the numerical model of MUE.

*Table 1.* Reference transport parameters of HTO, $^{36}Cl^{-}$ and $^{137}Cs^{+}$ for bentonite (B), granite (G) and filter (F) used for scoping calculations (porosity of Filter from García-Gutiérrez, personal communication. $D_e$ of Filter is computed from those in bentonite with Archie´s law and an exponent of 1.3).

| | Porosity | | | $K_d$ (m$^3$/kg) | | $D_e$ (m$^2$/s) | | |
|---|---|---|---|---|---|---|---|---|
| Tracer | B | G | F | B | G | B | G | F |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| HTO | 0.57 | 0.01 | 0.5 | 0.00 | 0.00 | $5.8\cdot10^{-11}$ | $1.5\cdot10^{-13}$ | $4.87\cdot10^{-11}$ |
| $^{36}Cl^-$ | 0.05 | 0.005 | 0.5 | 0.00 | 0.00 | $9.3\cdot10^{-13}$ | $2.3\cdot10^{-14}$ | $7.81\cdot10^{-13}$ |
| $^{137}Cs^+$ | 0.57 | 0.01 | 0.5 | 1 | 0.225 | $8.03\cdot10^{-10}$ | $6.0\cdot10^{-13}$ | $6.76\cdot10^{-10}$ |

Note: B=Bentonite, G=Granite, F=Filter

## 4. Numerical results

Time evolution of tracer concentrations was calculated at all sampling sections for all tracers. Fig. 4 illustrates the time evolution of normalized tracer concentrations at the tracer injection section. Chloride is an anion suffering anion exclusion and its concentration after 1 year is 99% of its initial value while neutral HTO has decreased to 95 %. $^{137}Cs^+$ reduces to 0.9% of its initial concentration.

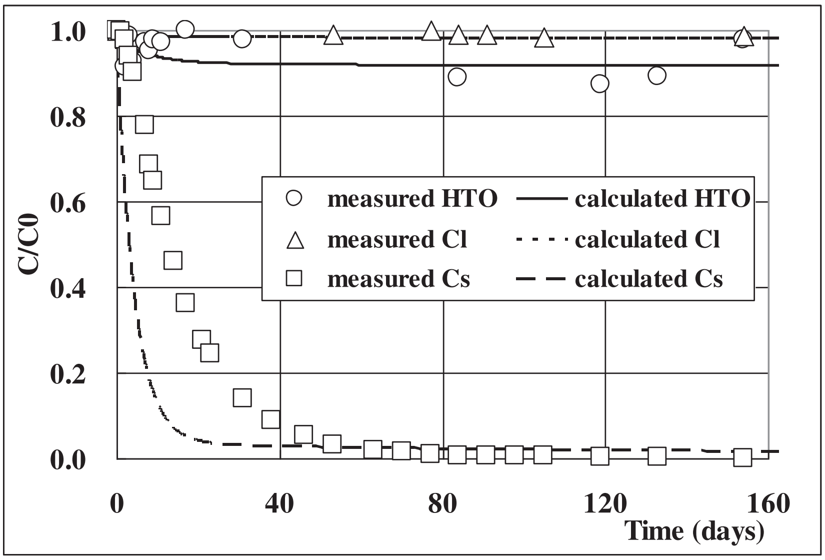



***Fig. 4.*** Time evolution of normalized tracer concentrations in tracer section, measured (symbols) and computed (lines).

Measured data in tracer section shows a lot of scatter for HTO. Chloride data remains constant and does not show major changes. Computed HTO and $^{36}Cl^-$ values with reference parameters reproduce the measured data. However, there is a clear discrepancy for $^{137}Cs^+$ between calculated values and the measured data, indicating $^{137}Cs^+$ data in the injection chamber need to be interpreted (Fig. 4).

$^{137}Cs^+$ concentrations in borehole are clearly sensitive to the effective diffusion coefficient of the filter (Fig. 5). They are slightly sensitive to the distribution coefficient of bentonite (Fig. 6) and lack sensitivity to the distribution coefficient and effective diffusion coefficient of granite (figures not shown here).

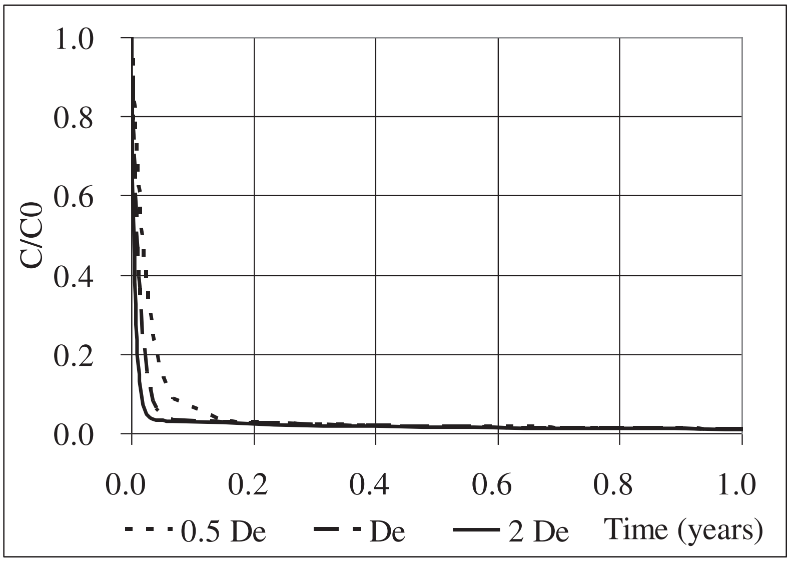



***Fig. 5.*** Sensitivity of $^{137}Cs^+$ concentration in the tracer section to changes in effective diffusion coefficient of filter.

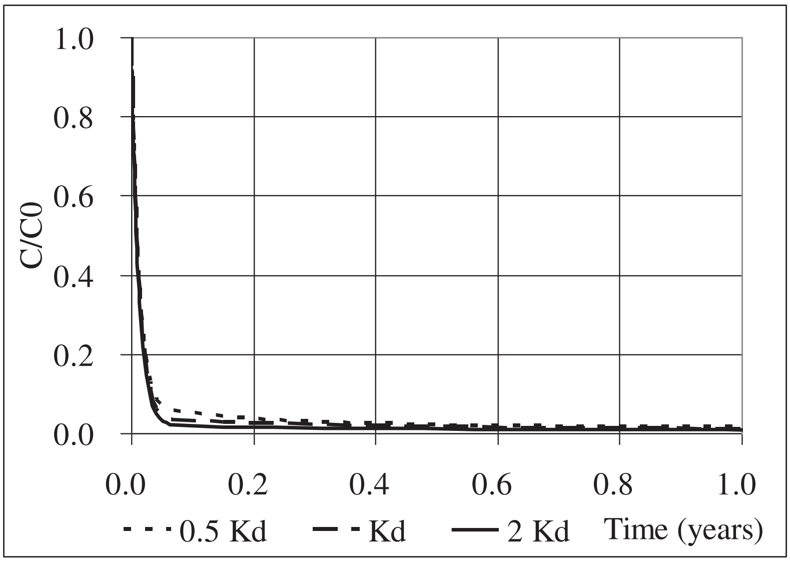
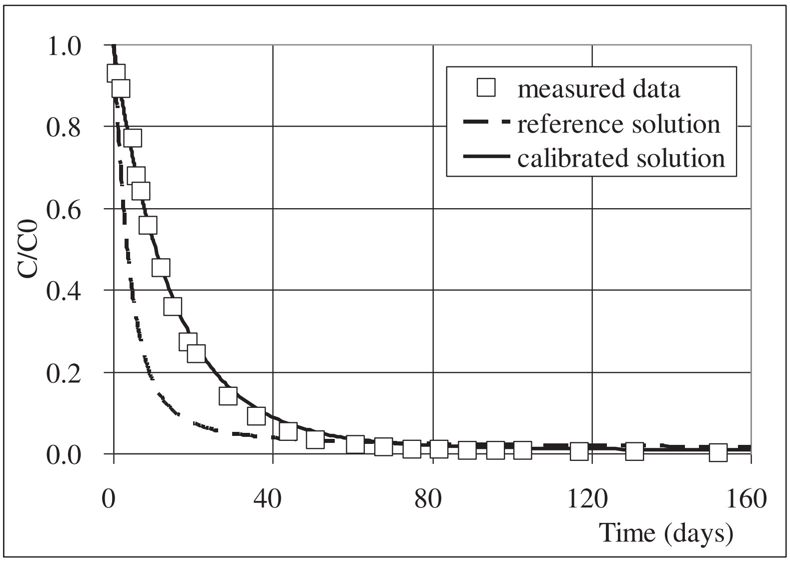



***Fig. 6.*** Sensitivity of $^{137}Cs^{+}$ concentration in the tracer section to changes in distribution coefficient of bentonite.

Based on the results of sensitivity analysis, measured $^{137}Cs^{+}$ data in the injection chamber have been interpreted by estimating De-filter and Kd-bentonite and taking into account of the uncertainties in the values of initial activity $C_0$ and the value of $t_0$ (time at which tracer injection starts). Results indicate that the optimum $C_0$ is equal to 19000 cpm/mL (the value of the raw data is 18752.05 cpm/mL) and $t_0$ equal to the raw data time minus 2 days. The difference of the initial time indicating that there is a lag of time about 2 days between the diffusion occurred and measurement performed. A perfect fit is obtained for $^{137}Cs^{+}$ data with De-filter of 2.03 ·10-10 $m^2/s$ (0.3 times its reference value) and Kd-bentonite of 5 $m^3 \cdot Kg^{-}$ (5 times its reference). Fig.7 illustrates the comparison between model results and measured data, also shown is the reference solution for the purpose of comparison. It should be noticed that $^{137}Cs^{+}$ data in injection system provide information only about filter and bentonite parameters but not on granite parameters.



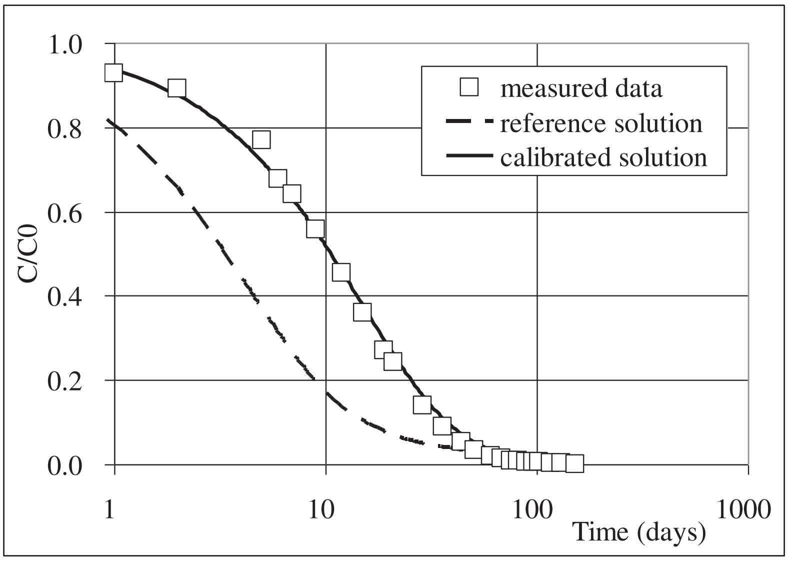



***Fig. 7.*** Comparisons of measured (symbols) and computed (lines: with reference and calibrated parameters) $^{137}Cs^{+}$ normalized concentrations.

## 5. Conclusions

A 2-D axi-symmetric model has been used to analyze tracer diffusion across the bentonite/granite interface. Numerical solutions of HTO and $^{36}Cl^{-}$ reproduce the measured data with reference parameters. Results of sensitivity analysis show that normalized $^{137}Cs^{+}$ concentrations in the tracer section are very sensitive to effective diffusion of filter and slightly sensitive to distribution coefficient of bentonite, but no sensitivity to the parameters of granite, indicating the uncertainties in granite parameters do not affect the estimation of filter and bentonite parameters.

There are some uncertainties in filter properties which is known affect significantly $^{137}Cs^{+}$ dilution. Interpretation of $^{137}Cs^{+}$ dilution shows it is controlled mostly by effective diffusion of filter and slightly affected by distribution coefficient of bentonite. Optimum value of $C_0$ is equal to 19000 cpm/mL and t0 equal to the raw data time minus 2 days. The best fit is obtained for $^{137}Cs^{+}$ with De-filter equal to 2.03 ·10-10 $m^2/s$ and Kd-bentonite equal to 5 $m^3 \cdot Kg^{-1}$. $^{137}Cs^{+}$ data in the injection system provide information only about filter and bentonite parameters but not on granite.

It should be noted that there exist uncertainties for the tracer activities induced by mixing in the sampling points. Measured activities in a sampling point are not the real diffusion activities instead of the activities suffering mixing. The numerical model presented here dose not account for the influence of mixing, such a model will be improved in the next work.

## Acknowledgements

This work has been funded by ENRESA and EU within FUNMIG (FUNdamental Processes of radionuclide MIGration) Project (Ref. FP6-516514) and also project of Safety evaluation of groundwater resources and research on pollution prevention and control technology in Beijing (D07050601510000). Partial funding for UDC contribution has been obtained also from Spanish Ministry of Science and Technology (CICYT Project CGL2006-09080), Xunta de Galicia (Incentive PGIDT05PXIC11801PM). We acknowledge Prof. Javier Samper, Miguel García-Gutiérrez, Tiziana Missana and Manuel Mingarro for providing experimental data and the contribution of S. P. Yi for final Cesium model fit.

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