

Diffusion data in granite**Recommended values**

Yvonne Ohlsson, Ivars Neretnieks

Department of Chemical Engineering and Technology,
Chemical Engineering, Royal Institute of Technology,
Stockholm, Sweden

October 1997

DIFFUSION DATA IN GRANITE

RECOMMENDED VALUES

Yvonne Ohlsson, Ivars Neretnieks

**Department of Chemical Engineering and Technology,
Chemical Engineering, Royal Institute of Technology,
Stockholm, Sweden**

October 1997

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.

Information on SKB technical reports from 1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26), 1981 (TR 81-17), 1982 (TR 82-28), 1983 (TR 83-77), 1984 (TR 85-01), 1985 (TR 85-20), 1986 (TR 86-31), 1987 (TR 87-33), 1988 (TR 88-32), 1989 (TR 89-40), 1990 (TR 90-46), 1991 (TR 91-64), 1992 (TR 92-46), 1993 (TR 93-34), 1994 (TR 94-33), 1995 (TR 95-37) and 1996 (TR 96-25) is available through SKB.

DIFFUSION DATA IN GRANITE

Recommended values

Yvonne Ohlsson, Ivars Neretnieks

**Department of Chemical Engineering and Technology
Chemical Engineering
Royal Institute of Technology
S-100 44 Stockholm**

October 1997

Keywords: Matrix diffusion, surface diffusion, anion exclusion, diffusivity, adsorption, radionuclide, radioactive waste

ABSTRACT (ENGLISH)

This report is one of the reports that form a basis for the safety report SR 97 (säkerhetsrapport 97). The project SR 97 shall together with other reports provide a full picture of the performance of the repository concept. SR 97 shall show the long term performance of a deep geologic repository for encapsulated spent fuel and other long lived wastes.

Diffusion data for radionuclide transport in the porous matrix of rock are proposed for Swedish rock and ground waters, for performance assessment. Suggested data are based on an experimental diffusion study by Johansson et al. (1996). Tritiated water was used as noninteracting diffusing species in stationary diffusion experiments in Äspö fine grained granite and diorite. These data, for tritiated water, were used as reference in our study. For other species the effective diffusivities could be predicted from knowledge of the relative behaviour of these species to that of tritiated water. The behaviour of these species is influenced by the difference in free water diffusivity and sometimes the existence of anion exclusion or surface diffusion. Apparent diffusivities are also calculated using sorption data provided by Carbol and Engkvist (1997) in addition to the effective diffusivities. Data are proposed for high saline and low saline ground water conditions.

ABSTRACT (SWEDISH)

Denna rapport är en av de rapporter som ligger till grund för säkerhetsredovisningen SR 97 (säkerhetsrapport 97). Projekt SR 97 skall tillsammans med andra redovisningar ge en helhetsbild av säkerheten hos förvarskonceptet. SR 97 skall redovisa den långsiktiga säkerheten för djupförvar av inkapslat utbränt bränsle och annat långlivat avfall.

Diffusionsdata för radionuklidtransport i poröst berg föreslås till säkerhetsanalys för svenska berg och grundvatten. Föreslagna data har baserats på en experimentell diffusionsstudie av Johansson et al. (1996). I denna studie användes tritierat vatten som icke-interaktiv diffunderande molekyl i stationära diffusionsexperiment i Äspö finkorning granit och diorit. Dessa data, för tritierat vatten, användes som referensdata i vår studie. För övriga ämnen kunde effektiva diffusiviteter beräknas genom kännedom om hur dessa uppför sig relativt det tritierade vattnet. Deras diffusion påverkas av sådant som skillnader i diffusivitet i fritt vatten och ibland av fenomen som anjonexklusion eller ytdiffusion. Även apparenta diffusiviteter beräknas mha sorptionsdata tillhandahållna av Carbol och Engkvist (1997), samt de effektiva diffusiviteterna. Data föreslås för grundvatten med höga och låga salthalter.

TABLE OF CONTENTS

SUMMARY AND CONCLUSIONS	v
1 INTRODUCTION	1
2 METHODOLOGY FOR PROPOSAL OF DATA	2
2.1 POROSITY	2
2.2 DIFFUSIVITY	2
2.2.1 Ground waters of high ionic strength	4
2.2.2 Ground waters of low ionic strength	5
3 PROPOSED DATA	7
4 DISCUSSION	9
5 NOTATION	11
6 REFERENCES	12
APPENDICES	
Appendix 1:	Discussion of data uncertainties
Appendix 2:	Paper presented at Geotrap Workshop #2. Paris, 9-11 June, 1997. Neretnieks I. Impact of uncertainties in chemical and other entities on radionuclide migration from a repository for spent nuclear fuel.

SUMMARY AND CONCLUSIONS

This report is one of the reports that form a basis for the safety report SR 97 (säkerhetsrapport 97). The project SR 97 shall together with other reports provide a full picture of the performance of the repository concept. SR 97 shall show the long term performance of a deep geologic repository for encapsulated spent fuel and other long lived wastes.

Matrix diffusion in rock is considered very important in retarding radionuclides escaping from a deep geologic repository for nuclear waste. Many experimental studies have been performed to gain knowledge of the diffusion and sorption processes, and to obtain reliable data for safety assessment calculations. In a compilation of porosity, sorption and diffusion data, Brandberg et al. (1991) proposed an effective diffusivity of $1 \cdot 10^{-13}$ m²/s for all nuclides in Finnsjön granite, as a representative value for the SKB-91 study. The representative porosity was 0.5 %

Extracting representative data from the vast amount of data available from different experiments, with different rock material and under different conditions, turned out to be very complicated. Experimental data from a large quantity of samples from the area of interest, using the same experimental technique and conditions, would be desirable. These data do not exist, wherefore we used another approach in order to propose diffusion data for rock, for performance assessment. The method is explained below.

In an experimental diffusion study, Johansson et al. (1996) measured the diffusion of both an uncharged molecule (HTO) and some cations in salty water in *the same rock samples* (Äspö fine grained granite and diorite). The relation between the effective diffusivity of the cations and tritiated water in the rock samples was about the same as the relation in free water. This means that the diffusion is, as expected in highly saline pore solutions, hindered only by the porous rock, and is not significantly affected by the negatively charged pore walls. The study also provided sufficient information about porosity, ground water composition etc. to make it serve as a basis for our report with HTO as a "reference molecule".

To determine the effective diffusivities of other species we then related them to this reference nuclide. This can be done if the differences in behaviour of other species, in relation to the reference molecule, is known. These differences can be deviations in free water diffusivity and sometimes anion exclusion or surface diffusion. In this manner, *effective diffusion coefficients* can be predicted for all nuclides of interest by measuring only the diffusion of a non charged non sorbing molecule and relating the others to this reference species. A retardation factor based on the proposed sorption coefficients for SR 97 (Carbol and Engkvist (1997)) is used for calculation of the *apparent diffusivities*.

The set of diffusion coefficients proposed is related to the reference molecule. Values are therefore *probable* values assessed from theory and from experiments with a limited number of rock samples. More investigations are still needed to obtain better knowledge on the various processes affecting diffusion in rock.

1 INTRODUCTION

Matrix diffusion has been concluded to be a most important retardation mechanism for nuclides escaping from deep bedrock repositories for nuclear waste. Many studies of matrix diffusion have been carried out since the late 1970:s, and experimental work is still going on and methods are being refined. Faster and in-situ experimental methods would be desirable in order to obtain site specific data and to make it easier to evaluate the phenomena affecting diffusion in porous rock.

In the SKB 91 study, Brandberg et al. (1991) compiled porosity, sorption and diffusivity data for Swedish rock and for bentonite clay. In the case of rock, an effective diffusion coefficient of $1 \cdot 10^{-13}$ m²/s was proposed for all nuclides in Finnsjön granite with a diffusion porosity of 0.5 %. Since then, more diffusion experiments have been carried out, and more knowledge of the various phenomena affecting diffusion in the porous rock has been obtained.

Diffusion studies have recently been reviewed by Ohlsson and Neretnieks (1995). Based on these and later studies, a set of diffusion data is now proposed for the SR 97 project. The large amount of data that is compiled and evaluated in the review forms a basis for the recommendations in this short report. It was found that the data for different nuclides are not easily compared. Often they do not seem to be consistent. This is to a large extent due to the fact that measurements have been performed on different pieces of rock. The variation of properties between different pieces is often large even if the pieces have been obtained from the same bore core and from locations near each other. We have therefore based our proposed data on the prerequisite that representative measurements of diffusion properties are available for at least one noninteracting species. This species can be e.g. tritiated water or some dissolved gas such as methane. Also electrical conductivity measurements can be used to obtain the diffusion information in the form of the formation factor. To obtain apparent diffusivities, sorption coefficient data must also be available.

Then, based on theoretical considerations and the observations and conclusions compiled in the report Ohlsson and Neretnieks (1995), diffusivity data can be estimated for all nuclides for which measurements are not available. We outline the reasons for our approach below.

The behaviour of the various species under different conditions is taken into account, and data are proposed for the different nuclides in the reference ground waters (Laaksoharju et al. 1996) in SR 97.

2 METHODOLOGY FOR PROPOSAL OF DATA

Data considered in this report are for diffusion in pristine rock, which means that the rock contains no weathered surfaces and is not situated close to any fractures, so that the porosity, structure and composition is that of the undisturbed mass of rock. Investigations of materials containing fracture surfaces mostly show larger porosities and also effective diffusivities than the intact rock, and in most studies, the surfaces do not seem to get clogged by fissure coating material (Skagius (1986)). Hydrothermally altered rock generally show larger porosities than the intact rock. However the new pores created are extremely small, wherefore effective diffusivities should not be radically different from that of the undisturbed rock. (Mazurek et al. (1997)).

2.1 POROSITY

The porosity of rock can be determined by many methods, the most common being the water saturation method. Here, the volume of the rock piece and the difference in weight between the dry rock and when saturated with water, are used to calculate the porosity. Various methods for measuring porosity have recently been evaluated by Rasilainen et al. (1996). The evaluated methods, among them the water saturation method, seem reliable and comparable. The proposed porosity for Swedish rock is 0.5 %. The chosen value is based on measurements on small samples (1-2 cm) of Äspö diorite used for through-diffusion experiments, Johansson et al. (1996). However, variations between 0.1 and 1 % are common and when site specific data are available these should be used.

2.2 DIFFUSIVITY

Matrix diffusion in crystalline rock has been studied for many years in different investigations with different aims. Sometimes the focus has been on evaluating certain phenomena and influences of different conditions, and sometimes the interest has been in producing data for safety assessment calculations. The richness of data provided in the scientific reports and papers, should make it possible to extract representative data for calculations. However, many factors complicate this and also suggest that it is not necessary to use all existing data to extract the appropriate ones for the case we are interested in. These factors can be differences in rock properties and structures, different experimental methods, not completely specified water and rock properties etc. Diffusion coefficients for a specific nuclide in granite can differ by factors of 10-100 or more between different studies, using different rock samples, even though experimental conditions otherwise seem to be similar. In gathering available data and trying to find representative data, we have

found that there are no studies that systematically have used the same rock sample to measure diffusivities and sorption coefficients for a majority of the nuclides of interest. This is of course understandable, but because the rock properties vary strongly between one sample and another, even if they have been taken at locations very near each other, this variation often overshadows the differences between different nuclides.

We have therefore been forced to use another approach which is outlined below, and described in detail with the appropriate equations later.

We propose that when a specific site is to be modelled, diffusion data for a noninteracting species, such as tritiated water, is used as a basis for all other species that have not been measured. The effective diffusivities for other species are then obtained by proportioning with the respective diffusivities in free water. This gives a correction of up to a factor 2-3 compared to the noninteracting species. In addition account is made for retardation of cationic species. A retardation factor based on the measured sorption coefficient is used for calculation of the apparent diffusivity.

For *low ionic strength* waters anions, such as iodide, chloride, perchlorate etc., have lower effective diffusivities by one order of magnitude, due to ion exclusion, Eriksen and Jansson (1996), Lehtikoinen et al. (1992). Also for low ionic strength waters some cationic species that sorb by ion exchange, such as cesium, strontium and sodium, have an effective diffusion coefficient that is one to two orders of magnitude higher than that of tritium due to surface diffusion. These species with these properties are specifically noted in the report. Also other species have been found to have surface diffusion in clays. Because there is not yet experimental evidence that this also is the case in granite, surface diffusion is not considered for these species in this report.

The detailed procedure and the rationale for the proposal is shown below. The proposal is consistent with measurements when made on the same rock samples. The use of one order of magnitude decrease and increase of the diffusivity due to ion exclusion and surface diffusion respectively is of course an approximate value. The use of *one order of magnitude* indicates its variability and uncertainty. For anions, one order of magnitude decrease seem to be a fairly good estimate of the effect. By using one order of magnitude increase for those nuclides that move by surface diffusion the effect is not overestimated and the values are thus conservative in the case of a rock mass which acts as a sink.

2.2.1 Ground waters of high ionic strength

The basic assumption for the subsequent treatment is that a noninteracting species (tritiated water is assumed to be such a species) will have an effective diffusivity that is less than in pore water by a factor equal to a rock property called the formation factor. The latter accounts for several mechanisms. The porous rock matrix decreases the available cross section for transport and the porosity is the volume fraction of the rock that allows transport. The tortuosity makes the transport paths longer and the constrictivity accounts for hindrance in the narrow passages underway. The formation factor can be measured by using e.g. tritiated water or a dissolved gas or some other noncharged species with a comparable size to the nuclides. The relation between these different entities is shown below.

The free water diffusivity D_w is $2.4 \cdot 10^{-9}$ m²/s for tritiated water and the formation factor F_f of around $4\text{-}5 \cdot 10^{-5}$ for Swedish fresh rock (Skagius (1986), Johansson (1996)) gives an effective diffusivity of around $1 \cdot 10^{-13}$ according to the relation:

$$D_e = D_p \varepsilon = D_w \varepsilon \frac{\delta_D}{\tau^2} = D_w F_f \quad (1)$$

where D_p is the pore diffusivity and D_e the effective diffusivity. ε is the rock porosity and $F_f = \varepsilon \cdot \delta_D / \tau^2$, the formation factor which takes into account the structure of the porous system in the rock. δ_D is the constrictivity and τ^2 the tortuosity.

In studying Sr^{2+} , Na^+ and Ca^{2+} diffusion in Äspö diorite and in fine grained granite (Johansson et al. (1996)) and using HTO as a non-sorbing reference, the relation between the effective diffusivity of the cations and HTO in the rock samples was about the same as the relation in free water. This means that the diffusion is, as expected in highly saline pore solutions, hindered only by the porous rock, and is not significantly affected by the negatively charged pore walls. The water used was the Äspö water, which is a highly saline water with the ionic strength 0.25. The suggested effective diffusivity is then given by:

$$D_e = D_{e,\text{HTO}} \frac{D_w}{D_{w,\text{HTO}}} \quad (2)$$

for waters of high ionic strength. Measuring the diffusivity of tritiated water in representative rock at a sampling location where the ground water is of high ionic strength, then gives representative effective diffusivities of all species of interest, knowing their free water diffusivity. Another way is to calculate the formation factor from electrical conductivity measurements which directly gives the formation factor, and using this value to calculate the effective diffusivity of each specie.

Sorbing species are retarded and this is accounted for in the apparent diffusivity. The apparent diffusivity, D_a , can then be calculated from the following expression:

$$D_a = \frac{D_e}{\varepsilon + \rho K_d} \quad (3)$$

knowing the sorption coefficient, K_d , the rock bulk density, ρ , and the rock porosity, ε . In the proposed data (table 2), this has been carried out using the study of Johansson et al. (1996) and sorption data proposed by Carbol and Engkvist (1997).

2.2.2 Ground waters of low ionic strength

In ground waters of low ionic strength, it has been found that the effective diffusivities of some species are lower, or higher, than expected. For anions the D_e is lower by a factor of about 10 or more, and for ion exchanging cations, D_e is higher by a factor of about 10-100. In the case of anions, this has been explained to be due to electrostatic repulsion of the negatively charged anions to the negatively charged pore walls, so that the anions do not have access to the whole pore volume, see Figure 1. The phenomenon is known as anion exclusion. At high ionic strengths the surface charge of the pore walls is suppressed and the effect is absent. Anion exclusion has been described in detail in a recent report on diffusion in compacted bentonite, where the effects are very pronounced (Yu and Neretnieks 1996).

For ion-exchanging cations, the phenomenon causing the increased effective diffusion is known as surface diffusion (Skagius (1986), Lehtikoinen et al. (1992), Yamaguchi et al. (1993), Neretnieks (1994), Eriksen and Jansson (1996)). The effective diffusion is then composed of two parts; diffusion in the free pore water, and diffusion in the sorbed phase, see Figure 1. Because of the higher concentration of cations in the sorbed phase than in the free pore water, this effect could contribute substantially to the diffusion. Equation 3 is therefore modified in the following manner:

$$D_a = \frac{D_e}{\varepsilon + \rho K_d} = \frac{D_p \varepsilon + D_s \rho K_d}{\varepsilon + \rho K_d} \quad (4)$$

where D_s is the surface diffusivity and $D_p \epsilon$ is the effective diffusion due only to transport in the free pore solution. The cations which in experiments have been found to behave in this manner are Cs, Na and Sr. In diffusion experiments in clay, also Pa and Ra have been found to diffuse in the sorbed state (Yu and Neretnieks (1996)). So far there is no experimental diffusion data for these species in granite.

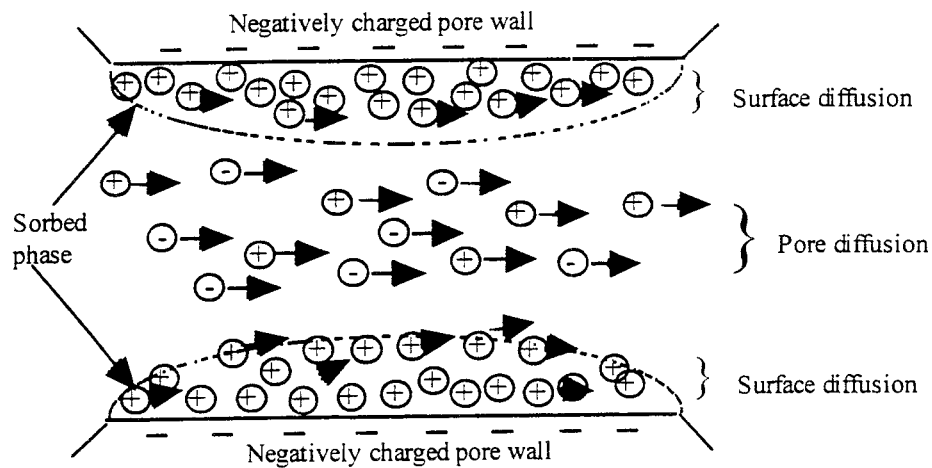


Figure 1. Schematic description of ions diffusing in a pore with negatively charged pore surfaces. Anions are restricted to a smaller pore volume due to electrostatic repulsion. Ion exchanging cations concentrate in the sorbed, but mobile, phase.

Proposed effective diffusivities of anions in waters of low ionic strength is suggested to be a factor of ten lower than in saline waters. For cations that have been found to migrate in the sorbed phase, the effective diffusivity is suggested to be a factor of ten higher than in saline solutions. There have been no systematic measurements of surface diffusion in different ionic strength pore waters covering a wide range of salinities.

In this report we mean by high ionic strength or high salinity a salt concentration of 10 000 mg/l or more. With low salinity waters we mean salt concentrations around 1 000 mg/l or less. There is a need to study these effects further.

3 PROPOSED DATA

Using available data and the reasoning about the effects of the negatively charged surfaces discussed in 2.1 and 2.2, makes it easy to set up a calculation scheme for the estimation of diffusion coefficients for different species. What is needed is the free water diffusivities (Gray (1972), Li and Gregory (1974), Skagius (1986)) and the K_d -values for each specie, and also the formation factor, either from electrical conductivity measurements or from measurements of the diffusion of tritiated water, HTO. This calculation scheme could look like the following Table 1, where IE denotes "ion exclusion" i.e. anions, SD denotes "surface diffusion" i.e. ion exchanging cations. f is the factor taking into account the effect of surface diffusion or anion exclusion as explained in section 2.2.2.

An advantage with this type of calculation scheme is that it is easy to modify. As more is known about the diffusion behaviour of the various nuclides, the factor f can be easily be changed in the scheme.

Table 1. Calculation scheme to obtain D_e and D_a when D_w , K_d , f , ϵ , ρ , and F_r are known

ionic strength: Species:	High				Low			
	D_w	$D_e=D_w \cdot F_f$	K_d	$D_a=D_e/(\epsilon+\rho K_d)$	f	K_d	$D_e=D_w \cdot F_f \cdot f$	$D_a=D_e/(\epsilon+\rho K_d)$
HTO					1			
IE					0,1			
SD					10			
other					1			

In Table 2 diffusivities for Swedish rock of 0.5 % porosity and the rock density 2750 kg/m³ (Johansson et al. (1996)), are proposed for the various reference waters in the safety assessment study SR 97. The K_d -values used in the table were proposed by Carbol and Engkvist (1997). For nuclides marked **, no sorption data were proposed. There are, though, diffusion data available for these nuclides, which can be interesting even though the nuclides are not important for safety assessment purposes.

Table 2 . Proposed data for a rock with D_e for HTO equal to $1 \cdot 10^{-13}$ m²/s.

Ionic strength:		High			Low			
Water:		KAS02, BF101			KF107, KG104			
Element/ chemical form or redox state	$D_w \cdot 10^9$ (m ² /s)	$D_e \cdot 10^{13}$ (m ² /s) $D_e = D_w \cdot F_r$	K_d (m ³ /kg)	D_a (m ² /s) $D_a = D_e / (\epsilon + \rho K_d)$	f	$D_e \cdot 10^{13}$ (m ² /s) $D_e = D_w \cdot F_r \cdot f$	K_d (m ³ /kg)	D_a (m ² /s) $D_a = D_e / (\epsilon + \rho K_d)$
HTO	2.4	1.0*	0	$2 \cdot 10^{-11}$	1	1.0	0	$2 \cdot 10^{-11}$
Ac/Ac (III)	(1)	0.4	3	$5 \cdot 10^{-18}$	1	0.4	3	$5 \cdot 10^{-18}$
Ag/Ag (I)	1.7	0.71	0.05	$5 \cdot 10^{-16}$	1	0.71	0.5	$5 \cdot 10^{-17}$
Am/Am (III)	(1)	0.4	3	$5 \cdot 10^{-18}$	1	0.4	3	$5 \cdot 10^{-18}$
Br/Br**	2.0	0.83	0	$2 \cdot 10^{-11}$	0.1	0.08	0	$2 \cdot 10^{-12}$
C/HCO ₃ ⁻	1.2	0.50	0.001	$2 \cdot 10^{-14}$	0.1	0.05	0.001	$2 \cdot 10^{-15}$
Cd/Cd (II)	0.72	0.30	0.02	$5 \cdot 10^{-16}$	1	0.30	0.1	$1 \cdot 10^{-16}$
Cl/Cl ⁻	2.0	0.83	0	$2 \cdot 10^{-11}$	0.1	0.08	0	$2 \cdot 10^{-12}$
Cm/Cm (III)	(1)	0.4	3	$5 \cdot 10^{-18}$	1	0.4	3	$5 \cdot 10^{-18}$
Co/Co (II)	0.70	0.29	0.02	$5 \cdot 10^{-16}$	1	0.29	0.1	$1 \cdot 10^{-16}$
Cs/Cs (I)	2.1	0.88	0.05	$6 \cdot 10^{-16}$	10	9	0.5	$6 \cdot 10^{-16}$
Eu/Eu (III)	(1)	0.4	2	$7 \cdot 10^{-18}$	1	0.4	2	$7 \cdot 10^{-18}$
Ho/Ho (III)	(1)	0.4	2	$7 \cdot 10^{-18}$	1	0.4	2	$7 \cdot 10^{-18}$
I/I ⁻	2.0	0.83	0	$2 \cdot 10^{-11}$	0.1	0.08	0	$2 \cdot 10^{-12}$
Kr (inert gas)	(1)	0.4	0	$8 \cdot 10^{-12}$	1	0.4	0	$8 \cdot 10^{-12}$
Na/Na (I)**	1.3	0.54	-	-	10	5	-	-
Nb/Nb (V)	(1)	0.4	1	$1 \cdot 10^{-17}$	1	0.4	1	$1 \cdot 10^{-17}$
Ni/Ni (II)	0.68	0.28	0.02	$5 \cdot 10^{-16}$	1	0.28	0.1	$1 \cdot 10^{-16}$
Np/Np (IV)	(1)	0.4	5	$3 \cdot 10^{-18}$	1	0.4	5	$3 \cdot 10^{-18}$
Pa/Pa (IV,V)	(1)	0.4	1	$1 \cdot 10^{-17}$	1 [10]	0.4	1	$1 \cdot 10^{-17}$
Pd/Pd (II)	(1)	0.4	0.01	$1 \cdot 10^{-15}$	1	0.4	0.1	$1 \cdot 10^{-16}$
Pu/Pu (III,IV)	(1)	0.4	5	$3 \cdot 10^{-18}$	1	0.4	5	$3 \cdot 10^{-18}$
Ra/Ra (II)	0.89	0.37	0.02	$7 \cdot 10^{-16}$	1 [10]	0.37	0.1	$1 \cdot 10^{-16}$
Se/Se (IV,VI)	(1)	0.4	0.001	$1 \cdot 10^{-14}$	1	0.4	0.001	$1 \cdot 10^{-14}$
Sm/Sm (III)	(1)	0.4	2	$7 \cdot 10^{-18}$	1	0.4	2	$7 \cdot 10^{-18}$
Sn/Sn (IV)	(1)	0.4	0.001	$1 \cdot 10^{-14}$	1	0.4	0.001	$1 \cdot 10^{-14}$
Sr/Sr (II)	0.79	0.33	0.0002	$6 \cdot 10^{-14}$	10	3	0.01	$1 \cdot 10^{-14}$
Tc/TcO ₄ ⁻	(1)	0.4	0	$8 \cdot 10^{-12}$	0.1	0.04	0	$8 \cdot 10^{-13}$
Tc/Tc (IV)	(1)	0.4	1	$1 \cdot 10^{-17}$	1	0.4	1	$1 \cdot 10^{-17}$
Th/Th (IV)	0.15	0.063	5	$5 \cdot 10^{-19}$	1	0.063	5	$5 \cdot 10^{-19}$
U/U (IV)	(1)	0.4	5	$3 \cdot 10^{-18}$	1	0.4	5	$3 \cdot 10^{-18}$
Zr/Zr (IV)	(1)	0.4	1	$1 \cdot 10^{-17}$	1	0.4	1	$1 \cdot 10^{-17}$

* Measured value (Johansson et al. (1996))

** No sorption data are suggested for these nuclides

() Assumed values

[] Probable values but not used due to lack of experimental data.

4 DISCUSSION

In compiling data for safety assessment purposes, one confronts many contradictions in evaluating large amount of results and has to restrict and limit oneself to achieve comparable, consistent values founded on the theoretical understanding of the processes. Using data from many different studies gives a wide range of values, which are seldom comparable. All reported values are, of course, meaningful to study for the purpose of the understanding of the processes. Because of the variability of the rock samples they cannot simply be used to define the properties of a representative or average rock.

The first restriction in this compilation of diffusion data, was to limit ourselves to pristine rock, including no disturbed zones and no fractures. Then, we used one study which included the diffusion of a noninteracting species (HTO) and also provided sufficient information about porosity, ground water composition etc. From knowledge of the behaviour of other nuclides in relation to that of HTO, the effective diffusivity could then be predicted for these other nuclides and conditions.

The proposed data are then restricted to the unfractured host rock. For the disturbed zone and coated fissures, there seems to be no hindrance by clogging, but instead porosities and effective diffusion coefficients are the same or higher in these areas (Skagius (1986)). Mazurek et al. (1996) found that pore size distributions were affected by hydrothermal alteration, but that the newly created pores were extremely small.

The sorption data used for calculations of apparent diffusivities in table 2, have been supplied from the compilation of sorption data for SR 97 made by Carbol and Engkvist (1997). This means that these sorption data are not connected to the study on which estimations of effective diffusivities are based on in this report.

The results from experimental determinations have been questioned due to for example the small samples generally used in laboratory determinations, which could result in too high diffusivities due to the contribution from residual porosity which is less important in larger samples. In investigations of this effect, the decrease in diffusivity with sample length seems to disappear when pieces are longer than 4-5 cm (Johansson et al. (1996), Kumpulainen and Uusheimo (1989), Bradbury and Green 1986). The influence on the diffusion coefficient is commonly less than a factor two.

It has to be emphasised, that the proposed data are *probable* data which have been assessed from theory and from experiments with a limited number of rock samples from a limited area. More investigations are needed and development of reliable, preferably faster, in-situ and laboratory methods would be desirable. There are still considerable uncertainties in cases where there are not enough data to do accurate

predictions of behaviour, and more knowledge is essential. The use of a factor 0.1 and 10 taking into account anion exclusion and surface diffusion respectively shows the uncertainty in the understanding of the processes, but should not overestimate the effects. The simplicity of the kind of calculation scheme we suggest, facilitates adjustments as more becomes known about the phenomena.

5 NOTATION

D_a	Apparent diffusivity	m^2/s
D_e	Effective diffusivity	m^2/s
D_p	Pore diffusivity	m^2/s
D_s	Surface diffusivity	m^2/s
D_w	Diffusivity in free water solution	m^2/s
f	Factor taking into account ion exclusion and surface diffusion	-
F_f	Formation factor	-
K_d	Sorption coefficient	m^3/kg
δ_D	Constrictivity	-
ε	Porosity	m^3/m^3
ρ	Rock density	kg/m^3
τ^2	Tortuosity	-

6 REFERENCES

Bradbury M.H., Green A. *Retardation of radionuclide transport by fracture flow in granite and argillaceous rocks*. Chemical engineering division, UKAEA Harwell, United Kingdom 1986. EUR 10619

Brandberg F., Skagius K. *Porosity, sorption and diffusivity data compiled for the SKB 91 study*. SKB TR-91-16. Kemakta Consultants, Stockholm 1991.

Carbol P., Engkvist I. *Compilation of radionuclide sorption coefficients for performance assessment*. PI Chemical Consulting HB, Landvetter, Sweden. Feb. 1997

Eriksen T.E., Jansson M. *Diffusion of I, Cs⁺ and Sr²⁺ in compacted bentonite*. Royal institute of technology, Department of chemistry, Nuclear chemistry, S-100 44 Stockholm. SKB TR 96-16. 1996.

Gray D.E., *American Institute of Physics Handbook*, McGraw-Hill, New York, 1972.

Johansson H., Byegård J., Skarnemark G., Skålberg M. *Matrix diffusion of some alkali- and alkaline earth-metals in granitic rock*. Department of Nuclear chemistry, Chalmers University of Technology, S-412 96 Göteborg, Sweden. In symposium proceedings from Materials research society. Scientific basis for nuclear waste management XX. 1996 fall meeting, Dec 2-6. Boston, Massachusetts.

Kumpulainen H., Uusheimo K. *Diffusivities and electrical resistivity measurements in rock matrix around fractures*. Technical research centre of Finland. Reactor laboratory. Helsinki, Finland, Dec. 1989. YJT-89-19

Laaksoharju, Skårman, Gurban. *Vattendata för SR 97*. In preparation. 1996.

Lehikoinen J., Muurinen A., Olin M., Uusheimo K., Valkiainen M. *Diffusivity and porosity studies in rock matrix. The effect of salinity*. Technical research centre of Finland. Reactor laboratory. Espoo, Finland, Dec. 1992. Research notes 1394.

Li Y-H., Gregory S. *Diffusion of ions in sea water and in deep sea sediments*. *Geochimica et cosmochimica acta.* Vol. 38, p. 603-714. 1974

Mazurek M., Bossart P., Eliasson T. *Classification and characterization of water-conducting features at Äspö: Results of investigations on the outcrop scale*. Dec 1996. Swedish nuclear fuel and waste management co, Äspölaboratoriet, International cooperation report 97-01.

Neretnieks I. *Matrix Diffusion - How Confident are we?* Nuclear science and technology. Sixth EC natural analogue working group meeting. Proceedings of an international workshop held in Santa Fe, New Mexico, USA on 12-16 September 1994.

Ohlsson Y., Neretnieks I. *Literature survey of matrix diffusion theory and of experiments and data including natural analogues*. Department of chemical engineering, Royal institute of technology, Stockholm, Sweden. August 1995. SKB Technical Report 95-12.

Rasilainen K., Hellmuth K-H., Kivekäs L., Melamed A., Ruskeeniemi T., Siitari - Kauppi M., Timonen J., Valkiainen M. *An interlaboratory comparison of methods for measuring rock matrix porosity*. VTT research notes, Technical research centre of Finland. Espoo, Finland, 1996

Skagius K. *Diffusion of dissolved species in the matrix of some Swedish crystalline rocks*. PhD. Thesis. Department of Chemical Engineering, Royal Institute of Technology, Stockholm, Sweden, 1986

Yamaguchi T., Sakamoto Y., Senoo M. *Consideration on effective diffusivity of strontium in granite*. Journal of Nuclear Science and Technology. Tokyo Aug 1993. Vol. 30 (8) p.796-803

Yu Ji-Wei, Neretnieks I., *Diffusion and sorption properties of radionuclides in compacted bentonite*. SKB TR 97-12. 1997

Discussion of Data Uncertainties

General

The diffusivities in the rock matrix used in the simulations of nuclide migration are derived from measured values using mostly nonsorbing uncharged tracers. They are then adjusted to account for the effects of ion exclusion for negatively charged species and surface diffusion for positively charged species. In addition nuclide migration through the rock and the simulation results will depend on the retardation of diffusion due to sorption. Differences in the rock porosity and the rock density will only marginally influence the results and will not be further treated here.

The data are obtained in the following way. The effective diffusivity D_e of an uncharged solute is obtained from measured values of the formation factor, F_f , of the rock matrix and literature data on the diffusivity of the species in unconfined water, D_w .

$$D_e = D_w F_f \quad (1)$$

The formation factor is obtained by either diffusivity or electrical conductivity measurements. The formation factor is subject to uncertainty and also varies considerably from sample to sample, i.e. it has a natural variability. The diffusivity in unconfined water is subject to a much smaller uncertainty and will not be discussed further.

In diffusion of charged species, the negative anions and the positive cations are also influenced by the presence of the mineral surfaces which are negatively charged at the prevailing water compositions. A factor, f , that accounts for these effects is used when the salt concentration in the water is low.

$$D_e = D_w F_f f \quad (2)$$

It is only then that the effect is of importance. In more saline waters the effect becomes small and need not be accounted for. Typically the correction factor has a magnitude of 0.01-0.1, decreasing the mobility of anions and 10-100 increasing the mobility of some cations that are concentrated near the mineral surfaces by the charges in the electrical double layer. This applies typically to Cs, Sr and other alkaline and alkaline earth metals. These effects have not been found for the actinides except for protactinium. Thus a further major uncertainty is introduced by this effect.

In the formulation of the transport model and in some simulation programs the concept of an apparent diffusivity is used. This diffusivity also accounts for the seeming retardation of the diffusion due to withdrawal of the nuclide from the water by sorption on the mineral surfaces. It is accounted for by the sorption coefficient $K_v = \rho K_d$ in the following manner.

$$D_a = \frac{D_e}{(\varepsilon + K_w)} \quad (3)$$

The uncertainties in the sorption coefficients are discussed elsewhere.

Use in SR 97

The data will be used in the models FarF31 and CHAN3D in SR97. In both models the effective diffusivity enters the models through the following group of parameters.

$$\frac{\text{FWS}}{Q} \sqrt{D_e K_v} \quad (4)$$

It is seen that the *flow wetted surface*, FWS, and the water flow rate, Q, in a fracture have a dominating effect on the nuclide transport, Neretnieks (1997). Compared to these entities, uncertainties in effective diffusivities and sorption coefficients (K_d 's) are of less importance due to a square root dependence. The effect of an uncertainty of an entity on the nuclide migration is even more pronounced because the group of parameters enters into the model in a way that is extremely non-linear. For the transport in a single channel the following very simple expression shows how the effluent concentration is influenced by the parameters.

$$c_{\text{out}} = c_{\text{in}} e^{-\lambda t} \text{Erfc} \left[\frac{LW}{Q} \sqrt{\frac{D_e K_v}{(t-t_0)}} \right] \quad (5)$$

The transport of the nuclides in the complex network of pathways is modelled by much more complex models than the one above. However this very simple expression captures the influence of a change in parameter values in the more complex models surprisingly well. It can thus be used to assess the impact of the uncertainty of the matrix diffusivity and also the flow rate and the flow wetted surface.

Dependencies on Other Parameters, Ranges of Validity, Sensitivities

Effective diffusivities are influenced by, in addition to the rock matrix properties, the ionic strength and the composition of the ground water. The relations are somewhat complicated and exact predictions are difficult to perform. The tendencies and ranges are, though, fairly well known and therefore estimates of the effects are given in the form of a correction factor, f , to account for different salinities. For now, low ionic strength water means water with around 1000 mg/l of salt or less and high ionic strength meaning 10000 mg/l or more. A better understanding of these effects is forthcoming but not yet ripe to be expressed quantitatively.

In our recommendations we propose an f of 0.1 for anions in low salinity waters. For the performance assessment it matters very little what data are used for the anions because the only really important long lived radioactive anion is I-129. This will reach the effluent in the same concentration irrespective of the data used. The difference will be in the time it arrives. Its decay will not be influenced because of its long half life.

For the cations we propose an f -factor of 10. It is a best estimate as far as such an estimate can be made. The value is quite uncertain and could be somewhat smaller but could also be considerably larger. We have chosen the value to be cautiously conservative.

Characterisation of Uncertainties

The formation factor, which is the starting point for estimating the effective diffusivity has a natural variability even in locations very near to each other. This has been found in the laboratory (Skagius 1986) and in the field (Birgersson 1988). A variability along the flow path does not influence the results much as the system is near liner and the variations are averaged out. Only the mean value will matter Neretnieks (1997).

It is, however, difficult to separate out the variability from the uncertainty due to the following reasons. When rock samples are taken they are not taken in such a way that they can be considered to be representative of the rock near the surfaces where the water flows. A number of observations show that the rock near to fractures is more porous and has higher formation factors than rock further from the fracture. On the other hand, at Äspö recent observations show the opposite tendency at some locations. Also the rock stresses are higher at depth than what is normally used in laboratory experiments. This effect is accounted for using laboratory and field data on the influence of rock stresses. Skagius (1986) found that the diffusivities were reduced 20-70 % of the value when under 300-350 bar stress. Similar results were obtained by Birgersson et al (1988) in their field experiments in undisturbed rock at natural stress conditions at 360 m depth.

Variability also includes variations due to the inhomogeneity of the rock matrix, and of the differences between the rock material at different sites. Data are related to the specific site from which samples were taken for measurements. Experiments were carried out on small samples and it has been found that the diffusivity decreases somewhat when larger samples are used.

The measurement methods used for assessing the diffusivity, direct diffusivity measurement and electrical conductivity measurements, agree reasonably well. Compared to the other variabilities they are not deemed to be large sources of error.

Quantification of Uncertainties

It not possible to objectively assign numbers to uncertainties. Part of what we sometimes call uncertainties are in fact variabilities. These can be accounted for by making a sufficiently large number of measurements which will give the stochastic distribution. Uncertainties would be present if there are only a few measurements and when other factors which cannot be quantified can influence the results. Examples are pore water compositions along the flow path and the possible presence of fracture alteration materials along the pathway.

Nevertheless, if asked for reasonable estimate on the diffusion data based on only a limited number of measurements we would propose that the data be obtained by the procedure outlined above. The uncertainty of the mean of the nuclide diffusivities would be on the order of a factor 3 to 5 for high salinity waters. For low salinity waters the uncertainty would be on the order of a factor of 10 for charged species. In this statement probably a part of what is called uncertainty is variability.

Correlations

We use no correlation but a computation scheme to assess the diffusivities of such species that have not been experimentally measured on the site specific rock matrix.

Suggested *apparent diffusivities* are related to the K_d -values provided by Carbol and Engkvist (1997).

Treatment in Safety Assessment

The proposed data and procedures are a sort of best estimate values but with some bias towards being conservative. These should be used for a central case. A pessimistic case with a factor 3 and a factor 10 should then be used for high and low salinity waters respectively.

A stochastic analysis is not expected to give much more insights than what can be derived from the basic analytical solution shown above and discussed in the paper by Neretnieks (1997). The reason is that with the conceptual and mathematical models used the variabilities of diffusivity, sorption coefficient and flow wetted surface are effectively averaged along the pathway. The situation might be different if there were known correlations between the said variables. Even then the averaging of the group along the pathway would give better insights than numerical stochastic analysis.

References

- Birgersson L. *Diffusion in the matrix of granitic rock, field test in the Stripa mine. Licentiate treatise*. Department of Chemical Engineering, Royal Institute of Technology, Stockholm, Sweden, 1988.
- Carbol P., Engkvist I. *Compilation of radionuclide sorption coefficients for performance assessment*. PI Chemical Consulting HB, Landvetter, Sweden. Feb. 1997
- Neretnieks I. *Impact of uncertainties in chemical and other entities on radionuclide migration from a repository for spent nuclear fuel*. Geotrap workshop #2. Basis for modelling the effects of spatial variability on radionuclide migration. Paris 9-11 June, 1997.
- Skagius K. *Diffusion of dissolved species in the matrix of some Swedish crystalline rocks*. PhD. Thesis. Department of Chemical Engineering, Royal Institute of Technology, Stockholm, Sweden, 1986

APPENDIX 2

Impact of Uncertainties in Chemical and other Entities on Radionuclide Migration from a Repository for Spent Nuclear Fuel

Ivars Neretnieks

**Dept. Chemical Engineering and Technology
Royal Institute of Technology,
Stockholm,
Sweden**

Paper presented at Geotrap Workshop #2. Paris, 9-11 June, 1997

Impact of Uncertainties in Chemical and other Entities on Radionuclide Migration from a Repository for Spent Nuclear Fuel

Ivars Neretnieks

Dept. Chemical Engineering and Technology
Royal Institute of Technology,
Stockholm
SWEDEN

ABSTRACT

The migration of radionuclides in the geosphere is influenced by many factors. The primary factors that influence their mobility in the geosphere are: The water flowrate, the sorption properties in the rock matrix, the matrix diffusion properties and the flow wetted surface along the flowpath. There are also some secondary factors such as hydrodynamic dispersion, flowpath geometry, fracture filling materials etc.

The water flowrate and flowpaths are assessed by hydrodynamic modelling using data from primarily borehole measurements. Some inherent uncertainties emanating from both conceptual as well as measurement uncertainties will be briefly discussed.

Sorption data are uncertain due to several reasons. The water chemistry, especially the salinity of the water, its pH and redox conditions as well as encountered minerals can vary along the flowpath. In addition there is a variability i.e. uncertainty in the measured data. Small amounts of fracture filling and alteration materials will not much influence the migration over long times whereas larger amounts of strongly sorbing alteration minerals can have a strong impact on the short lived nuclides.

Diffusion of the nuclides into the matrix of the porous crystalline rock and sorption on the internal surfaces is a very strong retardation mechanism. Diffusion properties in the same rock matrix can vary considerably over short distances and there is also a considerable uncertainty in data.

The at present largest uncertainty is due to the largely unknown flow wetted surface encountered by the mobile water. This entity has been much less studied than sorption and diffusion properties. It is also the entity that is most difficult and expensive to assess because it must be done in the field and on a large scale.

The paper discusses the variabilities and uncertainties of the primary factors and exemplifies and illustrates the impact on the mobility of some of the nuclides of most concern by some sample calculations.

Introduction and background

To assess the impact of uncertainties in chemical and other transport parameters on the migration of radionuclides it is necessary to understand how these mechanisms influence transport. The main mechanisms are: Sorption on fracture surfaces and in possible gouge material, diffusion into the rock matrix and sorption on the inner surfaces. The contact surface between the flowing water and the rock is thus a key entity. The water flux also is an important entity but lies somewhat outside the scope of this paper. Its impact will nevertheless be shown and compared to that of the other entities.

For illustration purposes the simplest of models is presented where the main mechanisms are accounted for. It starts with a very simple geometry to highlight the mechanisms and then is generalised to handle variable properties along the flowpath. Water flows in a fracture with an aperture $2b$ with a flowrate Q . The solute can sorb onto the fracture surface with a surface sorption coefficient K_a . There is some fracture filling material that can sorb the nuclides. The solute can also diffuse into the porous rock matrix which has a porosity ϵ . The solute can in addition sorb on the inner surfaces of the rock matrix. The distribution coefficient is $K_v = K_d \rho$. The surface sorption can also be thought of as resulting from very fast sorption in a thin layer with thickness d of some material that is readily accessed by the nuclide. K_a is then simply the product of the thickness of this material and its volume sorption coefficient. $K_a = d K_{da} \rho$.

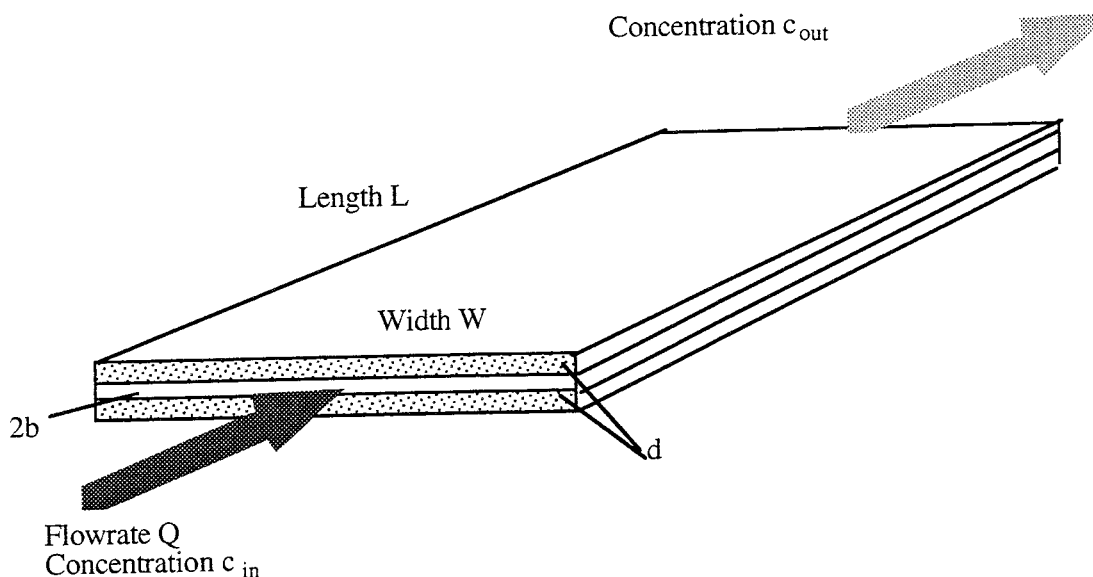


Figure 1. A fracture with aperture $2b$ and fracture alteration material of total thickness $2d$ has a water flow of Q , a length L and width W .

The rock on each side of the fracture has an altered region with a total thickness of $2d$. The flowrate of water in the fracture is Q m^3/s , the width is W m and the length is L m. The mass sorption coefficients of the alteration layer is K_{da} , that of the good rock is K_d and that of the filling material is K_{df} .

The concentration at the inlet of the fracture is suddenly raised to c_{in} and kept so except for the decay. The inlet concentration thus will decrease over time as $c = c_{in} e^{-\lambda t}$. If we assume that there is no diffusional resistance in the alteration layer and that this and the gouge material is instantaneously equilibrated with the water in each location, the outlet concentration from the fracture is obtained as [1]

instantaneously equilibrated with the water in each location, the outlet concentration from the fracture is obtained as [1]

$$c_{out} = c_{in} e^{-\lambda t} \operatorname{Erfc} \left[\frac{LW \sqrt{D_e K_v}}{Q \sqrt{t - t_o}} \right] \quad (1)$$

$K_v = K_d \rho$ is the volumetric sorption coefficient of the rock matrix, $K_{vf} = K_{df} \rho_f$ and $K_{va} = K_{da} \rho_a$. D_e is the effective diffusivity in the good rock and

$$t_o = \frac{LW}{Q} 2(b + bK_{vf} + dK_{va}) \quad (2)$$

$$t_o = t_w \left(1 + K_{vf} + \frac{d}{b} K_{va} \right) = t_w R_a \quad (3)$$

t_o is the tracer residence time if it were only affected by the water residence time and the retardation by fracture filling (gouge) and alteration materials.

The retardation factor caused by instantaneous reversible sorption on the fracture filling and alteration materials R_a is

$$R_a = 1 + K_{vf} + \frac{d}{b} K_{va} \quad (4)$$

Equation (3) warrants some comments. If the two volumetric sorption coefficients for the gouge and the alteration material are zero, the expression reduces to be the water residence time in the fracture. If the nuclide is sorbed on the gouge material, the second term in the parenthesis is not zero and the time t_o is the nuclide residence time. Because the volumetric sorption coefficient K_{vf} is much larger than 1, t_o will be very much larger than the water residence time and in fact the water residence time will in practice no longer influence the residence time of the sorbing nuclide. The same applies for the sorption in the alteration layer.

We note that the entity that has a dominating influence on the nuclide transport is the ratio of the flow wetted surface, FWS, to the water flowrate wetting this surface $2LW/Q$. Uncertainties in the matrix diffusivity and the matrix sorption coefficient D_e and K_v do not have as strong an influence as the flow wetted surface and the local flowrate because of the square root effect. This means that an uncertainty of a factor of ten in the FWS or the local flowrate will influence the group $\frac{LW}{Q} \sqrt{\frac{D_e K_v}{(t - t_o)}}$ by a factor of ten when an uncertainty in D_e and K will influence it by a factor of the square root of ten, which is slightly more than three.

Accounting for parameter variabilities along a flowpath

The very simple model presented above does not account for the variability of the entities along the flowpath. The width of the flowpath W can vary considerably, as can the matrix diffusivity, the sorption coefficients of the rock, the amounts of alteration material and gouge. Also the fracture aperture and the water velocity can vary.

Fortunately, it is possible to account for these variations in a simple manner. We follow a packet of water from inlet to outlet. The packet of water will encounter sections that are narrow and sections that are wide, it will be exposed to locally a large FWS as the flowpath widens and sections that have high and low diffusivities, sorption coefficients and different amounts of fracture filling materials etc. For such a complex flowpath the solution to the mass balance equations is [2]

$$c_{out} = c_{in} e^{-\lambda t} \operatorname{Erfc} \left[\frac{\psi(z)}{2\sqrt{t - \phi(z)}} \right] \quad (5)$$

Where

$$\phi(z) = \int_0^z \frac{R_a(z)}{u(z)} dz \quad (6)$$

and

$$\psi(z) = 2 \int_0^z \frac{W(z) \sqrt{D_e K_v}}{Q} dz \quad (7)$$

Equation (6) actually gives the total nuclide travel time in a pathway where there is no matrix diffusion and the retardation is caused by sorption on the filling and alteration materials. It is equivalent to Equation (2). For a sorbing nuclide the influence of the water residence is negligible. For short lived nuclides such as ^{137}Cs and ^{90}Sr the retardation due to sorption on the filling and alteration materials can be quite effective if their thicknesses are a mm or more. For long lived nuclides the matrix diffusion effects are often more important than the surface sorption.

The above treatment illustrates that variabilities along the flowpath in a variable aperture and variable width fracture can be accounted for. Equations (6) and (7) show that it is possible to account for variable rock properties by integration along the flow path. It is difficult to see how we will ever be able to obtain such detailed information. However, the equations show that if there is information available on the frequency distributions of K_v , D_e and W and if these distributions are not correlated, then it is correct to use the averages of W , $\sqrt{K_v}$ and $\sqrt{D_e}$ in Equation (7).

The above treatment can be extended to apply to the conditions in a stream tube with variable cross section A_o . Consider a streamtube, which is an imaginary tube within which the water flows. There is no transport of water or solutes through the wall of the tube and thus the water that enters one end will be the same water all along the tube and thus the water that enters cross section along the path. In the low conductivity "good" rock it will have a very large cross section and the water velocity will be low. As the water in the stream tube enters a high flow region its cross section will become small and the velocity will be large. Along the flowpath the water will move through the fractured rock and encounter varying amounts of local FWS. For a given location in the rock it has a specific FWS, a_r m^2 surface per m^3 rock volume. This entity may also vary along the flowpath. We deem it to be a rock property and assume that it can be measured.

Equation (7) can be rewritten to include a_r . It becomes

$$\psi(z) = \int_0^z \frac{a_r(z) \sqrt{D_e K_v}}{u_o(z)} dz = \int_0^z \frac{A_o(z) a_r(z) \sqrt{D_e K_v}}{Q} dz \quad (8)$$

$u_o(z)$ is the local flux, i.e. flowrate per cross sectional area. In the hydrology literature it is often called Darcy velocity. This entity is assessed by hydrological modelling in the form of fluxes along streamlines or in stream tubes. In this paper it is assumed to be known. Also the flowpath along which to integrate is assumed to be obtainable from the hydrological modeling.

These very simple equations give results that are very similar to those obtained by a very much more complex model, the Channel Network Model, CNM,[3,4]. In this model a multitude, hundreds of thousands, of independent channels form a three dimensional network. Each channel can have its own stochastically generated properties such as channel conductance, aperture, width etc. The more channels there are along the flowpath the more the solution resembles that of the

above equations. The above equations will thus well describe situations where a large number of channels have been encountered along the flowpath.

In this paper the influence of the variabilities and uncertainties in the sorption and diffusion properties and in the specific FWS, a_r , will be studied using the above equations.

Data, variabilities and uncertainties

By variability in data is meant the stochastic nature of the data due the differences between samples or measurement points and due to random measurement differences.

By uncertainties is meant here that there may be doubts if the method used to evaluate an entity from a measurement e.g. is correct and meaningful. An example is the hydraulic measurements using packer tests in boreholes to obtain some measure of a hydraulic conductivity. The methods may be reasonable or perhaps even correct for a homogeneous porous medium but it is questionable if they give meaningful results when applied to a fractured rock system.

The use of the notion of "Flow Wetted Surface" and the data that are available is another area that is questionable and uncertain. Another cause of uncertainty is if the samples or the experiment have used a representative piece of rock or location. This uncertainty arises in many field situations where the heterogeneity is very large and only a few measurements can be made.

In this paper it will be attempted to separate the variability from the uncertainty. As will be seen this is not easy and sometimes not possible.

Altered zone and fracture filling material

There is very little systematic information on the amounts and properties of fracture filling and alteration materials. Some recent observations at Äspö indicate that it is not uncommon at this site to find alteration materials with a thicknesses of 1 mm and porosity of 5 %. Also thicknesses of 5 mm and sometimes several tens of mm with porosities of 1-2 % have been observed by Tullborg [5]. Scoping calculations indicate that at least for the short lived nuclides ^{90}Sr and ^{137}Cs this can have strong retarding effect [6].

Matrix diffusion data

Recently Ohlsson and Neretnieks [7] compiled available data on matrix diffusion in crystalline rocks. The variability between different measurements for the same nuclides is very large. This variability is to a large extent due to the variability between different rock samples. This was noted already by Skagius and Neretnieks [8,9] in laboratory measurements and by Birgersson and Neretnieks [10] in field measurements. Porosities varied from less than 0.1 % up to 1 % with many data around 0.3-0.4 %. No formal statistical analysis of the data has been made but the variability in effective diffusivities was typically one to one and a half order of magnitude. It is deemed that most of the differences are due to stochastic variations and only a lesser part due to uncertainty.

Very seldom have measurements using different sorbing nuclides in the same rock sample been performed. This is because the experiments take a very long time. <for the sorbing nuclides a larger part is due to uncertainty because there is less data and the sorption mechanisms are not fully understood.

The effective diffusivity, D_e , which is used to assess the steady state flux of a diffusing species through a piece of the rock depends on the geometrical properties of pore network and on the diffusing species. It also depends on the diffusing species.

For a specie that does not interact with the mineral surfaces it is commonly assumed that a rock property called the formation factor F_f is an invariant. This entity multiplied by the diffusivity of the specie in bulk water will then give the effective diffusivity of the specie in the rock. F_f can be determined by diffusion experiments using e.g. tritiated water or methane or some other small non-charged molecule. This approach seems to be basically sound. Larger molecules will probably be influenced by steric hindrances as well, but for most nuclides of interest this is not a major concern.

There are two major additional effects that must be considered. One is the impact of the electrical double layer that can influence the flux of charged species. A high salinity will diminish the impact of the double layer whereas a low salinity will cause negatively charged (an-)ions to be repulsed from the naturally negatively charged mineral surfaces. This leads to what is called ion exclusion that prohibits the whole pore volume to transport anions. The opposite effect, a concentration of cat-ions at the surface leads to a higher flux in the pore because the concentration and thus the gradient will be larger than for an uncharged species with the same bulk concentration outside the piece of rock. The mobility in this more concentrated layer is called surface diffusion.

Figure 2 below illustrates the mean concentration profile of a non-charged, specie, an an-ion and a cat-ion outside and inside a rock sample.

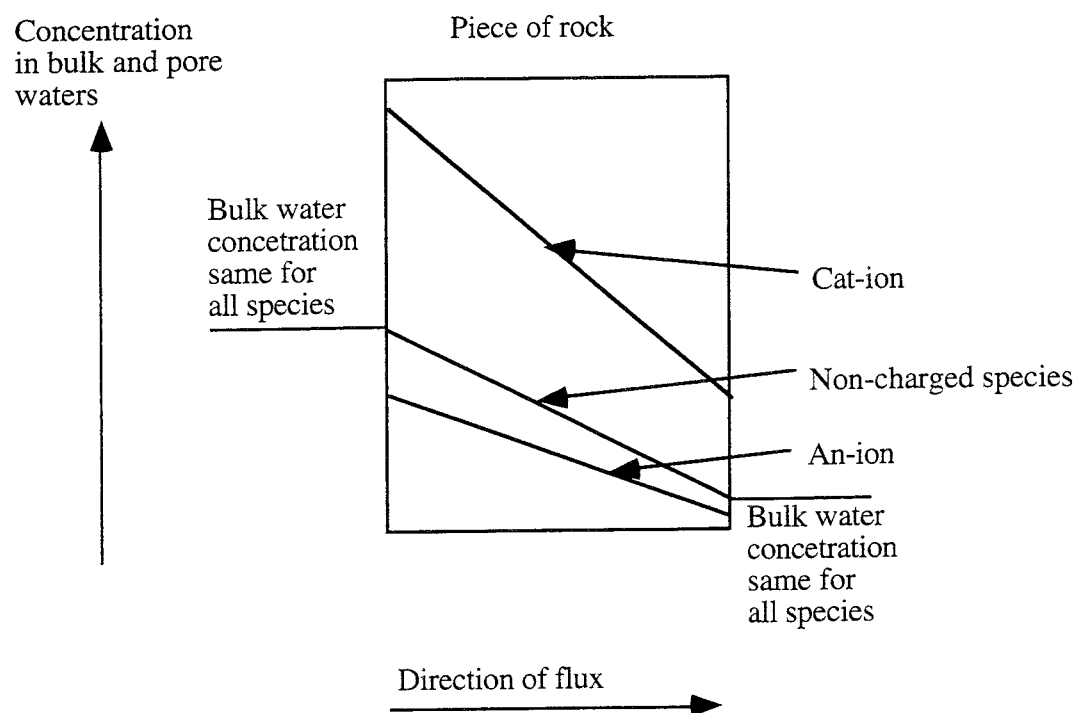


Figure 2. Concentrations in pore and bulk waters of neutral species, an-ions and cat-ions

These effects are clearly seen for diffusion in bentonite clay which has similar mineral surfaces as those of granites and gneisses.

A recent compilation of diffusivities in compacted bentonite has been made by Yu and Neretnieks [11]. For low salinity waters it was found that the an-ions iodide, chloride, carbonate and pertechnetate had one to three orders of magnitude smaller effective diffusivities due to ion exclusion than comparable non-influenced nuclides. Surface diffusion was found for cesium, protactinium, radium and strontium. One to two orders of magnitude increase in the effective diffusivity was noted. Other cationic nuclides were not found to have a clearly noticeable surface diffusion component. The ion exclusion as well as the surface diffusion effects disappeared at

higher ionic strength pore waters. High ionic strength or high salinity in this context is a concentration of 10 000 mg/l or more. With low salinity waters have salt concentrations around 1 000 mg/l or less. These are rather arbitrary values but they were chosen because a group of important data that were used had approximately these salinities. There is a definite need to study these effects further.

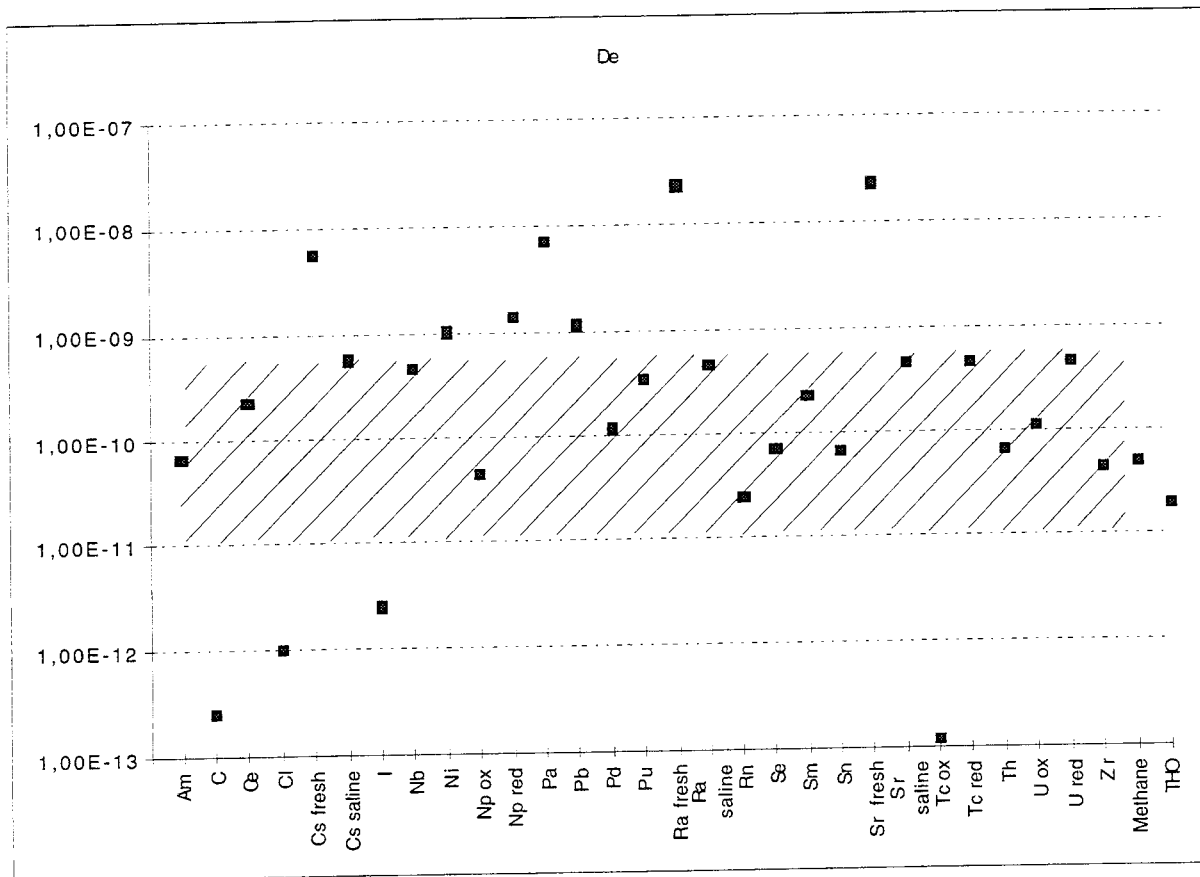


Figure 3. Effective diffusivities. Dotted lines at 10^{-9} and 10^{-11} indicate the span where neither ion exclusion effects nor surface diffusion effects are important.

Figure 3 shows a compilation of the diffusivity data in compacted bentonite. The variability of these data are nearly two orders of magnitude if one excludes the nuclides that either have ion exclusion or surface diffusion effects. A visual estimate of the standard deviation of these data is $\sigma=0.5$ on the $^{10}\log$ scale. Part of this variance is due to the differences between the nuclides so the variability for a given nuclide is probably less.

It has not been possible to make a similar compilation of diffusion data for the different nuclides in granite. In the earlier cited papers [8,9,10] where electrical conductivity measurements and diffusion measurements using iodide and an-ionic dyes were used similar variabilities were found for different rock pieces. A visual estimate of the standard deviation of these data is $\sigma=0.2-0.3$ in the $^{10}\log$ scale.

For use in a performance assessment for a known site we have recommended the following procedure[12].

Site specific data on the formation factor are determined for such a number of samples that a mean value and a standard deviation can be obtained. The formation factor can be measured using electrical conductivity measurements or direct steady state measurements of the diffusive flux through the samples. This is done for the different rock types that may be encountered by the nuclides. The water chemistry and the ionic strength is determined at the different locations. A table showing the calculation procedure can then be constructed. In the second column the bulk diffusivities of the nuclide is entered. This multiplied by the formation factor will give the effective diffusivity in column 4 if there are no ion exclusion or surface diffusion effects. This is for high ionic strength. For low ionic strength waters these effects must be accounted for and we suggest a decrease of a factor ten for those nuclides that are subject to ion exclusion and an increase by a factor of ten for those nuclides that are known to exhibit surface diffusion. This factor is given in column 7. There is however, a large uncertainty in the factor 10. It could well be a factor 100 as seen in Figure 3 above. The effective diffusivity is then entered in column 8.

For use in an ongoing safety analysis performed by SKB in Sweden we have suggested the use of formation factor of $4.2 \cdot 10^{-5}$. The effective diffusivities for the nuclides that will be used in the sample calculations are shown in Table 1.

Specie	D_w	K_d	$D_e = D_w \cdot F_f$	$D_a = D_e / (\epsilon + \rho K_d)$	K_d	f	$D_e = D_w \cdot F_f \cdot f$	$D_a = D_e / (\epsilon + \rho K_d)$
				High ionic strength			Low ionic strength	
HTO						1		
IE						0.1		
SD						10		
other						1		

where IE denotes "ion exclusion" i.e. anions, SD denotes surface diffusion i.e. ion exchanging cations.

Table 1. Calculation schedule to obtain D_e and D_a when D_w , K_d and F_f are known

Sorption data

Carbol and Engkvist ([13] have recently compiled sorption data for a large number of nuclides for the same types of waters as were used to estimate the diffusion data. The waters are reducing, with an Eh <200 mV and a pH ≥ 7. They are typical of the Gideå (low salinity) and Äspö (high salinity) site respectively.

Their data show that the sorption coefficient of mono- and di-valent cations decrease by a factor 5 to 10 in the high salinity waters compared to low salinity waters. They are in the range 0.1 m³/kg for Co(II), Ni(II), Pd(II), Ra(II) and Cd(II), 0.01 m³/kg for Sr(II) and 0.5 m³/kg for Cs(I) and Ag(I) for the low salinity waters. For tri- to penta-valent nuclides the sorption coefficients are much higher and are not as strongly influenced by the salinity of the water. The sorption coefficients range from 1-5 m³/kg with the higher value, 5, for the actinides Th(IV), U(IV), Np(IV) and Pu(IV), the value 3 for Ac(III), Am(III), Cm(III), the value 2 for Sm(III), Eu(III) and Ho(III). Zr(IV), Nb(V), Tc(IV) and Pa(IV,V) have $K_d = 1$ m³/kg. The anions I, Cl have $K_d = 0$ m³/kg. Carbonate has a K_d of 0.001 m³/kg.

The uncertainty interval has been estimated to be a factor between 4 and 50, for the different nuclides. A typical value for the uncertainty interval is a factor 10 between the highest and the lowest value in the span.

Uncertainty interval and distribution function

In very few instances a formal statistical analysis has been presented and there is little information on the form of distribution function and on the standard deviation of the distribution.

For illustration purposes it is assumed in this paper that the distribution is log-normal. The term uncertainty interval is not defined. Interviews with several colleagues working in this and related fields indicate that most of them agree to the following statement. "The uncertainty interval means that very few data points will be found outside the interval". When pressed somewhat more on how many points would be permitted to lie outside the interval the answers range from about 1 to no more than 10 %.

If 5 % lie outside the interval this will mean that the interval spans four standard deviations on the logarithmic scale. Thus for an interval of a factor of ten the standard deviation is $\sigma = 0.25$ on the ¹⁰log scale. For intervals 4, 20 and 50 they are 0.15, 0.33 and 0.43 respectively.

Often the proposed uncertainty intervals are asymmetric around the suggested value. This does not seem to indicate that the distribution is asymmetric, only that the use of the expressions is imprecise and blunt. It is unclear in this case what is uncertainty and what is variability.

In the examples a value of $\sigma=0.3$ will be used for D_e and K_d and an uncertainty interval of 3 up and down.

Specific flow wetted surface and conductivity distribution

The entity that is most poorly known is the flow wetted surface. There is no well tested and universally accepted method to determine this entity. There are also very few field measurements that have been used to determine the FWS in the form of the specific FWS a_r .

Abelin et al. [14] used the non recovery of a part of the tracer mass injected in a long time experiment in Stripa to estimate the FWS. Values ranges from 0.5 to more than 20 m^2/m^3 rock for five different flowpaths if virgin rock matrix properties were assumed. If the matrix diffusion had mostly taken place in somewhat altered rock adjacent to the fracture surfaces the values ranged from 0.2 to 2.7 m^2/m^3 .

Birgersson et al. [15] obtained a_r values ranging from 5 to 24 m^2/m^3 from the non-recovery of tracers in experiments in a fracture zone in the Stripa experimental mine.

Bore hole fluid injection measurements in the Äspö rock laboratory of fracture frequencies and conductivities in the observed fracture zones gave a value of a_r 1.2 m^2/m^3 . A $\sigma = 1.6$ on the ¹⁰log scale was found for the local conductivities. Gylling et al. [16] and Moreno and Neretnieks [4] analysed data from three different sites and found σ for the conductivities to be about 1.6-2-4. This shows that there will be a very large range of flowrates in different locations. It was found in simulation using the Channel Network Model that the standard deviation for local flowrates is approximately 0.85 of that for the conductivities.

From a geometrical point of view the flow wetted surface can be approximately estimated from the flowing fracture frequency in boreholes by the following expression [17].

$$a_r = \frac{\pi^2}{2} \cdot \frac{1}{H} \quad (9)$$

H is the average distance between conducting fractures in the borehole. There are two difficulties in using this method. One is that of cut off in the measurements. The low flow channels are not

detected. Moreno and Neretnieks [4] try to compensate for this by extrapolating the observed higher flow data to the low flow region by assuming a log normal distribution of flow rates.

The other difficulty is that the spacial resolution in the measurements is often very coarse. Commonly packer distances of more than 10 m are used in boreholes in the good rock. This means that one can in principle know if there is more than one conducting feature in that interval, not how many more. This may seriously underestimate the FWS.

There are not enough experiments where the FWS has been obtained using the geometric method and other methods that observe the impact of the FWS on the retardation or non recovery of tracers. In the few instances where this has been attempted the differences seem to be about an order of magnitude.

The data on the magnitude of the FWS are thus very uncertain. The variability in the form of the standard deviation of the local FWS/flowrate is perhaps better known. This information is at present used in the Channel Network Model. The simulation results for a tracer are much more influenced by the total FWS encountered along the flowpath than by the magnitude of the standard deviation.

For a standard deviation off the 10 log scale 1.6, the difference between the low and high values is more than one million. The low and high values are then taken at -2σ and $+2\sigma$. This span contains 95 % of all values.

In the examples a central value of $a_r = 0.4 \text{ m}^2/\text{m}^3$ will be used. A low value of 0.1 will also be used to illustrate the influence of the FWS on transport.

Other data

A water flux in the rock at repository depth of $0.1 \text{ l}/\text{m}^2/\text{year}$ is used. This would be obtained in a rock with a hydraulic conductivity of 10^{-9} m/s and a hydraulic gradient of 0.3%.

For use in Equation 1 for the single fracture description the fracture frequency is 1 per 5 meters for the chosen specific FWS. This gives a flowrate Q of $0.5 \text{ l}/\text{year}$ in a fracture 1 m wide.

In the example we study a fracture or streamtube somewhere near a degraded canister and assume it is 50 m long before reaching a region with different properties, e.g. a fracture zone. This would illustrate a near field fracture. The water may thereafter emerge into a region with high flux.

The fracture aperture is assumed to be 0.01 mm. This choice is not critical. Even very large variations do not influence the results. These values give a water residence time of 0.1 year. If the fracture aperture would be 1 mm for a loosely gouge filled fracture the residence time would be on the order of 10 years.

Summary of data used in the examples

Nuclide	$D_e \cdot 10^{13} \text{ m}^2/\text{s}$	$K_d \text{ m}^3/\text{kg}$	$^{10}\text{Log } \sigma_D^*$	$^{10}\text{Log } \sigma_K^*$
I Low salinity	0.08	0	0.3	0.3
I high salinity	0.8	0	0.3	0.3
Cs Low salinity	4	0.5	0.3	0.3
Cs high salinity	0.4	0.05	0.3	0.3
Pu and Np both salinities	0.4	5	0.3	0.3

*Standard deviations are examples for use in the examples. They are not based on statistical analysis

In the sample calculations it is assumed that the uncertainties in diffusivity and sorption data is a factor of three up and down.

	Entity	Value	Uncertainty span*
a _r	Specific flow wetted surface in good rock	0.4 m ² /m ³ rock	0.1-5
2b	Fracture aperture	0.01 mm	0.01-1α
L	Flow path length	50 m	
u _o	Water flux at repository depth	0.1 l/ m ²	-
d	Thickness of porous alteration material	0 mm	0-1#
	Thickness of porous gouge material in fracture	0 mm	0-2b#
ρ	Density of rock	2700 kg/ m ³	none

*Subjective values. This spans a conductive fracture distance of 1-50 meters

α Subjective values

Sorption coefficients are taken to be as for food rock

Nuclide	Halflife of nuclides years
¹³⁷ Cs	30
²³⁹ Pu	24 400
²³⁷ Np	2.1 * 10 ⁶

Table 2. Effective diffusivities, sorption coefficients and estimated standard deviations of some nuclides. Reducing water.

Influence of variability along the flowpath

It was demonstrated earlier, Equation (7), that variations along the flowpath can be accounted for as shown in the expression $\int_0^z \frac{A_o(z)a_r(z)\sqrt{D_e K_v}}{Q} dz$. In the streamtube, or we prefer to think of the flow as taking place in a succession of channels, Equation (8), the flowrate Q is the same all along the flowpath. It may be noted that $\int_0^z A_o(z)a_r(z) dz$ is the total FWS along the flowpath contacted by the water flowrate Q. The flowpath may have traversed good rock, fracture zones and other features. In principle it can be assessed by following the streamlines obtained from the hydrological modelling and from knowledge of specific FWS, a_r in the different regions. Provided there is no correlation between a_r and the sorption coefficient and the effective diffusivity the integration can be made as just described. At present there is no information available on any correlation between the entities and thus this assumption is necessary at present. There is also no known correlation between D_e and K_v. The integration can also here be made independently.

The average of the square root of D_e and K_v differs from the square root of the average of the entity. For the assumed log normal distribution and the value of σ= 0.3 the difference is only a few percent. However, the expected value of this entity is nearly three times that of the logarithmic mean in the distribution. There seems therefore to be a need to analyse the form of the distributions and what is actually meant by the values that are recommended for the different entities.

Although a part of the variability in the data are caused by natural stochastic variations that will be evened out along the pathway, some sample calculations will be made assuming that the variability is due to uncertainties.

Sample calculations

Influence of uncertainties in sorption, diffusion and flow wetted surface

For ^{137}Cs the effluent concentration after 50 m travel distance has decayed to insignificance, $\ll 10^{-9}$, for the central case. Only when all the variables are given the most unfavourable values simultaneously the concentration will become noticeable and be at most a few % of that at the inlet of the streamtube. Then the travel distance is 50 m, the diffusivity and the sorption coefficients are a factor 3 lower than the central value, FWS is at the lowest value and the water is saline. For a non saline water even the worst case combination will give a totally insignificant concentration.

For ^{239}Pu the effluent concentrations will decay to insignificance for the central case. A decrease of both the sorption coefficient and the diffusivity by the uncertainty factor 3 still gives no significant concentration. The same applies if only the FWS is given its lowest value. An increase

of the matrix diffusion group $\int_0^z \frac{A_o(z)a_r(z)\sqrt{D_e K_v}}{Q} dz$ by a factor of 10 gives noticeable concentrations. Any combination of variables that give factor of 10 increase will have the same effect. This includes also an increase of the flowrate Q .

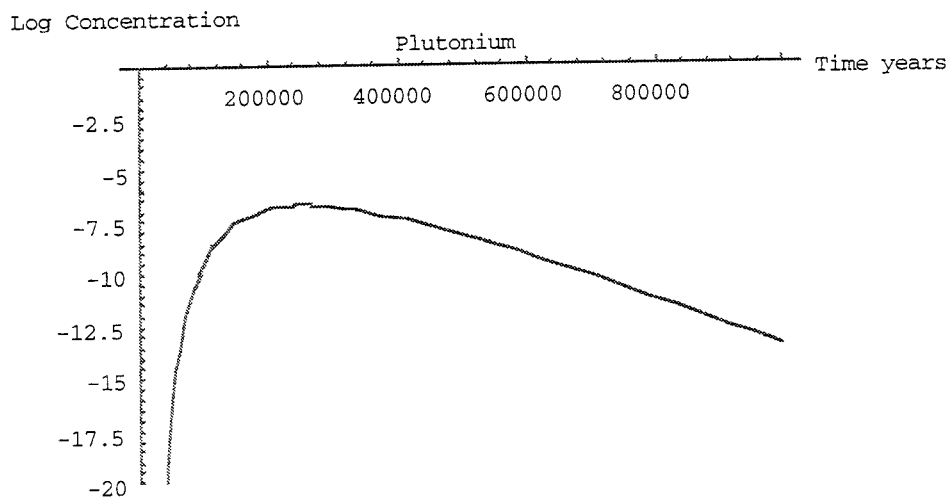


Figure 4. Outlet concentration for plutonium for a combined uncertainty of a factor 10 increase in matrix diffusion group.

For ^{237}Np the breakthrough curves are shown in Figure 5 for a factor of 3, lower curve, and factor of 10, upper curve, uncertainty in the group $\int_0^z \frac{A_o(z)a_r(z)\sqrt{D_e K_v}}{Q} dz$,

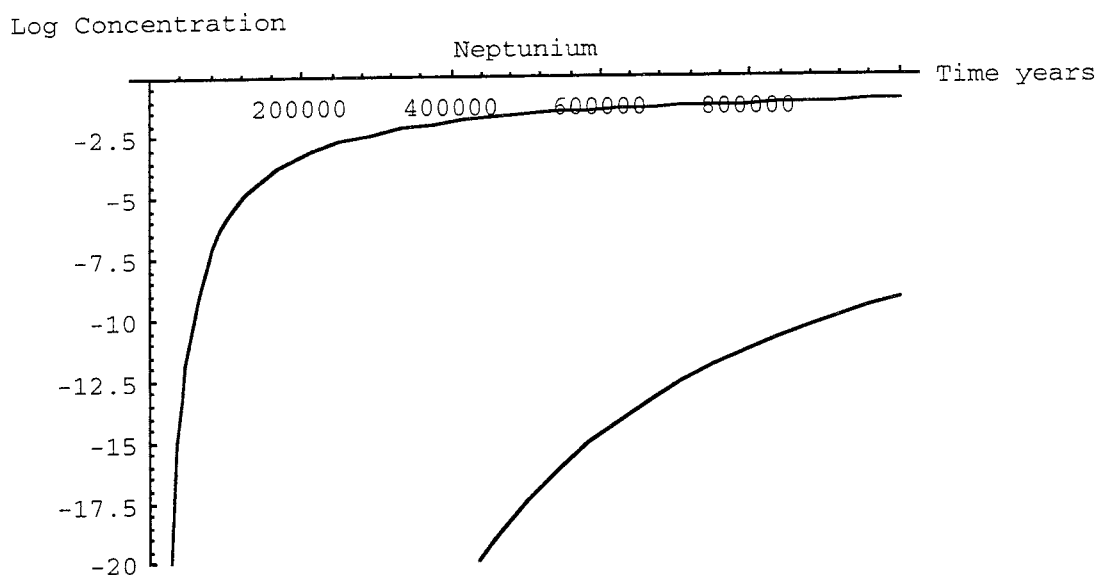


Figure 5. Outlet concentration for Neptunium. Upper curve factor 10, lower curve factor 3 uncertainty.

Influence of uncertainties of the presence of alteration and filling materials

Below is an example of the impact of fracture filling and fracture alteration material. The fracture filling material fills out all the fracture aperture, 0.01 mm and the alteration material is 1 mm thick. Both have the same volumetric sorption coefficients as the good rock and are instantly equilibrated. It is the fracture alteration material that causes 99% of the effect in this case.

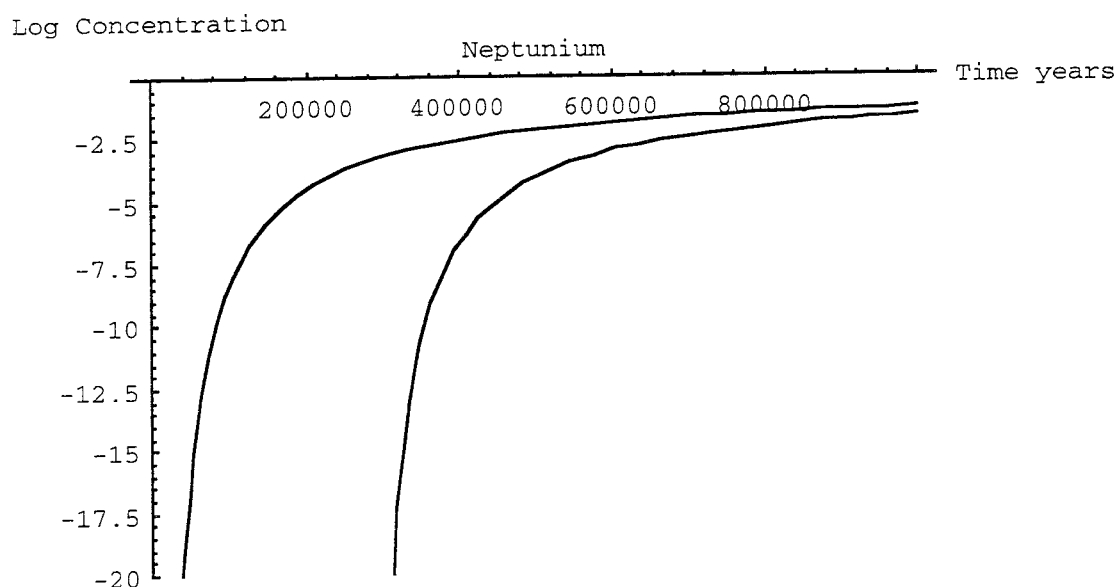


Figure 6. Outlet concentration for neptunium. Both curves factor 10 uncertainty for the matrix diffusion group. Right hand curve has 0.01 mm fracture filling and 1 mm alteration material.

Discussion and conclusions

There is at present very little information on the stochastic properties of most of the entities that influence the migration of radionuclides in fractured rock. Much of the observed variability in reported data on sorption and diffusion must at present be assumed to be uncertain. This may be due to systematic differences between different measurement techniques. The stochastic variability of these entities can be accounted for by integrating the properties along the flowpath as has been demonstrated.

The impact of systematic errors and uncertainties in these entities is somewhat diminished because they enter the expression by a square root. The local flowrate of water, Q , per Flow Wetted Surface, FWS, has a stronger impact. Furthermore this entity is much less studied and the magnitude of this entity is more uncertain. Fortunately theory indicates that the variability of this entity along the flowpath can be accounted for in a simple manner.

The by far most uncertain entity is the magnitude of the specific flow wetted surface. It also has the highest impact. It is recommended that this entity be studied further and that new techniques are developed to measure flowrate distributions in boreholes with a very high spatial resolution. Present day practice of using packer distances of 10 or more meters give data that can strongly underestimate the FWS.

In fractured rocks with fracture filling and porous fracture alteration materials of more than a fraction of a mm, such material can significantly retard the nuclides in addition to the matrix diffusion effect.

Notation

a_r	Specific flow wetted surface	m^2/m^3 rock
A_o	Cross section of stream tube	m^2
b	Half aperture of fracture	m
c	Concentration	mol/m^3
d	Thickness of alteration material	m
D_a	Apparent diffusivity	m^2/s
D_e	Effective diffusivity	m^2/s
F_f	Formation factor	-
H	Average distance between conducting fractures in borehole	m
K_a	Surface sorption coefficient for rock surface	m
K_d	Mass sorption coefficient for rock	m^3/kg
K_{df}	Mass sorption coefficient for gouge	m^3/kg
K_{da}	Mass sorption coefficient for alteration material	m^3/kg
K_v	Volumetric sorption coefficient for rock	m^3/m^3
K_{vf}	Volumetric sorption coefficient for gouge	m^3/m^3
K_{va}	Volumetric sorption coefficient for alteration material	m^3/m^3
L	Channel length	m
Q	Flowrate of water	m^3/s
R_a	Retardation factor	-
t	Time	s
t_o	Travel time of nuclide for plug flow	s
t_w	Travel time of water for plug flow	s
u_o	Water flux	$m^3/m^3 \cdot s$
W	Channel width	m
z	Flow path length	m

Literature

1. Neretnieks, I., Diffusion in the rock matrix: An important factor in radionuclide retardation? *J. Geophys. Res.* 85, p 4379-4397, 1980.
2. Carslaw H.S., Jaeger J.C. *Conduction of heat in solids*, 2nd ed. Oxford University press 1959
3. Moreno L. and I. Neretnieks, Flow and nuclide transport in fractured media. The importance of the flow wetted surface for radionuclide migration, In J.I. Kim and G. de Marsily (Editors), *Chemistry and Migration of Actinides and Fission Products*, *Journal of Contaminant Hydrology*, Vol 13, 49-71, 1993a
4. Moreno L. and I. Neretnieks, Fluid flow and solute transport in a network of channels. *J. Contaminant Hydrology* 14, 163- 192, 1993b
5. Tullborg Eva-Lena, Terralogica. Personal communication March 1997.
6. Neretnieks I. A note on the potential impact of fracture filling and alteration materials. Report prepared for SKB. Dept. Chemical Engineering and Technology, Royal Institute of technology, Stockholm Sweden. Feb 1997.
7. Ohlsson Y. and Neretnieks I. Literature Survey of Matrix Diffusion Theory and of Experiments and Data Including Natural Analogues. SKB TR 95-12
8. Skagius, K., and I. Neretnieks, Porosities of and diffusivities of some non-sorbing species in crystalline rocks. *Water Resources Res.* 22, p 389-398, 1986a.
9. Skagius, K., and I. Neretnieks, Diffusivity measurements and electrical resistivity measurements in rock samples under mechanical stress. *Water Resources Research* 22 (4), p 570-580, 1986b.
10. Birgersson, L., and I. Neretnieks, Diffusion in the matrix of granitic rock. Field test in the Stripa mine. *Water Resources Research* 26 p 2833-2841, 1990
11. Ji-Wei Yu and Neretnieks I., Diffusion and sorption properties of radionuclides in compacted bentonite, SKB TR 97-12
12. Ohlsson Y. and Neretnieks I. Diffusion data in granite, Recommended values SKB TR 97-xx, this report
13. Carbol P., Engkvist I. Compilation of radionuclide sorption coefficients for performance assessment. SKB TR 97-xx, in print
14. Abelin H., Birgersson L., Moreno L., Widén H., Ågren T., Neretnieks I. A Large Scale Flow and Tracer Experiment in Granite II. Results and interpretation. *Water Resources Research*, 27(12), p 3119-3135, 1991
15. Birgersson, L., L. Moreno, I. Neretnieks, H. Widén, and T. Ågren, A tracer migration experiment in a small fracture zone in granite, *Water Resour. Res.*, 29(12), 3867-3878, 1993
16. Gylling B., Moreno L., and Neretnieks I. A channel-network-model for radionuclide transport in fractured rock.- Testing against field data, Paper presented at the MRS Meeting on Scientific basis for nuclear waste management XVIII, Kyoto, Oct 24-26, 1994, *Proceedings* vol 353, p 395-402
17. Gylling B., Moreno L., and Neretnieks I. Modelling of solute transport in fractured media using a channel network model. Submitted to ICM 1997.

List of SKB reports

Annual Reports

1977-78

TR 121

KBS Technical Reports 1 – 120

Summaries

Stockholm, May 1979

1979

TR 79-28

The KBS Annual Report 1979

KBS Technical Reports 79-01 – 79-27

Summaries

Stockholm, March 1980

1980

TR 80-26

The KBS Annual Report 1980

KBS Technical Reports 80-01 – 80-25

Summaries

Stockholm, March 1981

1981

TR 81-17

The KBS Annual Report 1981

KBS Technical Reports 81-01 – 81-16

Summaries

Stockholm, April 1982

1982

TR 82-28

The KBS Annual Report 1982

KBS Technical Reports 82-01 – 82-27

Summaries

Stockholm, July 1983

1983

TR 83-77

The KBS Annual Report 1983

KBS Technical Reports 83-01 – 83-76

Summaries

Stockholm, June 1984

1984

TR 85-01

Annual Research and Development Report 1984

Including Summaries of Technical Reports Issued during 1984. (Technical Reports 84-01 – 84-19)

Stockholm, June 1985

1985

TR 85-20

Annual Research and Development Report 1985

Including Summaries of Technical Reports Issued during 1985. (Technical Reports 85-01 – 85-19)

Stockholm, May 1986

1986

TR 86-31

SKB Annual Report 1986

Including Summaries of Technical Reports Issued during 1986

Stockholm, May 1987

1987

TR 87-33

SKB Annual Report 1987

Including Summaries of Technical Reports Issued during 1987

Stockholm, May 1988

1988

TR 88-32

SKB Annual Report 1988

Including Summaries of Technical Reports Issued during 1988

Stockholm, May 1989

1989

TR 89-40

SKB Annual Report 1989

Including Summaries of Technical Reports Issued during 1989

Stockholm, May 1990

1990

TR 90-46

SKB Annual Report 1990

Including Summaries of Technical Reports Issued during 1990

Stockholm, May 1991

1991

TR 91-64

SKB Annual Report 1991

Including Summaries of Technical Reports Issued during 1991

Stockholm, April 1992

1992

TR 92-46

SKB Annual Report 1992

Including Summaries of Technical Reports Issued during 1992

Stockholm, May 1993

1993

TR 93-34

SKB Annual Report 1993

Including Summaries of Technical Reports Issued during 1993

Stockholm, May 1994

1994

TR 94-33

SKB Annual Report 1994

Including Summaries of Technical Reports Issued during 1994

Stockholm, May 1995

1995

TR 95-37

SKB Annual Report 1995

Including Summaries of Technical Reports Issued during 1995

Stockholm, May 1996

1996

TR 96-25

SKB Annual Report 1996

Including Summaries of Technical Reports Issued during 1996

Stockholm, May 1997

List of SKB Technical Reports 1997

TR 97-01

Retention mechanisms and the flow wetted surface – implications for safety analysis

Mark Elert

Kemakta Konsult AB

February 1997

TR 97-02

Äspö HRL – Geoscientific evaluation 1997/1. Overview of site characterization 1986–1995

Roy Stanfors¹, Mikael Erlström²,

Ingemar Markström³

¹ RS Consulting, Lund

² SGU, Lund

³ Sydkraft Konsult, Malmö

March 1997

TR 97-03

Äspö HRL – Geoscientific evaluation 1997/2. Results from pre-investigations and detailed site characterization. Summary report

Ingvar Rhén (ed.)¹, Göran Bäckblom (ed.)², Gunnar Gustafson³, Roy Stanfors⁴, Peter Wikberg²

¹ VBB Viak, Göteborg

² SKB, Stockholm

³ VBB Viak/CTH, Göteborg

⁴ RS Consulting, Lund

May 1997

TR 97-04

Äspö HRL – Geoscientific evaluation 1997/3. Results from pre-investigations and detailed site characterization. Comparison of predictions and observations. Geology and mechanical stability

Roy Stanfors¹, Pär Olsson², Håkan Stille³

¹ RS Consulting, Lund

² Skanska, Stockholm

³ KTH, Stockholm

May 1997

TR 97-05

Äspö HRL – Geoscientific evaluation 1997/4. Results from pre-investigations and detailed site characterization. Comparison of predictions and observations. Hydrogeology, groundwater chemistry and transport of solutes

Ingvar Rhén¹, Gunnar Gustafson², Peter Wikberg³

¹ VBB Viak, Göteborg

² VBB Viak/CTH, Göteborg

³ SKB, Stockholm

June 1997

TR 97-06

Äspö HRL – Geoscientific evaluation 1997/5. Models based on site characterization 1986–1995

Ingvar Rhén (ed.)¹, Gunnar Gustafson²,

Roy Stanfors⁴, Peter Wikberg⁴

¹ VBB Viak, Göteborg

² VBB Viak/CTH, Göteborg

³ RS Consulting, Lund

⁴ SKB, Stockholm

October 1997

TR 97-07

A methodology to estimate earthquake effects on fractures intersecting canister holes

Paul La Pointe, Peter Wallmann, Andrew Thomas,

Sven Follin

Golder Associates Inc.

March 1997

TR 97-08

Äspö Hard Rock Laboratory Annual Report 1996

SKB

April 1997

TR 97-09

A regional analysis of groundwater flow and salinity distribution in the Äspö area

Urban Svensson

Computer-aided Fluid Engineering AB

May 1997

TR 97-10

On the flow of groundwater in closed tunnels. Generic hydrogeological modelling of nuclear waste repository, SFL 3–5

Johan G Holmén
Uppsala University/Golder Associates AB
June 1997

TR 97-16

Groundwater flow through a natural fracture. Flow experiments and numerical modelling

Erik Larsson
Dept. of Geology, Chalmers University of Technology, Göteborg, Sweden
September 1997

TR 97-11

Analysis of radioactive corrosion test specimens by means of ICP-MS. Comparison with earlier methods

R S Forsyth
Forsyth Consulting
July 1997

TR 97-17

A site scale analysis of groundwater flow and salinity distribution in the Äspö area

Urban Svensson
Computer-aided Fluid Engineering AB
October 1997

TR 97-12

Diffusion and sorption properties of radionuclides in compacted bentonite

Ji-Wei Yu, Ivars Neretnieks
Dept. of Chemical Engineering and Technology, Chemical Engineering, Royal Institute of Technology, Stockholm, Sweden
July 1997

TR 97-18

Release of segregated nuclides from spent fuel

L H Johnson, J C Tait
AECL, Whiteshell Laboratories, Pinawa, Manitoba, Canada
October 1997

TR 97-13

Spent nuclear fuel – how dangerous is it? A report from the project "Description of risk"

Allan Hedin
Swedish Nuclear Fuel and Waste Management Co,
Stockholm, Sweden
March 1997

TR 97-19

Assessment of a spent fuel disposal canister. Assessment studies for a copper canister with cast steel inner component

Alex E Bond, Andrew R Hoch, Gareth D Jones, Aleks J Tomczyk, Richard M Wiggan, William J Worraker
AEA Technology, Harwell, UK
May 1997

TR 97-14

Water exchange estimates derived from forcing for the hydraulically coupled basins surrounding Äspö island and adjacent coastal water

Anders Engqvist
A & I Engqvist Konsult HB, Vaxholm, Sweden
August 1997

TR 97-15

Dissolution studies of synthetic soddyite and uranophane

Ignasi Casas¹, Isabel Pérez¹, Elena Torrero¹, Jordi Bruno², Esther Cera², Lara Duro²
¹ Dept. of Chemical Engineering, UPC
² QuantiSci SL
September 1997