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Äspö Hard Rock Laboratory

Test plan for the long term diffusion experiment

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October 1999

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This report concerns a study which was conducted for SKB. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

Abstract

A test plan has been developed for the Long-Term Diffusion Experiment (LTDE). The objective of the experiment is to study the magnitude and extent of matrix diffusion from a natural fracture surface, through the altered zone, into the intact matrix rock. Further, to compare obtained *in situ* values on diffusivity with that obtained from corresponding studies on core samples in the laboratory. The experimental concept involves drilling of a telescoped large-diameter borehole to intercept a target feature identified in a small diameter pilot borehole. The feature is packed off using a special packer which seals around the developed core stub. The borehole is further packed off with mechanical and inflatable packers to avoid effects of the acting hydraulic gradient. A tracer solution consisting of conservative and sorbing radioactive tracers will be circulated in the section with the natural fracture for a period of 3-4 years. A penetration due to diffusion in the order of decimetres is expected over the duration of the experiment. Subsequently, the rock volume subject to diffusion will be over-cored, sectioned and analysed for tracer activity/concentration. The *in situ* experimentation is supported by various types of mineralogical, geochemical and petrophysical analyses.

Sammanfattning

En testplan har tagits fram för Long-Term Diffusion Experiment (LTDE). Målet med försöket är att studera storleken på, och utsträckningen av, diffusion från en naturlig sprickyta, genom den omvandlade zonen, in i den intakta bergmatrisen. Vidare att jämföra värden från diffusivitet erhållna från fältförsök, med de värden som erhållits från laboratorieförsök på kärnprover. Det experimentella upplägget inkluderar borrhning av ett teleskoputformat borrhål med stor diameter där en identifierad målstruktur, som identifierats i ett närliggande pilotborrhål av mindre diameter, genomborras på valt djup. Målstrukturen isoleras med hjälp av en koppformad manschett som omsluter den kvarvarande kärstubben i borrhålet. Borrhålets yttre delar är manschetterad med mekaniska och uppblåsbara manschetter för att undvika att en hydraulisk gradient utbildas. En blandning av icke sorberande och sorberande radioaktiva spårämnen cirkuleras genom sektionen med den naturliga sprickan under en period på 3-4 år. Genomförda beräkningar visar att diffusionsdjup i storleksordningen några decimeter kan påräknas under experimentets varaktighet. Därefter överborras den intressanta bergvolymen och analyseras med avseende på koncentration/aktivitet. Fältdelen av experimentet understöds av olika mineralogiska, geokemiska och petrofysiska analyser.

Contents

Abstract	i
Sammanfattning	i
Contents	ii
1 Background	1
2 Objectives	2
3 Rationale	3
3.1 Relevance to repository performance	3
3.2 Current state of knowledge	3
3.3 Justification for the experimental work	6
4 Experimental Concept	8
4.1 Background	8
4.2. Experimental configuration	9
4.3 Expected outcome	12
4.3.1 Scoping calculations	12
4.4 Problem areas	16
4.4.1 Site-related problems	16
4.4.2 Experimental problems	17
5 Scope	19
5.1 Main project tasks	19
5.1.1 Site selection	19
5.1.2 Drilling of boreholes	19
5.1.3 Characterisation in boreholes	21
5.1.4 Geological characterisation of the drill core.	22
5.1.5 Petrophysical characterisation of core samples	23
5.1.6 Assessment of sample disturbance in core samples	23
5.1.7 Cleaning of boreholes	23
5.1.8 Instrumentation of boreholes	24
5.1.9 Supporting laboratory diffusion experiment	27
5.1.10 Selection of tracers	27
5.1.11 Sampling of the injection borehole	30
5.1.11.1 Analyses of tracer content in the drill core	30
5.1.12 Overcoring drilling	32
5.1.13 Analyses of tracer content in the drill core	32
5.1.14 Microbial investigations	32

6.	Time schedule	34
7	Project organisation and resource requirements	35
	7.1 Introduction	35
	7.2 Organisation	35
	7.3 Data management	36
	7.4 Reporting	36
8	References	37

1. Background

Transport of radionuclides in rock fractures is presently studied within the TRUE experimental programme. Specific matrix diffusion studies will require a somewhat modified experimental approach. Based on results from laboratory experiments (Byegård et al. 1998), the expected low matrix diffusivities for different radionuclides will make it difficult to observe matrix diffusion in the ongoing and planned dynamic experiments in the TRUE programme, given the practical time constraints of the planned experiments. Hence, a static long term diffusion experiment is proposed.

The chemical reactions involved in sorption of radionuclides can according to a simplistic view be divided in ion exchange, inner-sphere complexation and outer-sphere complexation. The mobility of radionuclides is strongly affected by the interaction with geologic materials. The transport of dissolved solutes can be regarded as a distribution between species in solution (ionic species, inorganic- and organic-complexes), mobile solid phases (particulate matter, colloids, precipitates etc.) and stationary solid phases (minerals). The distribution is often expressed as a distribution coefficient, K_d , which is an equilibrium constant for a reversible sorption process. Kinetic effects and irreversible sorption mechanisms are thus not included in a K_d value. Since most of the radionuclides in nuclear waste during their transport in the rock matrix are influenced by any of the sorption mechanisms mentioned above it is possible to take advantage of a combined diffusion and sorption experiment. By using sorbing radionuclides in a diffusion experiment one will automatically obtain a fixation of the tracer in the diffusion pathways. It is however important that one knows the sorption behaviour of the tracers, (e.g. specific sorption mechanisms, preferential "sites" and degree of reversibility or kinetics) in order to evaluate the diffusion pathways.

Matrix diffusion studies of radionuclides have been performed in several laboratory experiments (reviewed by Ohlsson and Neretnieks 1995) and also *in situ* (Birgersson and Neretnieks, 1990). Some experimental conditions such as pressure and natural groundwater composition are however difficult to simulate in a laboratory experiments. Investigation of rock matrix diffusion in laboratory scale implies that one uses rock specimens in which damage due to drilling and unloading effects (rock stress redistribution) may have caused irreversible changes of the rock properties. Investigation of matrix diffusion in non-disturbed rock is therefore preferably investigated *in situ*. Through the proposed experimental technique one will also get some information of the adsorption behaviour of some radionuclides both on natural fracture surfaces and on fresh granitic rock surfaces.

Scoping calculations for the planned experiment are made using the multi-rate diffusion concept which accounts for pore heterogeneity at different scales (Haggerty and Gorelick, 1998, Haggerty, 1999)

A proposal for a matrix diffusion experiment (MDE) for Äspö HRL has previously been presented by Birgersson et al. (1994).

2. Objectives

The objectives of the Long-Term Diffusion Experiment project (LTDE) are:

- To investigate the magnitude and extent of diffusion into matrix rock from a natural fracture *in situ* under natural rock stress conditions and hydraulic pressure and groundwater chemical conditions .
- To obtain data on sorption properties and processes of individual radionuclides on natural fracture surfaces and internal surfaces in the matrix.
- To compare laboratory derived diffusion constants and sorption coefficients for the investigated rock fracture system with the sorption behaviour observed *in situ* at natural conditions, and to evaluate if laboratory scale sorption results are representative also for larger scales.

3. Rationale

3.1 Relevance to repository performance

A deep repository for radioactive waste in granitic rock will not only serve as a long term physical barrier sheltering the waste containment in geological time scales, it will also act as a chemical barrier preventing migration of several radionuclides. A chemical barrier could be of significance in the case of waste containment failure releasing radionuclides to the groundwater. Transport of radionuclides by groundwater will occur by water flow in fractures and by diffusion in the pores of the rock matrix. In the case the pores are connected to each other, the rock matrix will serve as an important retardation barrier for radionuclides. The total amount of surfaces available for sorption will be much higher in the case of matrix diffusion, compared to the case of only fracture surface sorption. In fact, matrix diffusion is considered the most important factor for the natural granitic bedrock barrier for the repository performance (Neretnieks 1980).

3.2 Current state of knowledge

The impact of matrix diffusion on the migration rate of substances released from a leaking repository of spent nuclear waste was first pointed out by Neretnieks (1980). Matrix diffusion has mainly been studied in laboratory experiments and an elaborate review is given by Ohlsson and Neretnieks (1995). The main criticism against laboratory studies is that the rock samples used have undergone sample disturbance in the form of mechanical damage stress unloading effects which may have increased the porosity and the open pore connections. However, attempts to study the matrix diffusion by increasing the rock stress showed that the diffusion rate was only decreased by 20-70% (Skagius and Neretnieks, 1986). The degree to which the natural pore characteristics were reinstated can be a possible source of error. Laboratory experiments furthermore lack from the disadvantage that they can not reflect the time scale relevant for a repository of spent nuclear waste. It is therefore very important to combine the knowledge obtained from laboratory experiment with qualitative and quantitative information of matrix diffusion originating from natural analogue studies. A review of matrix diffusion studies in natural analogue studies is presented by Ohlsson and Neretnieks (1995). Smellie and Karlsson (1999) discuss the use of natural analogues in quantifying matrix diffusion and conclude that:

- From natural analogue studies there is a general consensus that matrix diffusion actually do occur.
- Penetration depths of mm:s to cm:s suggest interconnected pore systems within the rock mass adjacent to fracture systems
- Few analogue data exist from which to derive diffusion coefficients and those data that are available indicate a confusingly large range of values (10^{-19} to 10^{-9} m²/s)
- Laboratory-derived data must thus still be used for repository performance assessment, although they should be regarded as non-conservative because of the possibility of experimental artefacts.

An additional possibility of obtaining matrix diffusion information is to carry out laboratory experiments *in situ*. Such experiments have the advantage that they offer the possibility of maintaining the natural conditions with respect to groundwater composition and rock stress, respectively. The disadvantages are that a comparatively short time scale can be applied and that necessary excavation processes (drilling, etc.) may cause experimental artefacts. The latter is attributed to minor disturbance exerted close to the borehole. This disturbance is analogous to the disturbance developed around a tunnel excavated using a TBM machine. The results from the ZEDEX project at Äspö HRL (Emsley et al., 1998) indicate that the mechanically damaged zone is about 1% of the tunnel diameter (5m). The zone affected by stress redistribution (independent of excavation method) was in the case of ZEDEX found to extend radially 0.05-0.1 diameters from the tunnel perimeter.

In a similar way, sample disturbance along the periphery of a drill core is envisaged due to the mechanical damage of the drill bit and disturbance due to stress relief, respectively. Of the two, the former is expected to have the highest impact on possible enhancement of diffusivity. The damaged zone is expected to extend not more than 1% of the diameter of the core. Very little work has been made on disturbance along the periphery of a core specimen. Attention has however been put on “core discing” observed mainly in high-stressed rock (Ichida et al., 1995, Li, 1997), cf. Figure 3-1. An extensive literature review and numerical modelling of core damage and interpretation of in-situ stress is presented by Hakala (1999).

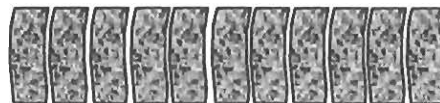


Figure 3-1 Schematic representation of core discing in high-stressed rock.

In the case of a remaining core “stub” in a borehole, so called “spalling cracks” may be formed in a high-stressed environment, cf. Figure 3-2. In the case of the situation at Äspö (assuming an uni-axial compressive strength $\sigma_c \sim 200$ MPa, such fractures (or embryos in the form of connected micro-cracks) are not expected to occur until an *in-situ* stress level of 80-100 MPa is reached (Li, pers. comm.). In this context it should be noted that the *in-situ* rock stress levels at this depth at the Äspö HRL are in the range 6-25 MPa.

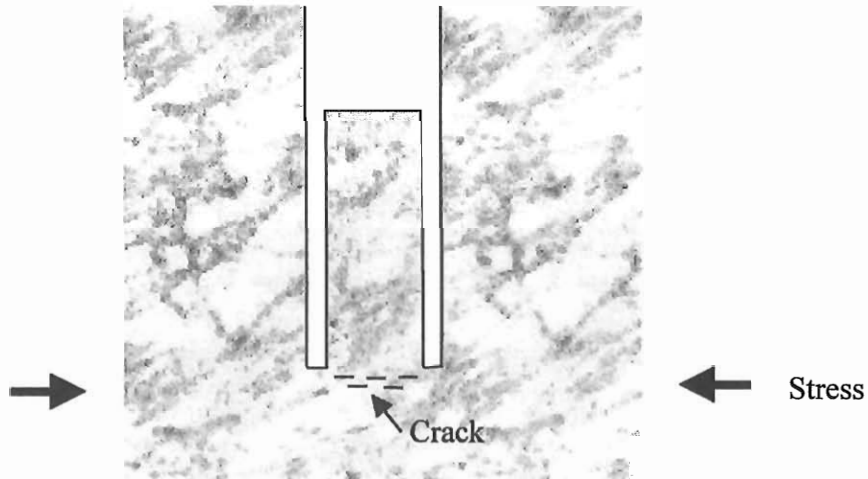


Figure 3-2 Schematic representation of development of “spalling cracks” in high-stressed rock (Courtesy by Shun-Lin Li, Technical University of Luleå).

In a series of laboratory experiments using He-gas targeted on effects of sample preparation (sawing) (Hartikainen et al., 1997) it is shown that the effect on porosity, diffusivity and permeability by successive sample reduction (approx. 60→30→15 mm) is the highest on the smallest samples.

Hartikainen et al. (1995) also reports results from a study where the successive change in porosity and diffusivity away from a natural fracture surface in a sample from the Finnish Palmottu site. The methodology employed He-gas and successive sample reduction, similar to the one outlined above. The results indicate increased values of porosity and diffusivity up to about 50 mm from the fracture surface into the rock matrix of the 78 mm long specimen. Diffusivity remains at an elevated level until a rapid drop is noted. The reduction in porosity is exponential, suggesting that no clear correlation between porosity and diffusivity in the altered zone.

3.3 Justification for the experimental work

So far, the only *in situ* experiment reported directly addressing matrix diffusion in granitic rock was performed in the Stripa mine by Birgersson and Neretnieks (1990). The tracers uranine, iodide and Cr-EDTA were injected to an isolated borehole section in granite during a contact time of <3.5 years. After the tracer solution had been removed, overcoring of the injection borehole was performed and the concentration profiles of the tracer in the rock matrix was measured. Measurable concentration of the tracers were found at distances <100mm from the periphery of the injection borehole. Severe variations in the concentration profiles were, however, observed in the different rock samples studied. Because of the existing hydraulic pressure gradient of the rock, the tracers were continuously injected with pressures 0.5-0.9 MPa over the natural water pressure. Thus, the experiment involved both advection and diffusion, which was subsequently addressed in the evaluation of the experimental data.

No *in situ* experiment can be found in the literature directly addressing matrix diffusion of sorbing tracers. The diffusivities determined in laboratory experiments found in the literature for substances present in the spent nuclear fuel (Olsson and Neretnieks 1995, references therein) are most often not very much lower the diffusivities of non-sorbing tracers studied in the same experiment (e.g., Ittner et al. 1990, Ittner et al. 1988,). In their work of recommending values for diffusion constants in granitic rock, Ohlsson and Neretnieks (1997) did not chose to concentrate on the experimentally obtained diffusivity values for the strongly sorbing substances. Instead the apparent diffusivities were calculated applying sorption data from Carbol and Engkvist (1997) in combination with diffusivities experimentally obtained for non-sorbing species. This resulted in diffusivities generally much lower than any experimentally obtained value. As an example, for Plutonium, Ohlsson and Neretnieks (1997) recommended an apparent diffusivity of $3 \cdot 10^{-18} \text{ m}^2/\text{s}$ applying a K_d of $5 \text{ m}^3/\text{kg}$ given by Carbol and Engkvist (1996). However, the lowest experimental value found in the literature is $1.6 \cdot 10^{-14} \text{ m}^2/\text{s}$ (Ittner et al. 1988). There are several explanations for the observed differences. The most important one is probably that penetration studies of sorbing tracers in diffusion experiments are disturbed by an unevenness and microfissures on the surface of the rock slab. The diffusivity may thus be overestimated; the obtained concentration profile may not have been caused by actual diffusion but instead be result of the unevenness of the rock. Furthermore, the database of Carbol and Engkvist (1996) is mainly extracted from batch sorption experiments and it has been shown (Johansson et al. 1997, Johansson et al. 1998) that batch sorption experiments tend to give higher sorption coefficients than what can be observed in diffusion experiments on corresponding geological material. The application of a batch sorption

K_d to calculate the apparent diffusivity of a tracer, requires that the porosity of the granite is homogeneously distributed in the rock matrix. Experiments studying the porosity distribution in the rock matrix have indicated a heterogeneous distribution of porosity in the matrix, e.g. Johansson et al. (1998).

Studies of the influence of the carbonate complexation on the diffusivity has recently been investigated by Yamaguchi and Nakayama (1998). Applying a water phase of 0.1M NaHCO₃ in a through diffusion experiment using a 0.5 cm thick granite slab, a breakthrough was obtained for Pu and U, which in their non-complexed form are considered to be strongly sorbing (e.g., Carbol and Engkvist (1996)). For Am studied under the same experimental conditions, no breakthrough was obtained. Using available thermodynamic data, the authors performed a speciation calculation and found that under the prevailing experimental conditions, the complexes Pu(IV)(OH)₂(CO₃)₂²⁻, U(VI)O₂(CO₃)₃⁴⁻ and Am(III)(CO₃)₃³⁻, respectively, should be the dominating species. The work indicates that complexation could play an important role for increasing the diffusivities of the actinides.

Mathematical solutions for the interpretation of the matrix diffusion as a heterogeneous process consisting of a multirate diffusion has been described by Haggerty and Gorelick (1995). The application of the model to interpret tracer experiment data from the Culebra dolomite at the WIPP site has been presented by McKenna et al. (1998). Application of this multirate model to the results of the TRUE-1 experiments at the Äspö Hard Rock Laboratory is also presently being conducted (McKenna 1999).

4. Experimental Concept

4.1 Background

Several concepts have been applied in the studying of matrix diffusion in laboratory experiments. An important part of the proposed experiment is to compare data and experimental concepts applied at the laboratory scale and the *in situ* scale. It would thus be an advantage if several of the laboratory experimental concepts presently proposed for extracting data for matrix diffusion could be tested in an analogous *in situ* configuration. A short review of the most commonly used laboratory experimental methods will therefore be presented.

In through-diffusion studies, a rock slab is mounted between two containers of water; one of the containers spiked with tracers, the other containing non-spiked water (e.g., Skagius 1986, Johansson et al. 1997). The concentration increase in the originally non-spiked water is followed in a long time perspective. From the concentration versus time plot, the effective diffusivity and the rock capacity factor (equal to the porosity in the case of using a non-sorbing tracer) can be evaluated. This technique has mainly been applied to diffusion of non-sorbing tracers, since the time to obtain data is long for a non-sorbing tracer (months) and can be very long (years) if sorbing tracers are used.

An alternative technique is to put the rock slab in the tracer spiked solution with one side of the slab in contact with the water and the others sealed. The diffusion is allowed to proceed for a certain time after which the slab is separated from the solution. The slab is thereafter ground or sawed and thereafter the concentration profile of the diffusing tracer is determined. This technique has mainly been applied to diffusion of sorbing tracers (e.g., Allard et al. 1985, Ittner et al. 1988) mainly since this technique offers possibility to study the penetration in mm per year scale; which is often obtained by sorbing tracers. Comparison of studies of penetration depth with through-diffusion technique has been conducted by Byegård et al. (1998). The penetration profile obtained for Ba and Cs in the penetration studies indicated that two diffusivities had to be applied in order to explain the results. The faster one of these two diffusivities correlated well to the diffusivity obtain in the through diffusion experiment.

A third tracer method of studying matrix diffusion is to let the spiked water diffuse into the rock matrix. The water phase is after a given time exchanged with non-spiked water and thereafter the concentration increase

caused by back-diffusion of the tracer is measured. An alternative is to study the out-diffusion of tracers naturally present in the matrix at the start of the experiment (Van der Kamp et al. 1996, Novakowski and Van der Kamp, 1996). The disadvantage of such a method is that the method measures the matrix diffusion indirectly; no actual observation of the tracers penetrating the rock matrix is observed.

Since tracer methods in order to measure matrix diffusion are rather time-consuming, alternative methods have been developed such as electrical resistivity measurements (e.g., Skagius and Neretnieks 1986, Ohlsson and Neretnieks 1998) and gas diffusion studies (Väätäinen et al. 1993). Since these methods are less time consuming, they can offer possibilities to measure a large number of rock samples and thus obtain diffusivity measurements and statistics over a large rock volume, e.g., over a repository. However, for the acceptance of these methods, intercalibration between them and conventional tracer diffusion experiments are essential (Ohlsson and Johansson, 1999).

Efforts have been made to use data from the uranium series disequilibrium in order to investigate matrix diffusion in crystalline rocks (e.g., Smellie et al. 1986). By measuring the different isotopes in the ^{238}U decay series as a function of the distance from a natural fracture, information of the matrix diffusion characteristics was obtained.

4.2 Experimental configuration

The concept presumes the existence of a pilot borehole (drilled within the SELECT-2 programme) which has identified a suitable fracture for further study. A large diameter borehole (approx. 200 mm in diameter, telescoped from a larger diameter) is drilled through the identified fracture plane some 10 - 20 m into the rock (Figure 4-1). In the bottom of the hole a stub including the target fracture plane remains. This stub, some 50 mm long, will be packed off and isolated so that only the fracture surface is in contact with the groundwater (same concept as used in the REX-experiment at the Äspö hard Rock Laboratory, Puigdomenech et al. 1999). The groundwater will be continuously circulated and injection of the tracers will be performed over a four year period. The intersection of the target fracture by the pilot borehole will be used as a pressure reference, i.e., the pressure in the experimental section will be set so that no pressure difference will be obtained along the experimental borehole. After the four years period, overcoring of the injection borehole and extraction of the stub will be performed. The axial diffusion through the fracture rim into the intact matrix rock will be studied by performing analyses of the penetration profile of the different tracers used. This should be done by extracting small diameter cores parallel to the diffusion direction which

should be cut in to small slices and analysed for their content of tracers. Furthermore, during the injection phase measurement of the tracer concentration should be conducted, either by on-line detection or by sampling.

Experimental concept

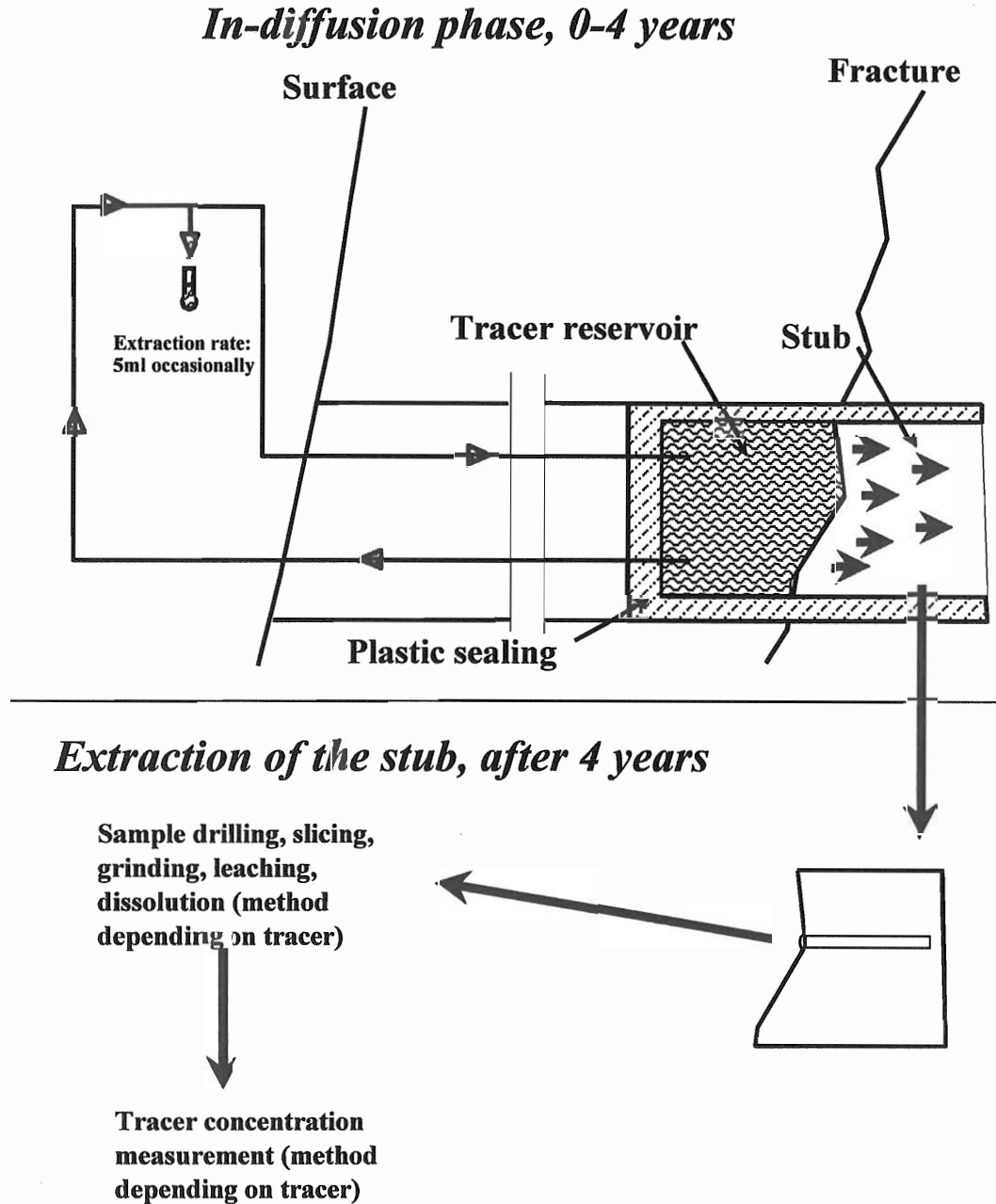


Fig. 4-1 Schematic of LTDE experimental concept including injection borehole in contact with a fracture surface, combined with excavation and penetration profile studies

4.3 Expected outcome

The project is expected to provide quantitative and qualitative information of the matrix diffusion under natural stress and groundwater conditions, i.e.;

- To obtain diffusivities for non-sorbing tracers from the rim zone of a natural fracture into a presumed low-conductive rock under natural stress
- To obtain information on the existence of any limitations in extension of connected pores
- To obtain information on the diffusivity of slightly sorbing tracers, i.e., alkaline metals and alkaline earth metals, and from comparisons with the laboratory sorption data obtain general information of the sorptivity of the surfaces in the pores in the fracture rim zone and the rock matrix.
- To obtain information for elements relevant for a repository performance assessment case, i.e., the effects of complexation, sorption and diffusivity under natural chemical conditions and under natural rock stress.
- Post-experiment assessment of possible sample disturbance.

4.3.1 Scoping calculations

Scoping calculations have been performed in order to what maximum penetration depth could be observed of different tracers in a 4 years diffusion experiment. In the calculations, Fick's second law of diffusion (Crank 1975) has been applied, i.e.;

$$\frac{\partial C}{\partial t} = D_a \cdot \frac{\partial^2 C}{\partial x^2} \quad (4.1)$$

For the case of a presumably strongly sorbing tracer, e.g., Cs, U, Np and Am, it has been assumed that all tracer amount will sorb on the fracture surface. A solution to the diffusion equation has thus been chosen which involves the total amount of tracer diffusing in a semi-infinite cylinder of the rock (Crank 1975), i.e.;

$$C = \frac{C_0 \cdot V \cdot \left(\frac{d_{\text{samp}}}{d_{\text{tot}}} \right)^2 \cdot l}{\sqrt{\pi \cdot D_a \cdot t}} \cdot e^{-\frac{x^2}{4 \cdot D_a \cdot t}} \quad (4.2)$$

where:

- C - the amount of tracer (Bq or g) in a l m thick disc sample with a diameter of d_{samp} m at a distance of x m from the fracture surface.
- C_0 - the concentration of the tracer in the groundwater directly after injection (Bq/dm³ or g/dm³)
- V - the total groundwater volume used in the experiment (dm³)
- d_{tot} - the diameter of the stub (m)
- D_a - the apparent diffusivity of the tracer (m²/s)
- t - the diffusion time (s)

For the presumably non- and very weakly sorbing tracers, a solution to the diffusion equation has been applied which involves the water phase as a constant source. The amount of tracer diffusing into the rock and/or sorbing in the rock is thus considered as negligible compared to the amount in the water phase and the diffusion can be calculated according to:

$$C = C_0 \cdot (\varepsilon + K_d \cdot \rho) \cdot \left(\frac{d_{samp}}{2} \right)^2 \cdot \pi \cdot l \cdot \operatorname{erfc} \left(\frac{x}{2 \cdot \sqrt{D_a \cdot t}} \right) \quad (4.3)$$

where:

- ε - porosity of the rock
- K_d - sorption distribution coefficient (m³/kg)
- ρ - rock density (kg/m³)

For the scoping calculations of the LTDE experiment, the following premises have been assumed:

- The groundwater volume (V) has been set to 1 litre
- The diameter of the stub (d_{tot}) has been set to 0.2 m
- The sampling disc has been set to have a thickness (l) of 0.0005 m and a diameter (d_{samp}) of 0.04 m.
- The diffusion time (t) has been set to 4 years (=1.26·10⁸ s).
- The amounts of tracers used are based on Table 5-2.

- The apparent diffusivities, D_a , used for the different tracers used are given in Table 4.1. For some of the tracers, only a effective diffusivity, D_e , is available. In those cases, the apparent diffusivity has been calculated according to:

$$D_a = \frac{D_e}{(\varepsilon + K_d \cdot \rho)} \quad (4.4)$$

and the porosity in that case have been set to 0.004 and the rock density to 2700 kg/m³.

Table 4-1 Tracers used in the scoping calculations

Tracer	D_e (m ² /s)	K_d (m ³ /kg)	D_a (m ² /s)	Reference
²⁴ Na	6.7·10 ⁻¹⁴	1.4·10 ⁻⁶	8.6·10 ⁻¹²	Byegård et al. 1998
³⁶ Cl	1.2·10 ⁻¹³	0	3.0·10 ⁻¹¹	Ohlsson and Neretnieks 1997
⁹⁰ Sr	4.0·10 ⁻¹⁴	4.7·10 ⁻⁶	2.4·10 ⁻¹²	Byegård et al. 1998
⁹⁹ Tc	-	-	2.2·10 ⁻¹⁴	Ittner et al. 1990
¹³⁴ Cs	-	-	3.0·10 ⁻¹⁴	Byegård et al. 1998
²³⁷ Np	-	-	4.1·10 ⁻¹⁵	Ittner et al. 1988
²⁴¹ Am	-	-	1.0·10 ⁻¹⁵	Ittner et al. 1988

In the scoping calculations, only tracers have been included for which experimental diffusion data are available. The results of the calculations are given in Figure 4.1 (tracers to be measured radiometrically) and in Figure 4.2 (tracers to be measured mass-spectrometrically). Theoretically, it would be possible to observe matrix diffusion several dm:s in to the rock for the non-sorbing tracers and possibly also some mm:s for the most strongly sorbing tracers. However, using the diffusivity data calculated by Ohlsson and Neretnieks (1997) from the sorption data given by Carbol and Engkvist (1997) no penetration whatsoever of the most strongly sorbing tracers (Tc, Cs, Th, U, Np and Am) should be possible to observe.

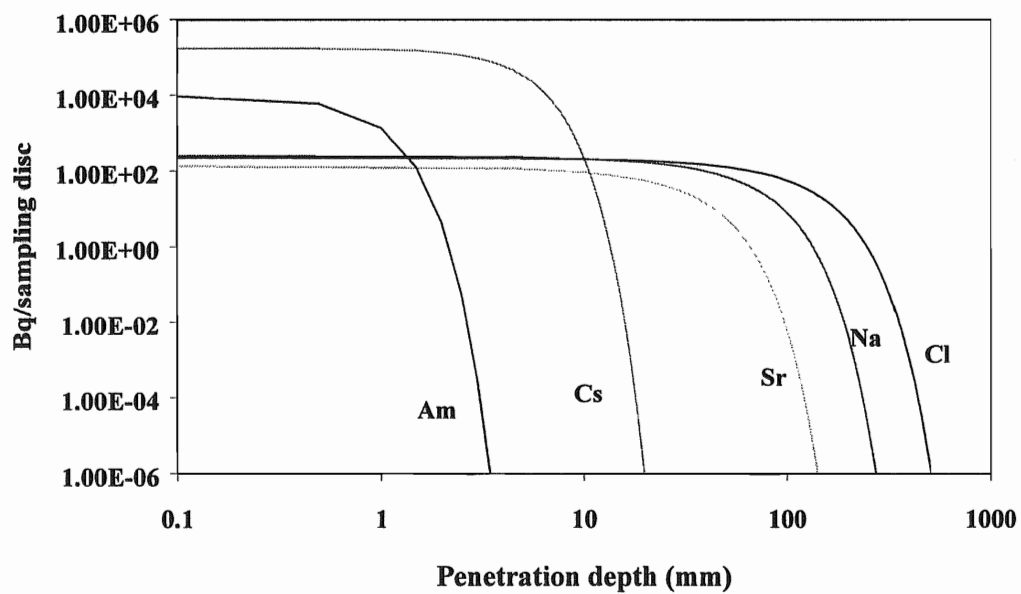


Figure 4-1 Penetration profile for the tracers to be measured radiometrically.

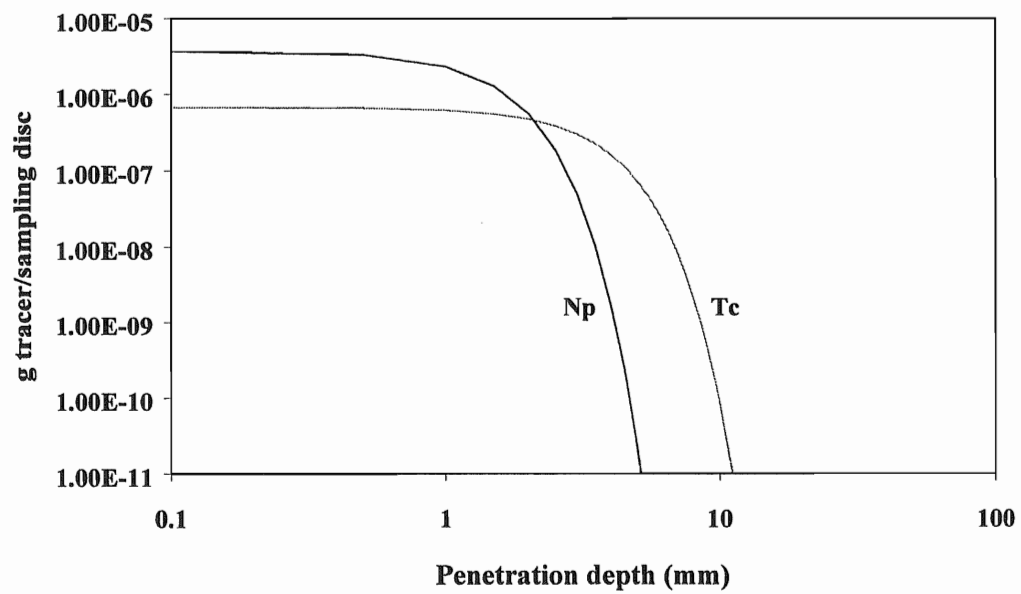


Figure 4-2 Penetration profile for the tracers to be measured mass-spectrometrically.

4.4 Problem areas

The problem areas can broadly be divided into those related to the geological and hydraulic conditions met at the proposed site. The other group of problems are related to the experimental performance once a site has been selected.

4.4.1 Site-related problems

It is foreseen that the selected fracture is located in a relative homogeneous bedrock. It is further foreseen that the fracture will be intercepted with a borehole geometry which will create a near orthogonal fracture surface in relation to the borehole axis. Further, that the fracture is intercepted at a depth ranging between 10 and 20 m. The latter implies that the fracture is located beyond the excavation disturbed zone (relative to the excavated tunnel) and at a distance which is accessible by large diameter drilling

If the above three constraints are full-filled, it is expected that the site will not provide any serious problems to the performance of the experiment.

In the event, a leakage occurs, the procedure outlined in the succeeding section will be put in effect. In the event a tracer is not reclaimed, it will migrate towards the closest major sink. It is expected that a portion of the tracer will sorb on the fracture surfaces along the flow path to the sink. The closest sink which is relevant to the identified experimental locations, cf. Section 5.1.1. is the TBM Assembly Hall.

4.4.2 Experimental problems

For the use of high amounts of radioactive tracers in the experiment, it is important that no leakage from the tracer reservoir (test section) occurs. However, safety precautions have to address how to handle a situation of failure. The following actions can at the moment be foreseen:

- A pre-test should be performed before the injection of radioactive tracer in order to verify that no loss of any water from the isolated section can be measured.
- An investigation should be performed in order to identify possible in-leakage points in the tunnel in the case of an experimental failure. This is preferably done by injecting a non-sorbing tracer in the borehole section without the isolation of the stub present. At this stage, a check could also be done in order control the hydraulic conductivity and pressure responses between the experiment borehole and the reference borehole.
- During the experimental phase of the experiment, an on-line measurement of the tracer concentration will be performed. The possibilities of installation of a system alarming in the case of lowered counting rate should be investigated.
- It is planned to install a radioactivity contamination detector (e.g., a scintillation detector). This detector should be connected to a loop in which the groundwater from the reference section and from the inner guard section will be alternately connected. The water from these volumes will then be circulated and measured for its content of radioactive tracers, in order to detect and measure any leak from the injection borehole. An alarm function will be connected to this detector.
- In the case of a severe leak due to failure in the experimental equipment, this can probably be observed by a drop of hydraulic pressure in the different sections. An alarm should be obtained in the case a drop of the pressure. Small leaks are difficult to detect since the obtained volume decrease in the experimental borehole can be masked by the transmissivity of the rock and/or the elasticity of the equipment. The best way of obtaining indications of small leaks is thus to perform radioactivity contamination measurements.

- During the whole experimental programme, water samples from the adjacent borehole sections should be collected and be measured γ -spectrometrically. This will give an indication of any small leak from the injection borehole.

The isolation of the volume between the stub and the borehole wall is essential, in order for the tracer spiked water not to penetrate that volume. If perfect isolation is not obtained, tracer-spiked water may penetrate the stub from the outside and difficulties in interpreting the diffusion may arise. This is preferably resolved by efforts for obtaining a perfect isolation. An other possible cause for enhanced diffusion along the walls of the stub is sample disturbance (damage and stress relief). For the two mentioned situations, exclusion of samples from the side of the stub in the penetration studies can also ensure that the concentration profile is not disturbed by diffusion from the water in contact with the side of the stub.

The choice of sealing material around the stub has to be done primarily for good isolation purposes, see above. However, for the use of strongly sorbing tracers, it is necessary that sorption on the material used in the borehole instrumentation has to be considered. It has been indicated that in the choice of sealing material around the water reservoir one is probably restricted to use some rubber material. If the sealing can not be done with a material with known low sorptivity (e.g., PEEK), the most strongly sorbing tracers will probably have to be excluded from that experimental concept.

Laboratory batch experiments in order to study the interaction of the tracers on the materials used for the instrumentation are planned to be performed in advance of the actual *in situ* diffusion experiments.

Some of the of tracers that are proposed to be used in this experiment hydrolyses strongly at the neutral groundwater pH ~ 8 (i.e., $^{229}\text{Th}^{4+}$, $^{233}\text{UO}_2^{2+}$ and $^{241}\text{Am}^{3+}$). Since hydrolysis favour sorption, it is likely that injection of tracers in a cocktail having that high pH will cause sorption of these tracers already at the inlet tubes. A way to overcome this problem would be to inject the tracers in a small volume of increased acidity. During the injection of this acidified tracer cocktail, efforts could be made to minimise the dilution during the transfer of the tracer to the isolated borehole volume. A plume of tracers would than be transported to the tracer reservoir at the fracture and would then be diluted at that point. It is probably necessary to afterwards slowly add sodium hydroxide to compensate for the acid injected during the tracer injection. One will have to make sure that the natural concentration of bicarbonate in the system can buffer the acid and base injected so that no severe pH changes occur during the injection.

5. Scope

5.1 Main project tasks

5.1.1 Site selection

A site selection project (SELECT-2) has been carried out to identify suitable fractures for LTDE (Winberg et al., in prep) and TRUE-2. Four boreholes have been drilled, KA2377A, KA2865A01, KXTT5 (at the TRUE-1 site) and KA3065A02. These boreholes have been subject to characterisation using borehole TV (BIPS), core logging (BOREMAP), borehole radar (not in KXTT5), and continuous flow logging (POSIVA difference flow meter) (not in KA2377A).

The results have shown that suitable candidates for the LTDE experiment are located in KA2865A01 and in KA3065A02, at depths ranging from 10 to 20 m. The part of the laboratory hosting the two boreholes is well known from the REX and TRUE-1 experiment, respectively.

Presently, the data from the two sites are evaluated to select the most suitable candidate fracture for LTDE.

5.1.2 Drilling

The experimental concept presumes that an existing pilot borehole can be used to identify the target fracture. The latter corresponds to one of the SELECT-2 triple tube 76 mm pilot boreholes discussed in the previous section.

Parallel and approximately <0.5 m away from the pilot borehole a large diameter borehole is drilled using conventional double core barrel technique.

The large diameter borehole which will host the experiment will be drilled in a telescoped fashion, stepping from the initial diameter (300 mm) to a diameter of the experimental chamber (196.5 mm) about 1-2 meters away from the fracture (diameter of the stub is 177 mm). The resulting slot between the borehole wall of the experimental chamber and the stub will be $(196.5-177)/2$ mm, or 9.75 mm. The nominal diameters of the borehole segments and the associated cores are presented in Table 5-1.

During the drilling, the core will be broken using a hydraulic jack. When approaching the target fracture plane the drilling will be carried out very carefully, drilling through and pass the fracture plane. It is expected that

the outer surface of the inner part of the borehole as well as on the surface of the stub will be smooth.

It is further envisaged that the remaining stub will be approximately 50 mm long. The latter facilitate optimal seal against the stub and the outer borehole wall. In addition the distance over which enhanced diffusion along the interface between the stub and the seal is minimal in relation to the scoped advancement of the diffusion front from the fracture, which is on the order of decimetres, cf. Section 4.3.1.

Table 5-1 Overview of diameters used when drilling the LTDE large diameter experimental borehole.

Outer segment		Inner segment	
OD	Core	OD	Core
300 mm	280 mm	196.5 mm	177 mm

5.1.3 Characterisation in experimental borehole

The large diameter corehole will be logged using the RAAX BIPS borehole imaging system. These measurements will form the platform against which all other measurements and loggings are referenced.

In addition a series of geophysical logging techniques will be employed to investigate the homogeneity in porosity and lithological will be employed. These techniques include;

- Normal resistivity, (volume penetrative)

When used for normal/lateral resistivity measurement, this probe measures the apparent resistivity in the bedrock. The normal resistivity log will indicate zones having anomalous resistivity such as fractures containing water and conducting minerals or the presence of mineralisation. Here, it is possible to make quantitative estimates of rock quality in general and also the width and resistivity of fractures. In many types of rock it is possible to find a direct relation between resistivity and porosity. For normal resistivity, the apparent resistivity of bedrock is measured with one current electrode and one potential electrode in the borehole. The other potential and current electrodes are situated on the surface at some distance from the borehole. With available resistivity probes a number of different layouts are possible, including Long (1.6 m) and Short normal (0.4 m). One restriction which may entail problems in this particular application is that the measurements requires the borehole to be filled with water.

Measurements of the resistivity of the boreholes will provide data to evaluate the matrix diffusivity by electrical conductivity methods, as been described by Ohlsson and Neretnieks (1998).

- Gamma-Gamma (density)

This probe is used to determine the density of rock by exposing it to a radioactive source and then measuring the reflected radiation. Changes in lithology and the presence of large fracture zones are indicated.

- Gamma (mineral assembly)

The gamma probe measures the natural gamma radiation in the borehole. Detectors, 1" diameter x 1.5" long NaI crystals, are used. Natural gamma radiation is used to detect boundaries between different types of rocks (for example). Total natural gamma radiation from potassium, uranium and

thorium is measured. Variations in the concentrations of these elements normally correspond to lithological changes in the rock.

The exact suite of logs has not been decided at this stage.

5.1.4 Geological characterisation of the drill core

The large diameter drill core will be logged using the BOREMAP system.. The results of this base logging will be used for a first screening of the boreholes with regards to suitability. In addition selected samples from the core will be subjected to mineralogical and geochemical analyses.

Parameters of importance for the Long Term Diffusion Experiment include rock textures, porosity and mineral composition. To meet these requirements a detailed description of the rock is needed. This include mineral composition, description of grain sizes and textures e.g. foliation, presence of mylonites and microfractures etc. Possible alteration of the rock is also important to determine, as it will affect the mineral composition (e.g. degradation of biotite to chlorite) and also the porosity.

Chemical whole rock analyses (ICP-MS and INA) and microscopy of the thin sections in transmissive light is the best way to complement the basic core logging. SEM/EDS studies of specific minerals or structures may be required. It is important that the samples selected for chemical and mineralogical identification correspond, as close as possible, to the samples analysed for petrophysical measurements (porosity, density etc), cf. Section 5.1.5. The number of analyses and thin sections that should preferably be studied from each borehole is dependent on the homogeneity of the rock and the degree of alteration, and should be adjusted as information from BIPS, basic core logging and the interpretations of the structural geology emerge.

Suggested mineralogical and geochemical analyses include :

- Whole rock analyses using ICP-MC/INA (Since Cs will be used in the experiment it is important to know the content of stable Cs in the rock, and this is best analysed using neutron activation (INA)).
- Microscopy of thin sections is important for determination of mineralogical composition and textures.

- Measurements of the isotopes in the ^{238}U -series. This is preferably done in the part of the drill-core that includes the opposite part of the fracture. By quantifying the uranium disequilibrium as a function of the distance of the fracture face, a natural analogue estimation of the matrix diffusivity may be obtained which should be interesting to compare with values obtained for the diffusivity of synthetic tracers.

5.1.5 Petrophysical characterisation of core samples

Measurements on selected samples will be made of

- porosity
- density
- permeability

Permeability and porosity will tentatively be assessed using He-gas as a medium (*pending results of ongoing experimental work on Åspö generic material*). The He porosity determinations will be compared with porosities obtained using the water absorption method.

5.1.6 Assessment of sample disturbance in core samples

In order to characterise sample disturbance two techniques will be considered, the first being a direct method involving thin section analyses in the microscope (done in conjunction with mineralogical analysis, cf. Section 5.1.4). The second technique, is an indirect geophysical measurements (micro seismic refraction) by which the depth of disturbance possibly can be assessed. It is expected that the resolution of the technique is in the order 1-3 mm.

Quantification of the pore structure and porosity will be made using impregnation of samples with ^{14}C -labelled methylmethacrylate (^{14}C -MMA method) (Hellmuth, et al, 1994) and Raisilainen et al, 1996). The primary objective of this method is to identify and visualise transport pathways. In addition total porosity can be quantified. The secondary objective, and new application of the method, is to assess sample disturbance along the perimeter of the experimental core.

5.1.7 Cleaning of borehole

The drilling of the boreholes will produce drilling debris which is not flushed out during the drilling operation. Small debris particles will

eventually sediment on the borehole walls and become a potential site for sorption of the injected tracers. The boreholes will therefore be cleaned with special cleaning equipment previously used in the REX Experiment. The method uses the section water in a closed circulation loop where the water is filtered. The active tool is a nozzle in the form of a brush which is applied to the walls of the borehole. Thus, no additional “foreign” water is added.

5.1.8 Instrumentation of boreholes

The instrumentation will be done in a large diameter borehole (approximately 200 mm, telescoped from a larger diameter, cf. Figure 5-1). The length of the borehole to the fracture plane is about 10 to 20 m. In the bottom of the hole a stub including the target fracture plane remains. This stub will be packed off and isolated so that only the fracture surface is in contact with the groundwater. It is important to get a good sealing along the cylinder area of the stub to get a axial diffusion and to minimise the water volume in contact with the fracture plane. The material of the sealing in contact with the water in this section should have no or only a negligible effect on the test. There will be three different PEEK-tubings coming in to the test section, one for pressure measurements and two for tracer circulation.

Outside of this section there will be a mechanically operated packer which will withstand a differential pressure of about 4.5 MPa. This packer will reduce the maximal differential pressure needed for the sealing around the stub. Also in the section inside this packer, three nylon tubes will emerge, one for pressure measurements and two for water circulation.

Hydraulically operated packers will be installed outside the mechanical packer, to reduce the pressure gradient along the hole. One hydraulic tube will be connected to each section around these packers, which could be used for pressure measurements.

The mechanical packer constitutes a fixed seal, independent of the hydraulic packers, which will facilitate a low hydraulic gradient across the stud and the test section.

The packers will be inflated with separate lines to reduce the effects of a failure in one packer. They will be operated by water pressurised with nitrogen. In order to avoid pressure gradients induced by the packers, the pressure vessel used for inflation needs to be as small as possible. Pressure changes induced by changes in the ambient temperature will be reduced by isolation of the pressure vessel and the inflation tubing.

At the top of the borehole there will be a unit including an outer seal and anchor device, packer expansion gear, piston pipe locking device which permit:

- Sealing function around the stub
- Operation of the mechanical packer
- Sealing of borehole
- Locking of the equipment

The matrix section will be connected to pressure transducers at the surface through ID 2 mm PEEK tubes. The matrix section will also be connected to the surface through two additional PEEK tubes used for circulating, injecting, and sampling of the section volume. The basic idea is to have a slowly circulating system where the tracer solution may be sampled and controlled over the duration of the experimental period, see Figure 5-1.

The surface part of the system includes a circulation loop with a pulsation free circulation pump (gear pump), a pressure regulator and some water chemistry sensors, tentatively including electrical conductivity, redox potential (Eh) and pH. The pressure regulator will in principle be a plunger that is moved back and forth depending on the pressure changes in the system caused by sampling, injection or by external circumstances such as temperature variations in the tunnel. The plunger will be regulated through a control unit where the input pressure is the pressure in the reference fracture. In addition, sampling and injection of tracer should be possible to perform. This is further described in Sections 5.1.9 and 5.1.10.

The pressure transducers are connected to a data logger for continuous storage of data. On-line monitoring of pressures will also be possible by connecting the system to the Äspö HRL Hydro Monitoring System (HMS).

The injection of tracer solution may be done in two different ways. The simplest way would be to inject a small volume of concentrated tracer solution either by a syringe needle through a septum (BAT system) in direct contact with the circulating loop. The other system is a cylinder with a double acting piston. When injecting a tracer volume, the same volume will be evacuated from the circulation volume. The system that will be used will depend on the final choice of tracers and other practical considerations.

Laboratory experiments studying sorption of the tracers proposed on the different materials (PEEK and polyurethane) will be performed.

As outlined in Section 4.4.2, pre-tests checking the performance of the equipment, and mapping possible outlets in the tunnel for tracer in the event of a failure will be performed.

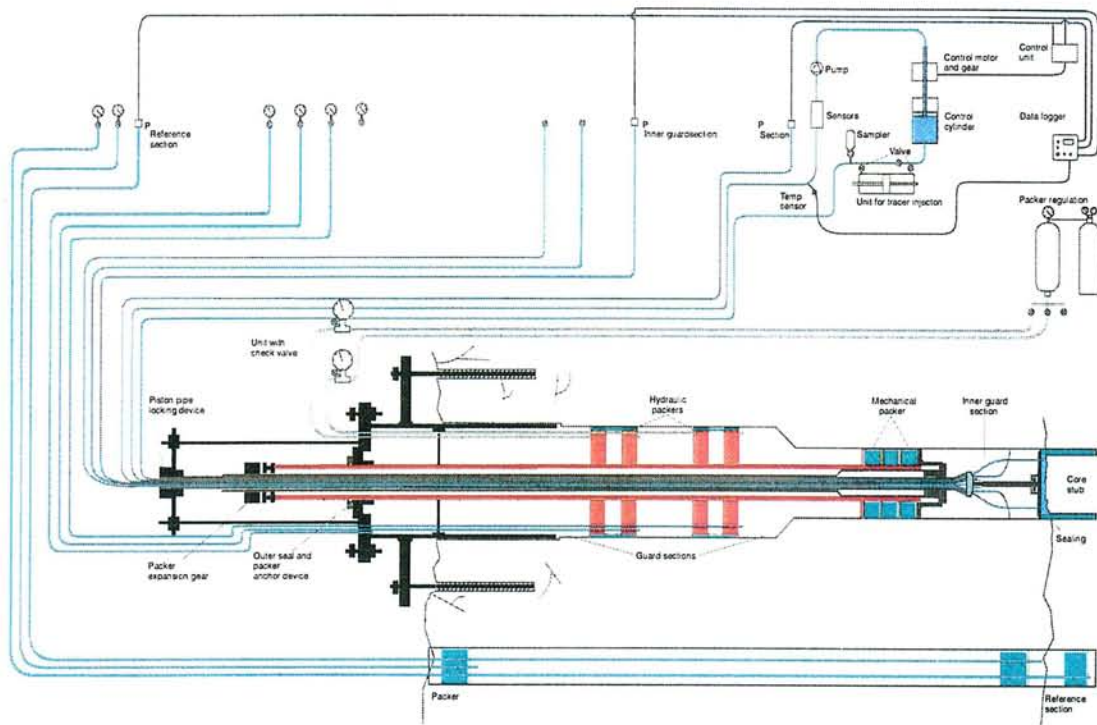


Figure 5-1 LDTE - Equipment for in-situ diffusion experiment.

5.1.9 Supporting laboratory diffusion experiment

One important aim of the experiment is to compare the validity of diffusion parameters obtained in laboratory with *in situ* data. Since the *in situ* data will be extracted only from one specific fracture surface, it is essential for comparative purposes to do laboratory analogue experiment with the same rock material. In this particular case it should be advantageous to use the opposite fracture surface (the one obtained as the end of the drill core) for laboratory experiment. Porosity and diffusivity of tritiated water should be determined according to the procedures described by Byegård et al 1998.

It is also proposed that parts of the core is exposed to other laboratory techniques that has been used for rock matrix diffusion studies, such as electrical conductivity measurements (Ohlsson and Neretnieks, 1998), gas diffusion (Väättäinen et al. 1993) and ¹⁴C-polymetacrylate impregnation (Hellmuth et al, 1994). A comparison of these laboratory techniques is presently being performed (Johansson and Ohlsson, 1999) using Äspö diorite and Fine grained granite which have been used in laboratory experiments. The Long-Term Diffusion Experiment therefore gives a unique opportunity for comparison between *in situ* techniques and laboratory studies used for matrix diffusion studies. Possibly the ¹⁴C-impregnation technique is only possible to apply on material which has no sorbed radionuclides. In this case the requested information can be obtained from the “mate of the stub”, cf. Section 5.1.1.

5.1.10 Selection of tracers

The tracers that are suggested to be used in the LTDE are given in Table 5-2.

A good non-sorbing to be used in the experiment would be tritiated water (HTO). However, since the analysis of the penetration of the tracers in to the rock involves cutting and sawing of thin slices of the rock, it is foreseen that the volatility of HTO will cause severe losses of that tracer. It is therefore proposed that instead use the presumed non-sorbing anion ³⁶Cl⁻ as a tracer to determine the maximum penetration depth of the water in to the rock. After chemical separation, the isotope ³⁶Cl⁻ is measured using liquid scintillation. However, by using accelerator mass spectrometry measurements in the order of 10⁻⁶ Bq/l of ³⁶Cl⁻ have been reported (Cornett et al. 1997) and it is thus possible to obtain an excellent dynamic range of that tracer. The accelerator mass spectrometry is, however, an expensive technique and these measurements can probably only be applied for a limited numbers of samples.

The weakly sorbing tracer $^{22}\text{Na}^+$ has been used in the TRUE-1 tracer programme. The $^{22}\text{Na}^+$ tracer has the advantage that the decay involves γ -emission. It is thus possible to measure the concentration of that tracer in intact slices of the sampled core, i.e., no leaching or dissolution is necessary. Accordingly, since results (Johansson et al. 1997) have shown that retardation of $^{22}\text{Na}^+$ compared to tritiated water is minor, the tracer is suitable to use in coarse scan of the sampled drill core.

The experimental concept offers the possibility of obtaining a natural fracture and low hydraulic conductive borehole section, respectively, with long time contact of a tracer spiked groundwater to the geological environment. It is therefore recommended to include radioisotopes that are of significant interest for a deep repository for nuclear waste in this experiment. The redox sensitive elements Tc, Np and U are of particular interest since, in the assumed prevailing reducing conditions in the groundwater, they are all expected to be reduced to their respective tetravalent form and thus to be sorbed or precipitated. These elements are thus proposed to be injected in their soluble oxidised forms, i.e., Tc(VII)O_4^- , Np(V)O_2^+ and U(VI)O_2^{2+} , respectively. The experimental concept will offer possibility of reduction/sorption kinetics *in situ*, and matrix diffusion properties of the sorbed species.

Furthermore, it is proposed to study the behaviour of some other elements of direct repository interest, e.g., Cs(I), Sr(II), Am(III) and Th(IV). These elements have fixed oxidation states and the behaviour of Am(III) and Th(IV) could to some extent be used to predict the behaviour of other elements of repository interest having the same oxidation state. The proposed experiment will offer the possibility both of studying the sorption kinetics of tracers and to see if there is any complexation *in situ* that increases the matrix diffusion of the presumably strong sorbing tracers.

The choice of sorbing tracers in the experiment is dependent on the outcome of the choice of sealing material to be used in the experimental concept. If it is found that the material used interacts strongly with any of the tracers, the tracers will probably have to be excluded. Batch sorption experiment will be performed on different material in order to investigate this issue.

In order to be able to, after the excavation process, study the penetration of these tracers in to the rock matrix, the elements are proposed to be used in their long-lived isotopic form, i.e., ^{134}Cs , ^{90}Sr , ^{99}Tc , ^{229}Th , ^{233}U , ^{237}Np and ^{241}Am . The amounts of tracer that should be injected are limited by the following restrictions:

- Not more than 10 ALI (annual limit of intake) should be used for any radioisotope (limitation for $^{90}\text{Sr}^{2+}$, $^{134}\text{Cs}^+$, $^{229}\text{Th(IV)}$ and $^{241}\text{Am(III)}$)
- A maximum concentration of $1 \cdot 10^{-6}$ M should be used of any tracer, unless the natural concentration of that element is higher (limitation for $^{99}\text{TcO}_4^-$, $^{233}\text{UO}_2^{2+}$ and $^{237}\text{NpO}_2^+$)
- Economical constraints limit the use of some tracer (limitation for $^{22}\text{Na}^+$, and $^{36}\text{Cl}^-$)

Table 5-2 Tracers proposed to be used in the experiment. In the calculation of the maximum amount to be used, it has been assumed that a total volume of 1 litre is used in the circulating water system.

Tracer	Maximum amount	Limitation	Measuring method	Half-life	ALI	Decay	Supplied by (e.g.)
$^{22}\text{Na}^+$	$7 \cdot 10^7$ Bq	Economical	γ -spec	2.6 y	$2 \cdot 10^7$ Bq	ϵ, γ	NEN
$^{36}\text{Cl}^-$	$4 \cdot 10^7$ Bq	Economical	Liq. Sc., AMS	$3 \cdot 10^5$ y	$6 \cdot 10^7$ Bq	$\beta^-, (\beta^+)$	NEN
$^{90}\text{Sr}^{2+}$	$1 \cdot 10^7$ Bq	10 ALI	Liq. Sc.,	29 y	$1 \cdot 10^6$ Bq	β^-	NC-CTH
$^{99}\text{TcO}_4^-$	$6 \cdot 10^4$ Bq	$\leq 1 \cdot 10^{-6}$ M	ICP-MS	$2 \cdot 10^5$ y	$1 \cdot 10^8$ Bq	β^-	NC-CTH
$^{134}\text{Cs}^+$	$3 \cdot 10^7$ Bq	10 ALI	γ -spec	2.1 y	$3 \cdot 10^6$ Bq	β^-, γ	Risö
$^{229}\text{Th(IV)}$	$2 \cdot 10^5$ Bq	10 ALI	γ -spec	7340 y	$2 \cdot 10^4$ Bq	α, γ	ORNL
$^{233}\text{UO}_2^{2+}$	$8 \cdot 10^4$ Bq/l	$\leq 1 \cdot 10^{-6}$ M	ICP-MS	$2.3 \cdot 10^7$ y	$5 \cdot 10^5$ Bq	α	NC-CTH
$^{237}\text{NpO}_2^+$	$6 \cdot 10^3$ Bq	$\leq 1 \cdot 10^{-6}$ M	ICP-MS,	$2.1 \cdot 10^6$ y	$2 \cdot 10^4$ Bq	α	NC-CTH
$^{241}\text{Am(III)}$	$3 \cdot 10^5$ Bq	10 ALI	γ -spec	430 y	$3 \cdot 10^4$ Bq	α, γ	NC-CTH

Abbreviations used:

ALI	Annual Limit of Intake (oral)
NEN	NEN Life Science products, commercial supplier of radioisotopes
ORNL	Oak Ridge National Laboratory, U.S.A.
NC-CTH	Chalmers University of Technology, Department of Nuclear Chemistry

5.1.11 Sampling of the injection borehole

The sampling methodology in the injection borehole employed during the injection phase will be similar for the two presented experimental concepts. The principle will be to extract a small volume (10 ml) from the circulating loop at, tentatively, 15 occasions during the injection phase. Thus the total volume removed from the system will be less than 150 ml and the pressure regulation plunger, cf. Section 5.1.8, handles the volume compensation. The sampling will be more intense in the beginning and gradually decreasing to one sample per year.

The sampling is made either through a HPLC sampling valve, where a fixed volume may be removed by manual switching of the valve, or by using a modified BAT system (syringe needle through a semi-permeable septum).

5.1.11.1 Analysis of the tracers in the injection water

Since the water in the injection section will be continuously circulated, it is proposed to use an on-line HPGe detector to measure the concentration of the γ -emitting tracers, i.e., $^{22}\text{Na}^+$ and $^{134}\text{Cs}^+$. The other γ -emitting tracers $^{229}\text{Th(IV)}$ and $^{241}\text{Am(III)}$ are used in much lower concentrations and they are, furthermore, expected to be lost in the water phase due to sorption. These tracers are therefore not possible to measure on-line and sampling and separation procedure will have to be applied for the measurement for these tracers and the tracers lacking γ -emission.

A proposed separation and analysis scheme is given in Figure 5.2. The methods proposed have been outlined by Ramebäck and Skålberg (1999) for the determination of diffusivity of long-lived radionuclides in bentonite backfill. Some modifications have been made for the suitability to the specific radionuclides used in this experiment, i.e.;

- Separation of $^{36}\text{Cl}^-$ by AgCl precipitation (Cornett et al 1996)
- Separation of Np and Th in their tetravalent form by HTTA-extraction (Starý 1964)
- Strip of UO_2^{2-} from HDEHP organic phase by addition of EDTA to the aqueous phase

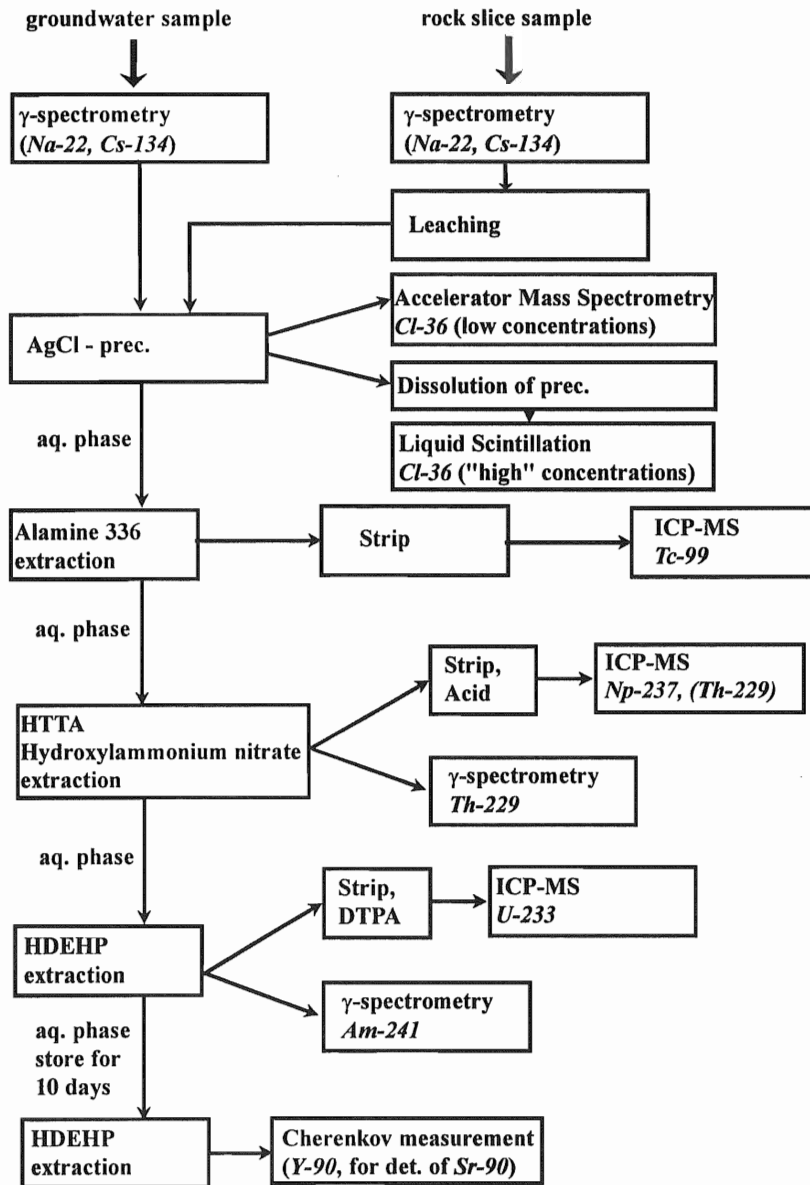


Figure 5-2 Analysis procedure proposed for the Long-Term Diffusion Experiment

5.1.12 Overcoring drilling

Once the experimental work is completed, the experimental chamber including the stub will be overcored using the diameter of the outer segment. The overcoring will be performed to about 0.5 metres beyond the fracture plane.

5.1.13 Analyses of tracer content in the drill core

After the overcoring has been performed, studies of the penetration depth of the used isotopes will be performed. Methods of measurements of the penetration profile of radioactive tracers after diffusion laboratory experiments have been presented by Ittner et al. (1988 and 1990), Albinsson et al. (1994) and by Johansson et al. (1998). In all these experiments only one isotope together with one given rock slab specimen were used and selective measurements and separations were thus not necessary. However, in the proposed experiment a mixture of radioisotopes will be present in the rock material. The same methods as presented for the separation and measurement of the radioactive species in the water phase (Figure 5-2) are proposed to be used in also for the rock samples.

5.1.14 Microbial investigation

The REX experiment has indicated that microbes play a role in changing the chemical environment over time. In order to assess the role of microbes, sampling for microbes in groundwater will be attempted as part of the sampling for analyses of tracer content in groundwater.

5.1.15 Post-excavation assessment of sample disturbance

It is assumed that the damage exerted by the drilling can provide enhanced diffusion along the periphery of the stub. Despite the fact that the stub is short in relation to the scoped diffusion depth, the experimental results may call for numerical analysis which takes into account the *in-situ* test geometry in relation to the existing *in-situ* stress situation. It is expected that input data to such an analysis can be obtained from the performed ZEDEX experiment (Emsley et al, 1997) and from the performed petrophysical and geophysical measurements, cf. Section 5.1.5 and 5.1.6.

Potentially, a numerical FEM model developed for analysis of the mechanical situation can also be employed to model the diffusion process, and possibly other processes.

Assuming that a constitutive relationship exists between seismic velocity (through elastic modulus and porosity) and diffusivity, also forward modelling of the diffusion process may be possible.

6. Time schedule

Site selection for the experiment will be carried out in mid September 1999. Drilling and characterisation will be performed starting November 1999, followed by characterisation. Instrumentation of the borehole will be performed in April 2000. The circulation of tracer solution is planned to start in May 2000.

Evaluation of analyses results from collected samples and supporting modelling will be carried out on a yearly basis for the duration of the experiment (4 years). The overcoring is planned to be carried out in January 2004. The final report is due in October 2004.

7. Project organisation and resource requirements

7.1 Introduction

The experiment constitutes a component of the Tracer Retention Understanding Experiment (TRUE) being conducted at The Äspö HRL.

7.2 Organisation

The project is organised as collaborative project between mainly Swedish universities and consultant companies. The project manager of the LTDE is Anders Winberg. Under the project manager a number of professionals support the project manager with different responsibilities and reports to the project manager;

Johan Byegård	Theoretical and laboratory aspects of diffusion and sorption processes. Scoping calculations. Tracer injection and radiation safety. Water and hard rock analyses for tracer content. Supporting laboratory experiments. Modelling of experimental results.
Peter Andersson	Instrument issues. Hydraulic testing.
Kent Hansson	Instrument design. Manufacturing of downhole and surface equipment. Testing of equipment before delivery.
Eva-Lena Tullborg	Mineralogy, geochemistry, petrophysics, uranium series measurements.
Torbjörn Hugo-Persson	Large diameter drilling technology. Over-coring technology.

7.3 Data management

The experimental results should be inputted in SICADA on a regular basis. The structure of the data tables will be agreed upon between the project group and the Äspö HRL database administrator.

7.4 Reporting

It is foreseen that the following reports will be produced during the course of the experiment.

- Drilling report (ITD) Feb 2000
- Site characterisation report (IPR) Mar 2000
- Instrumentation report (ITD) Dec 2000
- Mineralogy and geochemistry (ITD) Feb 2001
- Yearly progress reports (IPR) Dec 200X
- Report on supporting laboratory experiment (IPR) Dec 2003
- Report on overcoring drilling (ITD) Oct 2004
- Report on results of field experiment (ITD) Oct 2004
- Final Report of LTDE project (TR) Oct 2004

8. References

Albinsson, Y., Andersson, K., Börjesson, S., Allard, B., 1994 Diffusion of radionuclides in concrete and concrete/bentonite systems, Fourth international conference on the chemistry and migration behaviour of the actinides and fission products, R. Oldenbourg Verlag, p. 485-491 München 1994.

Allard, B., Ittner, T., Torstenfeldt, B., 1985 Migration of trace elements into water-exposed natural fissure surfaces of granitic rock, *Chem. Geol.* 49, 31-42

Birgersson, L., Neretnieks, I. 1990 Diffusion in the matrix of granitic rock: Field test in the Stripa mine, *Wat. Resour. Res.* 26(11) 2833-2842

Birgersson, L., Widén, H., Ågren, T., 1994 Matrix diffusion experiment, Initial planning and review, SKB Äspö Hard Rock Laboratory Technical Document 25-94-021, Stockholm 1994

Byegård, J., Johansson, H., Skålberg, M., Tullborg, E.L. 1998 The interaction of sorbing and non-sorbing tracers with different Äspö rock types, Sorption and diffusion experiments in the laboratory scale, SKB Technical Report TR-98-18, Stockholm 1998

Carbol, P., Engkvist, I., 1997 Compilation of radionuclide sorption coefficients for performance assessment, SKB Repot R-97-13, Stockholm 1997.

Cornett, R.J., Andrews, H.R., Chant, L.A., Davies W.G., Greiner, B.F., Imahori, Y., Koslowsky, V.T., Kotzer, T., Milton, J.C.D., Milton, G.M., 1997 Is ^{36}Cl from weapons' test fallout still cycling in the atmosphere? *Nucl. Instr. and Meth. B* 378-381

Crank, J., 1975, *The mathematics of diffusion*, 2nd edition, Oxford Univ. press, London.

Emsley, S., Olsson, O., Stenberg, L., Alheid, H.J., Falls, S. 1997 ZEDEX - A study of damage and disturbance from tunnel excavation by blasting and tunnel boring, SKB Technical Report TR 97-30, Stockholm 1997

Haggerty R., Gorelick, S.M., 1995 Multiple rate mass transfer processes for modelling diffusion and surface reactions in media with pore-scale heterogeneity, *Water Resour. Res.*, 31 2383-2400

Haggerty R., Gorelick, S.M., 1998 Modelling mass transfer processes in soil columns with pore-scale heterogeneity, *Soil Sci Soc J.*, 62(1) 62-74

Haggerty R. 1999, Application of the multirate diffusion approach in tracer test studies at Äspö HRL, to be published.

Hakala, M. 1999 Numerical study on core damage and interpretation of in situ state of stress. . POSIVA Technical Report POSIVA 99-25.

Hartikainen, K., Pietarila, H., Rasilainen, K., Nordman, H., Ruskeeniemi, T, Hölttä, P., Siitari-Kauppi, M, and Timonen, J. 1995 Characterization of the altered zone around a fracture in Palmottu Natural Analogue. *Mat. Rs. Soc. Symp. Proc.*, vol. 412, pp. 839-846.

Hartikainen, K., Hartikainen, J. And Timonen, J. 1997 Through-diffusion, permeability, channel-flow and in situ results for porosity and migration properties of rock samples by He-gas methods. POSIVA Technical Report POSIVA 97-13.

Hellmuth, K.-H., Lukkarinen, S., Siitari-Kauppi, M., 1994, Rock matrix studies with carbon-14-polymethacrylate (PMMA); method development and applications, *Isotopenpraxis Environ. Health Stud.* 30, 47-60

Ichida, T. et al. 1995 Observation of core discing and in situ stress measurements : Stress criteria causing core discing. *Rock Mech. And Rock Engineering*, pp.167-183.

Ittner, T., Torstenfeldt, B., Allard, B., 1988 Diffusion of neptunium, plutonium and americium in granitic rock, *Radiochim. Acta* 44/45, 171-177

Ittner, T., Torstenfeldt, B., Allard, B., 1990 Diffusion of strontium, technetium, iodine and cesium in granitic rock, *Radiochim. Acta* 49, 101-106

Johansson, H., Byegård, J., Skarnemark, G., Skålberg, M., 1997 Matrix diffusion of some alkali- and alkaline earth-metals in granitic rock, *Mat. Res. Soc. Symp. Proc.*, Vol. 465, 871-878

Johansson, H., Siitari-Kauppi, M., Skålberg, M., Tullborg, E.L., 1998 Diffusion pathways in crystalline rock - Examples from Äspö-diorite and fine-grained granite, *J. Cont. Hydrol.* 35, 41-53.

Li, Shun-Lin, Personal Communication, October 1999.

Li, Y. 1997 Drilling-induced core damage and its relationship to crustal in situ state of stress and rock properties. Ph D Thesis, U. of Alberta, Edmonton, Alberta, Canada.

McKenna, S.A., Meigs, L.C., Altman, S.J., Haggerty, R., 1998 Analysis of tracer tests with multirate diffusion models: Recent results and future directions within the WIPP project, In SKB TR-98-10, Stockholm 1998

McKenna, S.A., 1999 Solute transport modelling of the Äspö STT-1b tracer tests with multiple rates of mass transfer, Contribution to the Äspö Task Force, to be published

Neretnieks, I., 1980 Diffusion in the rock matrix - An important factor in radionuclide retardation? *J. Geophys. Res.*, 85 4379-4397

Novakowski, K.S., Van der Kamp, G., 1996 The radial diffusion method 2. A Semianalytical model for the determination of effective diffusion coefficients, porosity and adsorption, *Wat. Resour. Res.* 32(6) 1823-1830

Ohlsson, Y., Johansson, H., 1999 Comparisons of different laboratory experimental techniques applied for the determination of rock matrix diffusion - application on Äspö Hard Rock Laboratory rock material, to be published.

Ohlsson, Y., Neretnieks, I., 1995 Literature survey of matrix diffusion theory and of experiments and data including natural analogues, SKB Technical Report TR-95-12, Stockholm 1995

Ohlsson, Y., Neretnieks, I., 1997 Diffusion data in granite, recommended values, SKB Technical Report TR-97-20, Stockholm 1995

Ohlsson, Y., Neretnieks, I., 1998, Matrix diffusion studies by electrical conductivity methods. Comparison between laboratory and in-situ experiments, In SKB TR-98-10, Stockholm 1998
1997

Puigdomenech, I., Banwart, S.A., Bateman, K., Griffault, L., Gustafsson, E., Hama, K., Kotelnikova, S., Lartigue, J.E., Michaud, V., Milodowski, A.E., Morosini, M., Pedersen, K., Rivas Perez, J., Trotignon, L., Tullborg, E.L., West, J.M., Yoshida, H., 1998, Redox

experiment in the detailed scale - First Project Status Report, ÄHRL
Technical Report

**Rasilainen, K., Hellmuth, KH., Kievekäs, L., melamed, A.,
Ruskeenieniemi, T., Siitari-Kauppi, M., Timonen, J., Valkiaien, M. 1996**
An interlaboratory comparison of methods for measuring matrix porosity.
VTT Energy, 19996, VTTRN 1776.

Ramebäck, H., Skålberg, M., 1999 A separation procedure for the
analysis of ⁹⁰Sr, ¹⁵⁴Eu, and the actinides ²³⁷Np, ²³⁹Pu, ²⁴¹Am and ²⁴⁴Cm, J.
Radioanal. Nucl. Chem 240(2) 661-663

Skagius, K., Neretnieks, I., 1986 , Diffusivity measurements and
electrical resistivity measurements in rock samples under mechanical
stress, Water Resour. Res 22(4) 570-580

Skagius, K., 1986, Diffusion of dissolved species in the matrix of some
Swedish crystalline rocks, Thesis, Royal Institute of Technology,
Stockholm 1986.

Smellie, J.,A.T., MacKenzie, A.B., Scott, R.D., 1986 An analogue
validation study of natural radionuclide migration in crystalline rocks using
uranium-series disequilibrium studies, Chemical Geology 55 233-254.

Smellie, J., Karlsson, F., 1999 The use of natural analogues to assess
radionuclide transport, manuscript accepted for publication in Engineering
Geology'

Stary J., 1964, The solvent extraction of metal chelates, Pergamon Press,
Oxford

Van der Kamp, G., Van Stempvoort, D.R., Wassenaar, L.I., 1996 The
radial diffusion method 1. Using intact cores to determine isotopic
composition, chemistry, and effective porosities for groundwater in
aquifers, Wat. Resour. Res. 32(6) 1815-1822

Väätäinen, K., Timonen, J., Hautajärvi, A., 1993 Development of a gas
method for migration studies in fractured and porous media, Mat. Res.
Soc. Symp. Proc. Vol 294 851-856.

Yamaguchi, T. and Nakayama, S., 1998. Diffusivity of U Pu and Am
carbonate complexes in a granite from Inada, Ibaraki, Japan studied by
through diffusion, J Cont. Hydrol. 35, 55-65