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SR-Can

Data and uncertainty assessment

Matrix diffusivity and porosity in situ

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Executive summary

The molecular diffusion in microscopically small pores of crystalline rock matrices allows radionuclides to enter the stagnant matrix water. The water volume in the matrix is orders of magnitude larger than the mobile water in the flowing fractures. This effect will retard the nuclide migration. A further retardation effect is due to the sorption of the sorbing nuclides on the interior surfaces of the matrix. The internal surfaces are on the order of 100,000 times larger than the surfaces of the flowing fractures.

It is therefore important to ensure that matrix diffusion will take place in intact rock under the stresses which prevail at repository depths.

Laboratory experiments on drill cores have shown that the matrix porosity is connected over distances of at least several tens of centimetres for un-stressed samples. Samples that have been re-stressed to repository depth stresses have also been found to have connected porosity. Diffusivities in re-stressed samples were found to be up to three times lower than in un-stressed samples. This was found both using actual through-diffusion experiments as well as electrical conductivity measurements.

Diffusion experiments are very time consuming, costly and difficult to make in deeply lying rock under undisturbed conditions. An alternative way of measuring the mobility of charged species in porous rock is by using electric current to carry the ions. For bulk water this has a long-standing theoretical basis and is used to determine ion diffusivities. The method has also long been used in laboratory investigation to measure diffusivities in porous rocks. It has been shown to give electrical conductivities that agree well with that expected from diffusivity measurements. A number of tests have been made with AC (alternating current) and DC (direct current) to measure resistivities as well as using DC current to conduct ions through rock samples. These tests clearly confirm that the different methods give comparable results and show beyond doubt that the electrical conductivity method using AC gives the expected information on transport properties (diffusivities) of the pores of crystalline rocks.

The electrical conductivity method is much faster and can be used to measure large samples. It has recently been adopted for use in deep boreholes. Tens of thousands of measurements have been made at Simpevarp, Laxemar and Forsmark (Swedish sites) at depths of more than 1,000 m.

The results of these measurements form the basis for our proposed diffusion values to be used in Performance Assessment (PA) for the candidate sites (Performance Assessment (PA) used in this report is synonymous to Safety Assessment (SA) sometimes used by other authors). The in situ data are obtained essentially in undisturbed rock and have not been subject to either stress release or disturbances due to sample preparation. The small disturbance nearest the borehole is negligible because the electrical conductivity method samples rock extending to more than a metre from the borehole.

A large number of laboratory measurements have been analysed in order to ensure that other effects that cannot be controlled in the in situ measurements do not influence the down-hole data. No unexpected effects have been found.

Rock matrix porosity in situ measurements are extremely scarce. However, it has been possible to use some of the in situ measurements to estimate the increase in porosity when taking up rock from its natural environment to the laboratory. One example of such an investigation is briefly discussed to show how this was done.

In one in situ diffusion experiment performed at a depth of 360 m in granitic rock in Sweden the experimental conditions were such that it was ensured that any rock stress changes due to the presence of the drift and the presence of the borehole were avoided. The rock was thus subject to “virgin stress”. Over-coring after exposure to tracers for three and a half years and detailed sampling and measurement of the concentrations of the three tracers revealed that they had penetrated up to 40 cm into the rock matrix. This implies that the micropore network is connected over at least such distances. The observed distances agree with what was expected. It was found that the concentrations of the tracer in the samples nearest the thin injection hole were always about 30% of the injection concentration if the porosity of the samples as measured in the laboratory were used. Had the rock in situ been compressed and decreased the porosity by about a factor of three, when excavated a decrease to about 30% would result.

To summarise

The in situ measurements and observations clearly show that the micropore network in the matrix of the rock is connected over a large distance, more than a metre, and allows movement of dissolved ions. The electrical conductivity method gives diffusivities comparable to diffusivities obtained by direct diffusion measurements in laboratory tests. Laboratory measurements give much higher diffusivities than in situ tests. The in situ data are more appropriate for use in PA modelling.

Preface

It is our understanding that the main use of the data is for use in PA modelling of radionuclide migration through the bedrock from a potentially leaking repository.

The data on diffusive properties of the porous rock matrix and the porosity of the rock is then needed. These properties should be relevant to the rock under undisturbed conditions at the relevant depths. This implies that the data should be assessed for the prevailing rock stress, the water chemistry in the stagnant water in the micropores of the rock and other in situ conditions.

We will rely heavily on the large number of electrical conductivity measurements performed in deep boreholes in the candidate sites.

We will show that electrical conductivity measurements directly give information on the diffusion properties of the porous rock matrix. This is done by

- Theoretical arguments based on long-standing knowledge that ion mobility in an electric field can be used to determine the random movement of ions quantified as *diffusivity*.
- Direct comparison of transport by diffusion through rock samples in the laboratory and electrical conduction in an AC field. The methods give very similar results. The AC method is the method used in the field measurements.
- Direct comparison of AC and DC measurements, which gives the same results in the laboratory.
- Direct comparison between the rate of diffusion through a rock sample and the rate of ion transport through the sample in an electric field.

We will argue that the above issues convincingly show that the AC method applied in boreholes gives correct results for rock in situ.

The in situ data will therefore be the basis for suggesting data to use for PA. The causes of uncertainties in the field data will receive most of our attention.

A large number of laboratory and a few in situ measurements of diffusivities and porosities will be presented and used as supplementary information and for assessing other uncertainties that the field measurements cannot resolve. Most of the laboratory data are from Swedish and Finnish sites and rocks, which are deemed to have the largest relevance for the Swedish candidate sites. However, some investigations from other countries are also discussed briefly when they supply information that has not been found in the Scandinavian investigations.

The reader familiar with both PA modelling and matrix diffusion issues can, after having convinced him/herself that our basic method of in situ measurements is correct, go directly to Chapter 5 where the data of the in situ measurements are presented. In this chapter also our proposed values and stochastic uncertainties in the form of standard deviations of the measurements are presented.

In Chapter 4 the main sources affecting the uncertainties of the in situ measurements are discussed. The main uncertainty is due to the assumption that the water in the micropores of the rock matrix has the same electrical conductivity as that extracted from nearby fractures in the borehole. Other causes are also discussed. The information in Chapters 4 and 5 should give sufficient information for choosing data for use in the PA modelling.

It may help a reader not familiar in detail with how matrix diffusion is used in PA modelling to start with Chapter 2 on how the data is to be used in the PA models of SR-Can. Here it is also discussed how uncertainties and variabilities in data can impact the uncertainties in nuclide travel times.

Then proceed to Sections 3.1 and 3.2 that briefly describe the different experimental techniques used to obtain diffusion and porosity data as well as the models and techniques used to evaluate the measurements. A more detailed description of the in situ electrical conductivity measurements is given in /Löfgren and Neretnieks 2006/. Here also various sources of uncertainty for laboratory data are discussed.

The main sources of information are summarised in Chapter 3. In this chapter also the results and interpretations of the results are briefly discussed for each investigation. We found this procedure necessary because several issues and causes of uncertainties are directly connected to the investigation method used. They can only meaningfully be dealt with in the proper context. The majority of investigations are made using laboratory techniques. Many of the issues are not directly relevant to in situ data. However, for the sake of completeness and not to miss out potentially relevant questions for in situ data some effort is spent on the laboratory investigations.

We wish to emphasise that the in situ formation factors have been evaluated for depths larger than about 200 m.

We deem that we have captured the main causes of uncertainty that influence PA modelling and that they can be quantified or at least bounded for diffusion and porosity data. Nevertheless, we suggest the reader to also read Chapter 4 in a companion report on uncertainty issues concerning sorption data /Crawford et al. 2006/.

A short summary statement can be made here on the results. Several tens of thousands of measurements show that the formation factor varies somewhat but not very much between rock types and depths. The variability expressed as standard deviation is 0.2 to 0.7 on the ^{10}log scale.

The matrix porosity is assessed from nearly only laboratory measurements and is between 0.02 and 0.2%. Here the uncertainty is caused mainly by the need to account for the influence on rock porosity caused by de-stressing the samples when coring and taking them up from the undisturbed stress in situ.

For PA modelling this uncertainty has little influence, being overwhelmed by the uncertainties in flowpath distributions and flow porosities.

Contents

1 Introduction

This report concerns data and uncertainty assessment of diffusion properties in crystalline rocks in support of the SKB Safety Assessment SR-Can.

1.1 Background

SKB, the Swedish Nuclear Fuel and Waste Management Co, is currently preparing license applications to locate, build and operate (i) an encapsulation plant in which the spent fuel will be emplaced in canisters to be deposited in the deep repository; and (ii) the deep repository for spent nuclear fuel. Several investigations are conducted in support of these license applications.

1.1.1 Planned safety assessments

SKB is currently pursuing site investigations for a deep repository in the municipalities of Östhammar and Oskarshamn. The investigations are conducted in two stages, an initial phase followed, if the expected site suitability is confirmed, by a complete site investigation phase. The aim is to build a deep repository at one of these candidate sites, provided that the bedrock and other relevant conditions are found suitable.

Two safety reports will be produced within the next few years: one that originally was to be used for the application to build an encapsulation plant, SR-Can; and one for the application to build the repository, SR-Site. SR-Can is based on site data from the initial site investigation phase and SR-Site on data from the complete site investigation. After an initial phase of the SR-Can project, an SR-Can Interim report has been produced, with the main purpose of demonstrating the adopted methodology, so that this can be reviewed before it is used for the applications.

1.1.2 Assessing input data – need for traceable expert decision

All input data related to quantitative aspects of the safety assessment will exhibit uncertainties. The quality of the results of any calculation in the assessment will, among other factors, depend on the quality of the input data and on the rigor with which input data uncertainties have been managed. A common and methodological philosophy for the determination of input data and the subsequent management of data uncertainty is therefore required.

In SR 97, a standardised procedure was employed for all input data to radionuclide transport calculations. The outcome was presented in the SR 97 Data Report /Andersson 1999/. The uncertainty treatment in SR 97 is discussed by the SKI/SSI review /SKI and SSI 2001/. The authorities have since conducted some investigations on Expert Judgement /e.g. Wilmot and Galson 2000, Wilmot et al. 2000, Hora 2002, Hora and Jensen 2002/. Also SKB has continued the development work /Hedin 2002, 2003/.

Among other things the reviewers require *quantification of uncertainties* into a form suitable for probabilistic assessment and *traceable records on the expert input* to data selection and uncertainty assessment. A new procedure, based on the one used in SR 97 and taking into account review comments is therefore established for SR-Can.

1.2 Scope and Objectives

This report supplies data with uncertainty to the following SR-Can parameter(s):

- Formation factors of crystalline rocks *under in situ conditions for the sites* being investigated.
- Porosity of the rock matrix *under in situ conditions for the sites* being investigated.

This information is directly used to determine nuclide specific effective diffusion coefficients used in PA models for nuclide transport in the rock.

2 Use in SR-Can

2.1 Applicable SR-Can conditions

We think it will aid the readability of the report that we mention some modelling aspects, uncertainties and data sources already in this chapter although these issues are treated in depth in other chapters and sections.

2.1.1 Conditions for which data are supplied

The importance of matrix diffusion

In the safety assessment of a final repository for spent nuclear fuel, the surrounding rock acts as a barrier for the retardation of released radionuclides. The retardation process in water-conducting fractured rock is due to sorption of radionuclides on the fracture walls combined with diffusion into the fracture coatings and further into micropores in the rock matrix where they can be further adsorbed. The micropores in the crystalline rocks are filled with stagnant groundwater. Under naturally prevailing hydraulic gradients the flow of the groundwater in the rock matrix can be neglected. A dissolved species in the groundwater will mainly be transported by diffusion in the micropores in the rock matrix. This transport process is termed matrix diffusion in the literature /Neretnieks 1980/. Diffusion into rock matrix plays an important role in retarding the release of radionuclides into the biosphere /Neretnieks 1980, SKI 1996, SKB 1999, JNC 2000/.

As the water residence time from the repository depth to the ground surface could be as short as about ten years /SKB 1999/, the retention of radionuclides in the rock matrix is of great importance from the viewpoint of safety assessment. When matrix diffusion is accounted for, the residence time for non-sorbing radionuclides can be enhanced by up to two orders of magnitude /Paper IV in Löfgren 2004/. For sorbing radionuclides, the enhancement can be even many orders of magnitude larger.

The key entities needed when assessing the diffusive properties of a rock are the effective diffusivity D_e ($m^2\ s^{-1}$) and the storage capacity α (–) for the species in the rock matrix. For non-charged species the effective diffusivity can be obtained from the product of the diffusivity of the species in free solution D_w ($m^2\ s^{-1}$) and the formation factor F_f (–). A prerequisite for this is that the characteristic length of the pores is much larger than the size of the species. Otherwise size exclusion effects may occur. For charged species, the diffusion may be decreased by ion exclusion or enhanced by surface diffusion. The storage capacity of the rock matrix ($\alpha = \varepsilon + K_d\rho$) is the combined storage capacity due to dissolved species in the pore water and interaction of species with mineral surfaces by sorption. The key entity for storage of non-sorbing solutes in the pore water is the porosity ε (–).

We alternatively use the terms *formation factor* F_f, and *matrix diffusion properties or data*. The former is a material property that can be assigned numeric values. It can be used to calculate the effective diffusion coefficients for different solutes including radionuclides. The latter has a more general meaning and may include effects such as ion exclusion, surface diffusion or other effects that are not included in the term *formation factor*.

Conditions

The matrix diffusion data delivered in this report is to be primarily used for PA modelling. The focus is therefore on data for in situ conditions for the candidate sites at Oskarshamn and Forsmark.

For PA, matrix diffusion data representing the rock at in situ conditions is needed. The data should represent the rock volume of about 1 km^3, which is of interest for a KBS-3 type repository /SKB 1999/. Both data concerning rock that is adjacent to fractures and rock at some distance from fractures are of interest. The properties of the different rock types at depths varying from a few hundred meters to a kilometre or more below the ground surface are thus the main target. Furthermore, primarily the unaltered rock not immediately adjacent to fractures is targeted although the rock near fractures is also discussed. The latter may influence the retardation of nuclides but it is difficult, not to say impossible, to ensure that the flowpaths will contact mostly the more porous altered rock with enhanced diffusion properties. Also, the in situ techniques used to determine the formation factor for the rock some distance from fractures cannot as yet be unambiguously used to obtain formation factors for the altered rock. This approach is deemed to underestimate the actual retardation of nuclides migrating in flowing fractures.

We will rely heavily on in situ data in this report because these measurements have been obtained at the appropriate temperature, water chemistry[1] and rock stresses and with only slight disturbances of the rock nearest the borehole. As the in situ measurements reach rock more than a metre from the borehole we deem the disturbance to be negligible.

The data are to be used for small ionic species that do not exhibit ion exclusion or surface migration effects. The influences of such effects are discussed in Section 4.2. This will normally apply to all nuclides of interest. Nuclides carried by colloids and very large molecules such as fulvic and humic acids may not access the matrix porosity as readily as small molecules due to size exclusion effects. This is not further discussed in the present report.

There are other secondary uses for the data also mainly aimed at confidence building for use of data and models in PA. Depending on the intended usage, different conditions may be required. Some such issues are briefly discussed here.

Some areas where the data may be used in addition to PA include:

- Interpretation of laboratory and in situ experiments.
- Interpretation of natural analogues.
- Understanding the paleohydrology of a site.

Matrix diffusion data and their uncertainties discussed in this report can be of some value for interpreting laboratory and in situ experiments and for interpreting natural analogues and the paleohydrology of a site.

It may be difficult to assess at what conditions the studied processes occurred. Both spatial and temporal conditions may be needed. It is important to discuss if the proposed conditions are reasonable, if they correspond with the condition used when obtaining the matrix diffusion data delivered in this report, and whether other conditions could be proposed if using other conditions for the matrix diffusion data.

We mention some examples where there may be considerable differences between PA needs and the uses in other supporting modelling and investigations. For example in in situ tracer experiments performed over a year or less the tracer will penetrate a very short distance into the matrix compared to what is expected in PA time scales. The rock thus contacted may be the altered rock and not that deeper in that will influence the nuclide transport over PA time scales.

Another example is the observations of limited penetration depth observed in Uranium series dis-equilibration. This has been used as an argument that a nuclide only may penetrate a few centimetres. This, on the contrary, indicates that the expected retardation for a reactive nuclide actually takes place on a PA time scale /Löfgren and Neretnieks 2006/.

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[1] The uncertainty due to lack of actual chemical composition of the pore waters is discussed in /Löfgren 2004/ and /Löfgren and Neretnieks 2006/.

A third example is when paleohydrologic information of salt displacement is used to compare with expected displacement of salt water by melting ice water or rain penetrating deep into the rock. The water that is sampled at depth has mostly travelled in the highly conductive fracture zones. Exchange with matrix waters will be strongly influenced by the crushed rock in and adjacent to the zones and not with the rock far from such zones where the repository will be located.

Thus the information supplied for PA use cannot directly be used for the other purposes and more importantly vice versa.

To aid understanding the following discussion we summarise some basic concepts here. A more detailed discussion of concepts and models will be given later.

Below the most important conditions needed when presenting matrix diffusion data, in the content of material properties of the rock, are listed. When assessing the radionuclide retention due to matrix diffusion, not only the diffusive properties of the rock matrix are of interest. The retention capacity of the bedrock also depends on what flow paths the solutes are transported along, on its way from the repository to the biosphere. Here the length of the flow path, the Flow Wetted Surface (FWS) and the flow rate along a path are key entities. The influence of these entities on retention is discussed later in Section 2.2.1.

The following factors can influence the matrix diffusion properties. They also influence the interpretation and presentation of the data.

Rock type

Results from recent studies suggest that there may be a correlation between the rock type and the formation factor and porosity /e.g. Löfgren 2004/. Even though this correlation may be weak and rock type classification is not an absolutely clear-cut science, the rock type is an important condition for both the formation factors and porosities presented in this report.

Rock type heterogeneity

The geology of a site may be lithologically homogenous or heterogeneous. This mainly influences the measurements in the following ways. Firstly, the more heterogeneous the bedrock is, the greater the natural variation of the formation factor and porosity is likely to be. Therefore, more data points may be required to obtain results that represent the site.

Secondly, depending on the ratio between the scale of heterogeneity and the scale of the sample, the obtained results may differ. If the scale of the sample is on the same scale as the heterogeneity, it is more likely that “extreme” formation factors and porosities are obtained. If the scale of the sample is much larger than the scale of the heterogeneity, some averaging will occur.

Fracture heterogeneity

The degree of fracturing relates both to the advective and the diffusive transport of solutes in fractured crystalline rock. The fracture characteristic that relates to advective transport is discussed in Section 2.2.1 of this report.

For the diffusive transport, fractures that hold stagnant water will increase the storage capacity of the rock matrix. Firstly, the porosity of the rock matrix is increased. Secondly, more rock matrix may be utilised for sorption and storage in the pore water, especially if limited pore connectivity is assumed. Therefore, the extent of which the fractures, holding stagnant water, are included in the rock matrix is an important condition both for the formation factor and porosity.

Groundwater chemistry

For certain species, the matrix diffusivity may be changed by the groundwater chemistry, or more correctly by the pore water chemistry.

For non-sorbing species this especially applies to anions, which are subjected to anion exclusion. The anion exclusion is more important in pore waters of low ionic strength than in pore waters of high ionic strength.

For sorbing cations, the diffusive flux through the rock matrix may be enhanced by diffusion in the electrical double layer. The same applies for an electro-migratory flux. The flux may be more or less enhanced depending on the ionic strength and concentration of solute in the pore water.

As the pore water chemistry may be important for the migration of certain species in the rock matrix, the groundwater chemistry is assessed to be an important condition, especially in respect of ionic strength and electrical conductivity.

Temperature

The effective diffusivity is to some extent dependent on the temperature. Therefore the temperature is a reasonable condition for the diffusive properties of rocks presented in this report. However, as the temperatures of interest range only over less than tens of Kelvin, corrections due to the temperature would only influence the results marginally.

Degree of alteration

The rock adjacent to water-bearing fractures could have been altered in different processes including hydrothermal alteration, metamorphism, and weathering. It is frequently reported that the rock in the alteration zone has a higher porosity and formation factor than the undisturbed rock /e.g. Byegård et al. 2002/. Therefore, the degree of alteration is an important condition for both the formation factors and porosities presented in this report.

Rock stress

In a number of important applications, for example in performance assessments, diffusive properties of rock that is subjected to rock stress are needed. In situ the rock stress derives from the pressure from the overburden as well as from lateral stress induced by tectonic processes. The higher the rock stress, the more likely it is that the porous system is compressed. Therefore, both the formation factor /Skagius 1986/ and the porosity /Möri et al. 2003/ are likely to be reduced in stressed rock compared to destressed rock. The rock stress is an important condition for both the formation factors and porosities presented in this report.

2.2 Models

2.2.1 Models used for transport of radionuclides in the geosphere

The following models are used to calculate solute transport and radionuclide migration in SR-Can.

- FARF31
- CONNECTFLOW
- DarcyTools
- Chan3D

FARF31 /Norman and Kjellbert 1990, Elert et al. 2004/ is the main code for simulation of radionuclide transport from the repository to the biosphere in the performance assessment. It is based on modelling flow and transport in a dual porosity medium using the Advection-Dispersion, AD, formulation, and accounts for chain decay and diffusion in the rock matrix and linear sorption.

The groundwater flow is modelled through a discrete fracture network (DFN) where individual fractures are represented explicitly /SKB 2006a/. The conceptualisation of a migration path used in SR-Can project is slightly different than in a continuum-based groundwater flow model such as HYDRASTAR /Morris and Cliffe 1994/, previously used in SR 97. Rather than macroscopic streamtubes encompassing both rock and flow paths, the equation in the DFN model describes flow paths through the actual open pore space, i.e. through the connected fracture network /RETROCK 2004/. The entities calculated in the DFN-based groundwater flow models are the advective travel time and transport resistance.

CONNECTFLOW /Hartley and Holton 2003/ combines the facilities of NAMMU /Cliffe et al. 1999/ which is based on a finite element application to continuum- porous media models and NAPSAC /Hartley et al. 2002/ for modelling discrete fracture networks. CONNECTFLOW is used in essentially the same way as HYDRASTAR for flow but particle tracking can be done in the fracture network. CONNECTFLOW will also be used in the PA to track paths from individual canisters intersected by one or more fractures.

The other three programs are used for supporting calculations to interpret field tracer tests, paleohydrological observations, impact of transient flow conditions and a variety of other tasks including scoping calculations to gain insights into different questions.

In each model stream tubes or stochastic flowpaths can be defined and the following mechanisms that influence the nuclide transport can be included.

1. Advective transport by the flowing water.
2. Hydrodynamic dispersion in the flowing water.
3. Diffusion of the nuclides in and out of the porous rock matrix of the rock “blocks” in the streamtube or the rock adjacent to the flowpaths.
4. Sorption of nuclides on micropore surfaces in the rock matrix.
5. Radionuclide decay, including chain decay and generation of daughter nuclides.

A type of numerical implementation of FARF31 /Vahlund and Hermansson 2004/ can also account for nuclide transport by colloids.

In this report data for Item 3, diffusion in the rock matrix are discussed.

Item 4, sorption, which is closely related to diffusion, is discussed in another report /Crawford et al. 2006/.

DarcyTools /Svensson et al. 2004/, CONNECTFLOW, and Chan3D /Gylling 1997/ are at present used mostly for supporting simulations such as studies of paleohydrological flow and salt transport in the rock simulating what may have happened after the last ice age regarding flow when land rises and hydraulic boundary conditions change, transport of nuclides that diffuse in the matrix during changes of flowrates (Chan3D), evaluation of tracer tests such as those at Äspö and other supporting studies. These include investigations of whether thin zones of altered rock adjacent to the flowing fractures can influence the nuclide retardation, whether fracture and channel network models behave similarly to porous media models, whether correlations between fracture sizes and transport properties influence the Residence Time Distribution (RTD) of the nuclides etc. DarcyTools is also used for all Open Repository simulations within SR-Can. Further, a number of simple models which capture the main processes and have analytical solutions are used for exploratory studies of nuclide migration in fractured rocks.

The by far most important mechanism for retarding the nuclide transport compared to the water movement is matrix diffusion. For sorbing nuclides sorption further dramatically enhances retardation.

The core of all these models is the mechanism of nuclide diffusion and sorption in the rock matrix coupled to the advective flow of the mobile water.

These processes are technically handled somewhat differently in the different computational programs (codes). They all give essentially the same results when simulating the same problem.

The simple analytical solutions give good insights into the importance of the different mechanisms and a short description is given below as a basis for later discussion on how diffusion data and the uncertainties and variabilities of data influence the RTD of the nuclides.

2.2.2 Basic conceptual model and some analytical solutions

Basic equations

The aim of this section is to highlight the mechanisms that are most important for retarding the radionuclides in PA situations.

It will be shown that:

- The contact area between the flowing water and the rock is highly important.
- The large variations of diffusion and sorption parameters due to rock heterogeneities along the flowpaths are evened out and that the higher values in the parameter distributions contribute most to the retardation of the nuclides.

In previous PA modelling of far-field fluid flow and solute transport, both continuum porous medium (CPM) and discrete fracture network (DFN) approaches have been used /Hartley et al. 2004/. In the SR-Can project the DFN approach is used, especially in the local scale /SKB 2006b/. The differences of the two approaches lie mainly in the modelling of the flowpaths. In the CPM models, the concepts of streamtubes (often one-dimensional) are used. It is often assumed that mixing of solutes occurs only within a streamtube and there is no mixing of solutes between streamtubes. In the DFN models, however, one flowpath usually consists of a series of connected sub-flowpaths in a network of fractures. The water residence time and the flow-wetted surface (FWS) are obtained in the CPM models by averaging over the varying cross-section area of the streamtube, while in the DFN models they are obtained by summation of the values in the sub-flowpaths. Such a flowpath is equivalent to a streamtube. In both approaches the underlying mechanisms for solute (radionuclide) transport are essentially the same /RETROCK 2004/. In the following, we use the streamtube concept to illustrate the basic mechanisms of solute transport and retention.

The streamtube concept is used to describe the flow in fluids and also in porous media where it is implicitly assumed that the solid particles making up the porous medium are so small that their local influence on the flow field can be neglected when averaging the velocities in the streamtube.

The commonly used advection-dispersion model, accounting for interaction with the rock matrix, can be formulated in mathematical terms by the following differential mass balance. The conventional mathematical form in continuum formulation, neglecting hydrodynamic dispersion, is

$$\frac{\partial c}{\partial t} + u\frac{\partial c}{\partial x} = a_w D_e \frac{\partial c_p}{\partial z}(z=0) \qquad \text{(Equation 2.1)}$$

$$\frac{\partial c_p}{\partial t} = \frac{D_e}{\alpha}\frac{\partial^2 c_p}{\partial z^2} \qquad \text{(Equation 2.2)}$$

where

c is concentration in the flowing water (mol m^{-3}),

x is coordinate in flow direction (m),

u is water velocity (m s^{-1}),

a_w is the FWS per volume of water (m^2 m^{-3}),

c_p is concentration in pore water in rock matrix (mol m^{-3}),

D_e is effective diffusivity (m^2 s^{-1}),

z is distance into rock matrix (m),

t is time (s).

The rock capacity factor α is

$$\alpha = \varepsilon_p + K_d \rho = \varepsilon_p + K \cong K \qquad \text{(Equation 2.3)}$$

where

ε_p is matrix porosity (–),

K_d is mass based sorption coefficient (m^3 kg^{-1}),

ρ is rock density (kg m^{-3}),

K is volume based sorption coefficient (–).

This is the basic equation upon which most solute transport models are based. The models differ mostly in how the equation is implemented in different conceptualisations of the rock mass.

Hydrodynamic dispersion is neglected in this simplified analysis because we want to highlight the impact of matrix diffusion on the RTD of a solute. Furthermore, the conventional way of using hydrodynamic dispersion is increasingly challenged.

The equation describes a mass balance of a solute. It was originally formulated for transport in a porous medium but can be applied to fractures in fractured rocks also. It will form the basis for further discussions.

The streamtube concept is illustrated in Figure 2-1. Mass can enter and exit at in- and outlet only, not over boundaries. The flowrate q is constant in a streamtube. The cross-section A, water velocity and contact surface between water and rock fracture, a_w, can vary along the streamtube.

It is obvious that for fractured rocks this is a challengeable concept because fractures and thus flow will go in and out through the streamtube walls. It is defended by the argument that if the streamtube size is made large the flow over the boundaries will be small compared to the flow through the streamtube.

In Equation (2.1) the water flows in the x direction. The last term accounts for the exchange of the nuclide with the porous rock matrix. The diffusion is directed in the z-direction into the rock matrix.

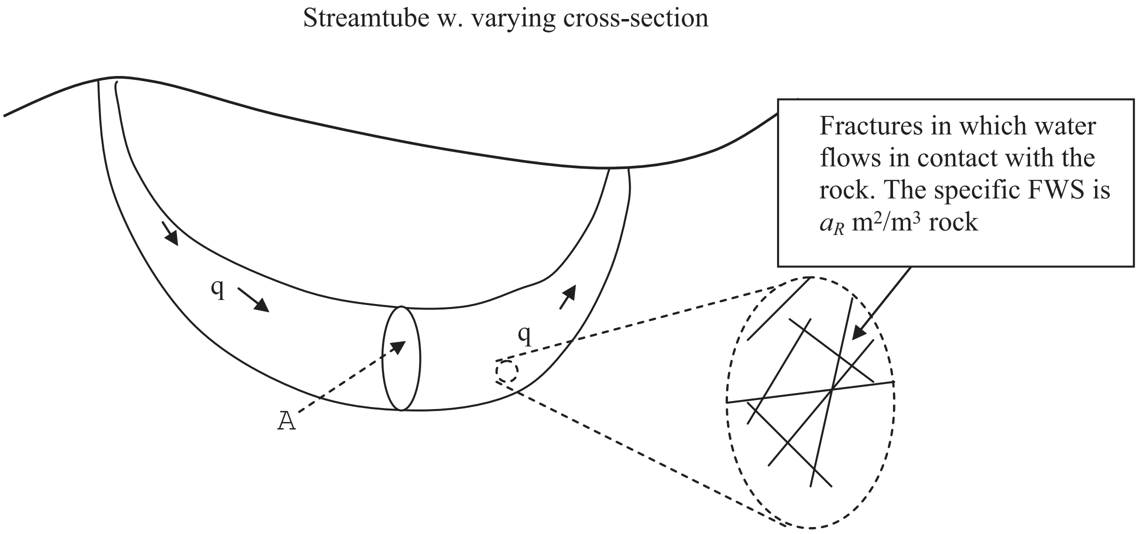


Figure 2-1. *Illustration of the concept of a streamtube.*

Intuitively one expects that if the same water package with flowrate q is in contact with a larger FWS the exchange of solute between the flowing water and the rock matrix will be larger. It should be noted that when the volume of the streamtube is known the water residence time in it is simply obtained as the ratio of mobile water volume V_m to the flowrate q, i.e. $t_w=V_m/q$. In the same way with a total FWS in the streamtube that the stream encounters being A_q, the ratio of $A_q/q=Va_R/q$ where V is the total volume of the streamtube and a_R is the FWS per unit volume of the rock. V_m/V is the flow porosity ε_f, not further needed in this discussion. *The emphasis is on the interaction with the matrix*. It should be noted that in the DFN approach, the ratio of A_q/q is obtained directly from the flow simulations. Also the specific FWS, a_R, can be calculated in the DFN models and be compared with the conductive fracture frequency (CFF) obtained from borehole data.

We intentionally simplify the reasoning and do not here discuss the fact that different fractures in the fracture network will carry different flowrates and thus have different nuclide retardation properties. A more detailed description on these issues can be found in /RETROCK 2004/.

The entity A_q/q has the highest impact on the RTD of the nuclides in the streamtube in addition to the sorption coefficients and the diffusivity in the rock matrix.

For illustration purposes we choose a streamtube in a fracture that can vary in aperture and where therefore the FWS will be the two sides of the fracture that the water package contacts. It is shown that under some simplifying conditions (neglecting hydrodynamic dispersion) variations in water velocities do not influence the retardation of the nuclides /RETROCK 2004/. Only the total FWS encountered by the stream matters. The total FWS will be the integral of the surface along the flowpath. This is shown in Figure 2-2 below.

This is readily seen in the solutions of Equations (2.1) and (2.2), Equations (2.4) and (2.5) below. It is exemplified for the case when the rock matrix extends very far compared to the penetration depth of the solutes and when a constant concentration stream is fed at the inlet. The reasoning applies also to cases where the matrix has a limited extent and for chain decay but the equations become increasingly complex /Neretnieks 2006/.

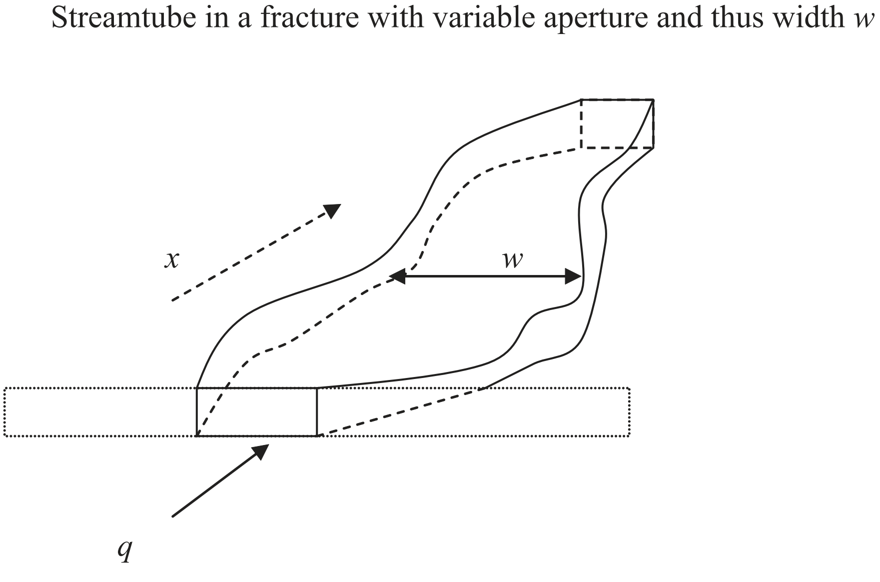


Figure 2-2. *A channel in a fracture with variable aperture where a stream with constant flowrate q is followed. The stream contacts a FWS made up of the two sides of the fracture.*

$$\frac{c}{c_o} = Erfc\left(\frac{X}{2(t-t_w)^{\frac{1}{2}}}\right) \qquad \text{(Equation 2.4)}$$

where

$$X = \int_0^x \frac{a_w\sqrt{D_e\alpha}\,dx'}{u} = \frac{2}{q}\int_0^x w(x')\sqrt{D_e\alpha}\,dx' = \frac{2}{q}\int_0^x w(x')MPG(x')dx' \cong \frac{2}{q}\int_0^x w(x')\sqrt{D_eK}\,dx' \qquad \text{(Equation 2.5)}$$

For constant material properties of D_e and K along the pathway, X simplifies to

$$X = \frac{2}{q}MPG\int_0^x w(x')dx' \cong \frac{A_q}{q}\sqrt{D_eK} \qquad \text{(Equation 2.6)}$$

where A_q is the FWS the stream with flowrate q encounters along the flowpath. The last terms in Equations (2.5) and (2.6) apply to sorbing species. This way of describing the entities involved shows that by following a constant stream, i.e. using the streamtube concept, the physical entities that influence the matrix interaction are clearly seen.

Inspection of Equation (2.4) reveals that there are only two entities that need to be known to obtain the RTD in a streamtube under these simplified conditions, namely the water residence time t_w and the entity X. The former is obtained from the mobile water volume in the streamtube (information of rock porosity is needed). The entity X is obtained by integrating along the streamtube (or flowpath) as shown in Equation (2.5). In practice the variation of K and D_e along the path is not known and averages of these entities are used.

This leads to an important consequence for determining the mean and variance of D_e and K to be used in PA simulations. The very large variations of these entities along a flowpath will be considerably evened out, the higher part of the distributions will have a larger impact than the lower end and the variance of the averages will be much smaller than the variance of the distribution of individual measurements. This effect is illustrated below for D_e distributions.

For visualisation purposes we determine the time of arrival $t_{0.00001}$ of a fraction of the inlet concentration at a point downstream in a flowpath. Equation (2.4) is solved for example for $c/c_o = 10^{-5}$

$$t_{0.00001} = t_w + 0.1\frac{X^2}{4} = t_w + 0.025(\frac{A_q}{q})^2 KD_e \qquad \text{(Equation 2.7)}$$

Equation (2.7) is only modified by the constant, in this case 0.1, if another fraction of the concentration is chosen. For example for $c/c_o = 0.16$, the constant is equal to 1. The form of the equation is the same. The first term represents the residence time of the water and the second term the residence time due to delay in the matrix.

For PA applications and sorbing nuclides usually t_w can be neglected compared to the matrix residence time.

Inspection of Equation (2.7) reveals that a variation or uncertainty in K or D_e will give the same variation in the arrival time whereas the same uncertainty in A_q/q will have a square influence. At present the uncertainty in the latter entity is deemed to be larger than that for K or D_e and will contribute more to the total uncertainty of the residence time distribution along a path.

Probability distribution for D_e

The distribution of matrix diffusion data is well described by a log normal distribution with mean μ and standard deviation σ in ^{e}log space. See Chapter 5.

$$f(D_e) = \frac{1}{\sigma D_e\sqrt{2\pi}}e^{-\frac{(\log(D_e)-\mu)^2}{2\sigma^2}} \qquad \text{(Equation 2.8)}$$

The arithmetic mean of D_e is

$$\overline{D}_e = e^{\mu + \frac{\sigma^2}{2}} = e^{\mu} e^{+\frac{\sigma^2}{2}} \qquad \text{(Equation 2.9)}$$

This implies that the arithmetic mean increases by a factor of $e^{\frac{\sigma^2}{2}}$ over the logarithmic mean. For a (typical) σ value of 0.5 in the common logarithmic (^{10}log) scale, the value in the natural logarithmic (^{e}log) scale will be 2.303*0.5. The increase is then by a factor of 1.94, which nearly doubles the value. (The term 2.303 is used to convert from ^{10}log to ^{e}log).

The standard deviation in linear space s of D_e is

$$s = \overline{D}_e \sqrt{(e^{\sigma^2} - 1)} \qquad \text{(Equation 2.10)}$$

with the above σ, $s = 1.66\,\overline{D}_e$.

Figures 2-3 and 2-4 show the probability and the distribution functions of D_e for $\mu = 2.303 {*} {}^{10}\mathrm{Log}(10^{-14})$ and $\sigma = 2.303{*}0.5$. The plot clearly shows that the higher values will contribute much to the mean. The latter is the cumulative function of the former. These figures illustrate the variations of the diffusivity caused by heterogeneities in a rock type. Different rock types may have different distributions.

Integration of D_e along the flowpath

In the PA simulations representative data for the various parameters will be chosen. This may e.g. be based on the mean values of the distributions. In addition, the impact of variations and uncertainties of the parameter values will be studied by selecting values both smaller and larger than the mean.

Assuming that in a flowpath the water will contact the heterogeneous parts of the rock according to the distribution of the heterogeneities the above reasoning can be used to reduce the uncertainties of $\overline{D}_e$ along the path as shown in equation 2.5.

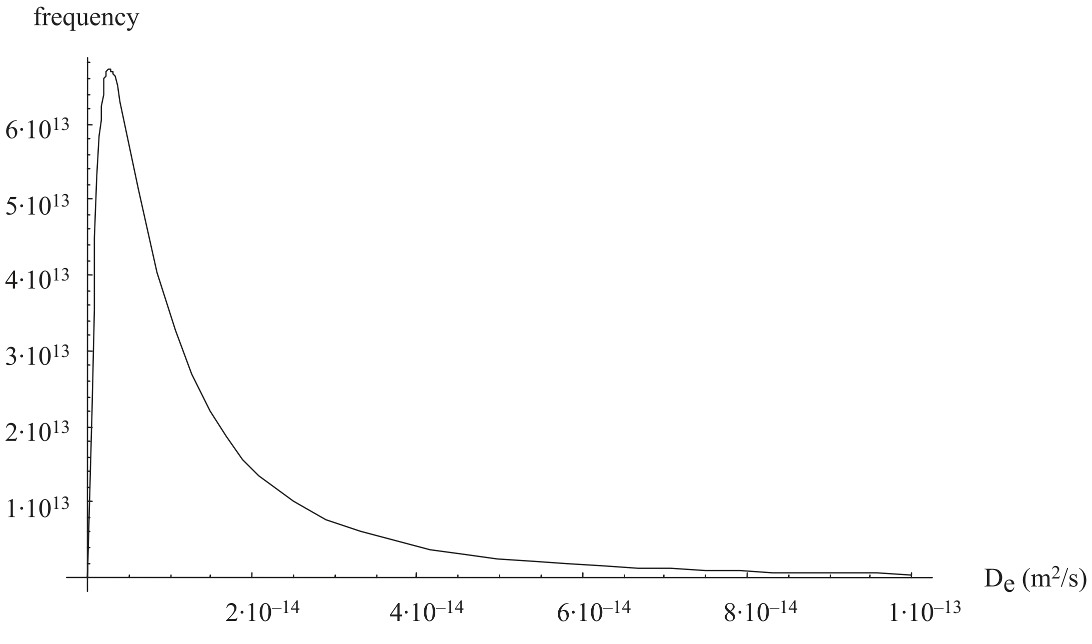


Figure 2-3. *Probability function of D_e with the above data.*

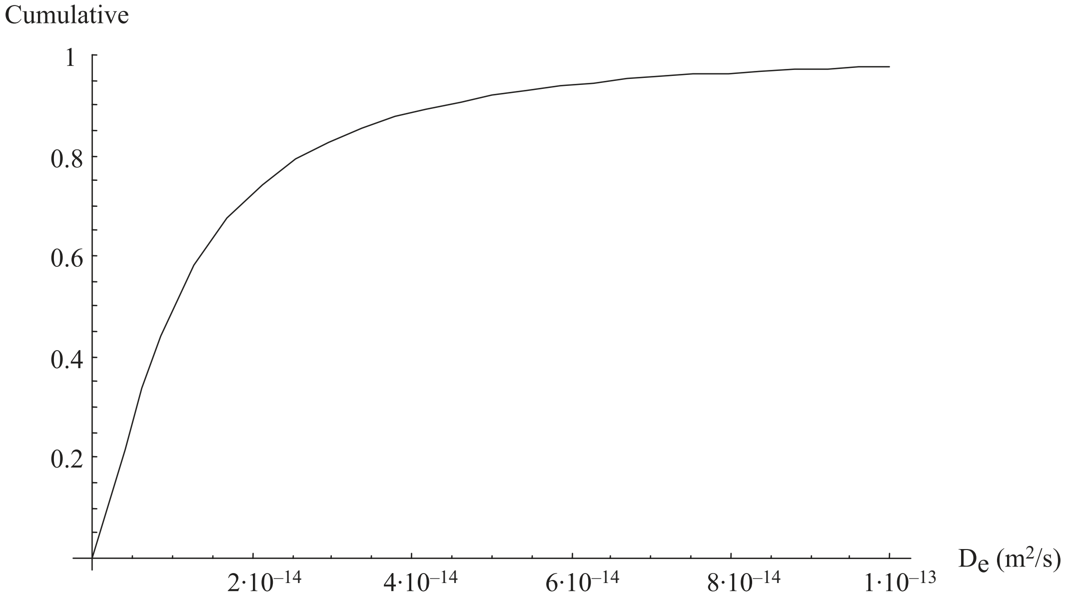


Figure 2-4. *Distribution function of D_e with the above data.*

For the above reasoning it is inherently assumed that the local D_e along the flowpath is the same in the diffusion direction, that is, into the rock matrix. This, however, also must be expected to vary. Below we present a short discussion of what influence this can have on the assessment of D_e values used for transport modelling.

The heterogeneities in the rock cause much of the variation in the diffusivities. The measurements in the deep boreholes, which are typically made every 10 cm, reveal that very nearby samples may have very different diffusivities /Löfgren and Neretnieks 2005ab/. The same observation can be made for nearby samples in the laboratory, where the sample length is only a few centimeters /Löfgren 2001/.

From the observation that the variation in the diffusivity typically is larger for the smaller samples in the laboratory than for the larger samples in situ, one may conclude that the scale of the heterogeneity is on the same scale, or smaller, as that of the laboratory samples. There seems to be more averaging when measuring in situ, as compared to in the laboratory.

If assuming that the scale of the heterogeneity is much smaller than the scale of the samples, one may be able to assess the variation of a specific rock property for a certain sample size based on its variation for another sample size. This is straightforward when it comes to a property like the matrix porosity. Each sample could then be divided into a number of equally sized unit cells. If the porosity of each unit cell is known, the average porosity of the sample can be calculated. Larger samples would contain a larger number of unit cells and, as there would be more averaging, the variation in the average porosity for different samples would be smaller. The relation between the variation (expressed as standard deviation) of the porosity for the larger and smaller sample would then be:

$$s_{\text{large}} = \sqrt{\frac{V_{\text{small}}}{V_{\text{large}}}} \cdot s_{\text{small}} \qquad \text{(Equation 2.11)}$$

where s_{large} and s_{small} are the standard deviation of the large and small samples respectively and V_{large} and V_{small} are the volumes of the large and small samples respectively.

For diffusion through a porous medium, it is not as straightforward. The reason is that in order for a species to diffuse from one unit cell to another, both cells have to be accessible for diffusion. One can thus not obtain the average diffusivity of the sample by simply averaging the diffusivities of the unit cells. If assuming one-dimensional diffusion through the rock one may suggest that the relation between the variations of two sample sizes is:

$$s_{large} = \sqrt{\frac{L_{small}}{L_{large}}} \cdot s_{small} \qquad \text{(Equation 2.12)}$$

where L_{small} and L_{large} is some characteristic length of the small and large samples respectively. As it is not known exactly how to average the diffusive properties of the rock matrix, there is a need to investigate the three dimensional aspects of the problem. The averaging may have a great consequence for the variance of the effective diffusivity used in the performance assessment.

It should be noted that the above averaging of diffusivity is made along the direct of diffusion, i.e. in the direction more or less perpendicular to the flowpath in the DFN approach.

Consider an example where the distance between flowing fractures is 30 m. A non-sorbing tracer would access all the pores in the rock in PA time scales and an average over this distance is sought. Assuming that Equation (2.12) could be used, the PA scale variance would only be a fraction of the laboratory scale variance, where the sample length typically is 3 cm.

$$s_{30} = \sqrt{\frac{0.03}{30}} \cdot s_{0.03} \approx 0.03 \cdot s_{0.03} \qquad \text{(Equation 2.13)}$$

This is a considerable decrease in uncertainty. For sorbing nuclides, which are only expected to penetrate centimetres to tens of centimetres over PA time scales the depth averaging would not apply. However, the averaging along a flowpaths still applies. This considerably decreases the impact of variability and uncertainty of the matrix diffusion data on solute transport.

2.3 Results of sensitivity analysis – if any?

2.3.1 Sensitivity of the matrix diffusion data

The sensitivity of nuclide residence time to matrix diffusion has been discussed above in a general sense. See e.g. Equation (2.7) and the following discussion. Below some further, more detailed, issues are discussed.

Other factors related to matrix diffusion may have impact on the tracer residence time

In a large perspective, the tracer residence time depends on the total area of the flow-wetted surface (FWS) (which in turn depends on the length of the flow path) and the material properties group (MPG) parameters including the diffusivity and the sorption coefficient. As has been discussed in Sections 2.2.1 and 2.2.2, the FWS may have even a larger impact on the tracer residence time than the MPG parameters. Large uncertainties are also related to the FWS.

The importance of the scale of pore connectivity in the rock matrix

In the models of matrix diffusion it is often assumed that micropores in the rock matrix are connected over large distances in rock under natural stress. Laboratory demonstrations of pore connectivity and through-diffusion experiments do not suffice to prove this. In addition, rock samples used in laboratory experiments have been inevitably subjected to stress release, damages during excavation and sample preperation, etc and the results of laboratory experiments may not be representative for those of the undisturbed rock. Field scale experiments in rocks at large depths have thus been conducted to determine the connectivity of the micropore systems in unaltered granitic rock matrix /Birgersson and Neretnieks 1990/. The results revealed that micropore connectivity extends to at least 40 cm. In dense granitic bedrock, matrix diffusion is a very slow process and in some cases indirect indications of the pore connectivity and matrix diffusion have been used to support unlimited connectivity of the micropore structure. The results of the resin impregnation method /Möri et al. 2003, Ota et al. 2003/ suggested an interconnected pore system within the rock several metres away from a water-conducting shear zone.

Based on the analogy between the diffusion process and the electrical conductivity process in pore water, /Löfgren 2001/ has performed in situ electrical conductivity borehole logging in granitic rocks /Löfgren et al. 2001, Löfgren and Neretnieks 2002, 2003, Löfgren 2004/. Their results indicate that the unaltered rocks have micropore connectivity up to at least metres.

The impact of rock type and especially rock heterogeneity on the results of safety assessment

Due to rock heterogeneity in any candidate site, the diffusivity (or the rock formation factor) varies over some 6 orders of magnitude. The impact of this variation, however, could be possibly evened out along any flow path and the final result of the tracer residence time may depend just on the arithmetic mean value of the data. See discussions in Sections 2.2.1 and 2.2.2.

Presence of altered rock

In many fractures alteration of the rock nearest the fracture has been observed. The extent of alteration can vary from practically nothing to millimetres and sometimes several centimetres. Commonly the altered rock has increased porosity and therefore diffusivity. This could enhance the uptake of nuclides into the matrix. However, as flowpaths are not known in detail it cannot be assured that these positive effects can be included in PA modelling.

The presence of open fractures with stagnant water and alteration zones around fractures are found to have increased electrical conductivity in the down-hole measurements. These data cannot be included in the database for undisturbed rock properties. It may be tempting to argue that flowpaths also should encounter the same frequency of altered rock as found in boreholes and that these data should be included. We have decided not to do this and present the data for undisturbed rock separated from data on sections where fractures are present. There are several reasons for this. The main reason is that the geometries for the diffusion are different when diffusion is into the undisturbed rock in the first case and into a stagnant water in a fracture or in a thin altered zone and then further from these regions into the undisturbed rock perpendicular to the first direction in the second case. Another reason is that there is limited information on the correlation between diffusion and sorption in the altered regions.

3 Sources of information

3.1 Databases

3.1.1 Databases for matrix diffusion and rock porosity

In this report site-specific formation factors and porosities are presented from three subareas, the Forsmark, Simpevarp and Laxemar subareas. This section gives a short description of the different subareas. It also gives a short description of the campaign wherein data have been obtained and references to publications where more relevant information can be found. Furthermore, some generic formation factors and porosities are given and the sources and databases from which they have been compiled are described.

Forsmark

A geological interpretation of the rock surrounding the boreholes KFM01A, KFM02A, KFM03A, KFM04A, and KFM05A can be found in /Carlsten et al. 2004ab/ and /Thunehed and Keisu 2004/. A Site Descriptive Model of the Forsmark site can be found in /SKB 2006/.

Intrusive igneous rocks dominate the Forsmark region, whilst supracrustal rocks form a subordinate component. Apart from some younger granites and pegmatites, which display only a weak foliation, all these rocks were more or less affected by ductile deformation and underwent a recrystallisation at depths probably greater than 15 km and under amphibolite-facies (> 500–550°C) metamorphic conditions. Generally, medium-grained metagranite-granodiorite dominates the site. Amphibolite, pegmatitic granite and a fine-medium grained metagranitoid occur as subordinate rock types.

Site-specific formation factors, characteristic for the rock surrounding the boreholes KFM01A and KFM02A, have been obtained both in the laboratory and in situ. The in situ campaign wherein the formation factors were obtained is described in:

- **/Löfgren and Neretnieks 2005b/.** Forsmark site investigation: Formation factor logging in situ and in the laboratory by electrical methods in KFM01A and KFM02A. Measurements and evaluation of methodology. SKB P-05-29. Svensk Kärnbränslehantering AB.

In situ formation factor data between the borehole lengths 116 m–995 m and 145 m–999 m are tabulated in the SICADA database for KFM01A and KFM02A respectively. Martin Löfgren and Ivars Neretnieks, Chemical Engineering and Technology, Royal Institute of Technology in Stockholm, Sweden have compiled the data.

The in situ formation factors are based on in situ rock resistivity and in situ groundwater electrical conductivity measurements. The in situ rock resistivities were measured by the Antares Dual-Laterolog, a focused tool that was used in the short mode (DLLs). The rock resistivity was measured every 0.05 m in the borehole. The data from the in situ rock resistivity measurements can be found in the SICADA database. The electrical conductivity of the groundwater was measured on groundwater extracted from fractures at specific depths. In doing this, a drawdown was used at the surface. The electrical conductivity of the groundwater was either measured in the hydrogeochemical program or by the Posiva different flow meter. As only a number of measurements could be performed per borehole, some inter- and extrapolation was needed. For further references, consult /Löfgren and Neretnieks 2005b/.

The laboratory campaign, wherein laboratory formation factors have been obtained, is described in:

- **/Thunehed 2005/.** Forsmark site investigation: Resistivity measurements on samples from KFM01A and KFM02A. SKB P-05-26. Svensk Kärnbränslehantering AB.

The formation factor was measured by electrical methods. The measurements were performed by GeoVista AB, Luleå, Sweden. Laboratory formation factors obtained between the borehole lengths 101 m–980 m and 100 m–1,001 m are tabulated in the SICADA database for KFM01A and KFM02A respectively. The rock samples were, in general, taken every 20 m from the drill cores.

The porosities from the drill core samples described above were supplied by Johan Byegård of Geosigma, Kungälv, Sweden.

Simpevarp

The two boreholes investigated, KSH01A and KSH02, are situated on the Simpevarp peninsula. The geology of the Simpevarp peninsula is described in /Curtis et al. 2003/ and /Bergman et al. 2000/. The rock types in this area are members of the so-called Småland-Värmland intrusion, which is dominated by ca. 1,700–1,800 million year old granites and related metavolcanites. The Småland granite has intruded the metavolcanite to such an extent that the metavolcanite has re-crystallised and developed a more granitic structure. Thus, for the most part, the area containing the metavolcanite is best described as a complex mixture of granite and metavolcanite. Red-greyish red, fine-grained granite occurs throughout the peninsula, commonly as irregular dykes and veins in the older rock mass. Due to the complex mixture of rock types, the site has to be considered as lithologically inhomogeneous.

Fracture zones oriented against all four cardinal points confine the Simpevarp peninsula. Within the area, minor fractures occur frequently. The rock surrounding KSH01A is dominated by dioritoid, quartz monzonite to monzodiorite, granite to quartz monzodiorite and fine-grained mafic rock. The rock surrounding KSH02 is dominated by dioritoid and fine- to medium-grained granite.

Site-specific formation factors, characteristic for the rock surrounding the boreholes KSH01A and KSH02, have been obtained both in the laboratory and in situ. The campaign wherein the formation factors were obtained is described in:

- **/Löfgren and Neretnieks 2005a/**. Oskarshamn site investigation: Formation factor logging in situ and in the laboratory by electrical methods in KSH01A and KSH02. Measurements and evaluation of methodology. SKB P-05-27. Svensk Kärnbränslehantering AB.

In situ formation factor data between the borehole lengths 162 m–1,001 m and 180 m–999 m are tabulated in the database SICADA for KSH01A and KSH02 respectively. Martin Löfgren and Ivars Neretnieks, Chemical Engineering and Technology, Royal Institute of Technology in Stockholm, Sweden have compiled the data and performed the laboratory work.

The in situ formation factors are based on in situ rock resistivity and in situ groundwater electrical conductivity measurements. The in situ rock resistivities were measured by the Century 9070 log. The rock resistivity was measured every 0.1 m in the borehole. The data from the in situ rock resistivity measurements can be found in the SICADA database. The electrical conductivity of the groundwater was measured on groundwater extracted from fractures at specific depths. In doing this, a drawdown was used at the surface. The electrical conductivity of the groundwater was either measured in the hydrogeochemical program or by the Posiva different flow meter. As only a number of measurements could be performed per borehole, some inter- and extrapolation was needed. For further references, consult /Löfgren and Neretnieks 2005a/.

Laxemar

Site-specific formation factors, characteristic for the rock surrounding the borehole KLX02 have been obtained both in the laboratory and in situ. The formation factors were obtained in the sections 350–400 m and 720–800 m. The campaign wherein the formation factors were obtained is described in:

- **/Löfgren 2001/.** Formation Factor Logging in Igneous Rock by Electrical Methods. Licentiate Thesis, Dept. of Chemical Engineering and Technology, Royal Institute of Technology, Stockholm, Sweden. ISBN: 91-7283-207-X

Martin Löfgren and Ivars Neretnieks, Chemical Engineering and Technology, Royal Institute of Technology in Stockholm, Sweden have compiled the data and performed the laboratory work.

The in situ formation factors are based on in situ rock resistivity and in situ groundwater electrical conductivity measurements. The in situ rock resistivities were measured by the Antares Dual-Laterolog, a focused tool that was used in the long mode (DLLl). The rock resistivity was measured every 0.05 m in the borehole. The data from the in situ rock resistivity measurements can be found in the SICADA database. The electrical conductivity of the groundwater was measured on groundwater extracted from fractures at specific depths. In doing this, a drawdown was used at the surface. The electrical conductivity of the groundwater was measured by the Posiva different flow meter. As only a number of measurements could be performed per borehole, some inter- and extrapolation was needed. For further references, consult /Löfgren 2001/.

Porosity and Formation factor data obtained in the laboratory and in situ by methods based on diffusion

Diffusion data of sorbing and non-sorbing tracers in crystalline rock matrix have been obtained by direct diffusion measurements mostly in 1980's by researchers in Sweden, Finland, the United Kingdom and Switzerland. The methods for determining the data were through-diffusion, in-diffusion using rock samples with thickness up to a few centimetres, and diffusion coupled with sorption in crushed rock with particle sizes of less than 1 mm.

In the following we will mainly present diffusion data in the forms of porosity and rock formation factor. Discussions of relations among porosity (including transport porosity and porosity of the dead-end pores), storage capacity, effective diffusivity, apparent diffusivity and rock formation factor can be found in Section 3.2.1: Modelling used to support the data.

As this report is intended to review and recommend data in support to the SKB Safety Assessment SR-Can, we are concerned mainly with the matrix diffusion data measured for crystalline rocks at some Swedish and Finnish sites. Matrix diffusion data measured by diffusion methods in laboratory are also available for crystalline rocks at the Swiss sites /Carlson and Platz 1985, Smith 1990/, at the Japanese sites /e.g. Idemitsu et al. 1992, Yamaguchi et al. 1993, 1997/, as well as at the Canadian sites /Cramer et al. 1997, Vilks et al. 2003, 2004/ which are not specifically dealt with in this report. It should be noted that all the other results that are not discussed in this report do not show any contradictions to the main conclusions of this report.

The porosity and the rock formation factor of a crystalline rock are essentially the properties of the rock itself. To measure them different tracers are used in the laboratory diffusion experiments. For reasons that will be discussed in more detail in Section 4.2.1, the measured values of the porosity and rock formation factor will to some extent depend on the tracers used. The tracer-dependent variations are, however, often less than the natural variations of the rock properties themselves due to heterogeneities of the rocks. In the following we will therefore focus mainly on data on porosities and formation factors obtained by using non-sorbing tracers, since the procedures for obtaining the data are more or less straightforward and are not complicated by the sorption properties that are more difficult to measure and are entangled with the diffusion properties.

In this report, most of the diffusion data measured in laboratory by diffusion method are compiled directly from original publications, and no specific databases are referred to. The reasons are: (1) So as to critically review the data and evaluate the uncertainties related to them, much more detailed information concerning the measurement conditions of the data is needed. (2) More data have emerged in recent years since the latest compilations of some databases, e.g. the data obtained by in situ electrical measurement of rock formation factor, and those data need to be included in this report.

/Ohlsson and Neretnieks 1995/ have reviewed and compiled diffusion data in crystalline rocks for the Swedish Nuclear Fuel and Waste Management Co. (SKB). /Valkiainen 1992/ has made a similar review of the diffusion data, emphasising several factors that may result in variations of the diffusivity: the rock sample thickness, the heterogeneity of the fracture surface coating and altered zone, the ionic strength and salinity of the pore water, etc.

/Brandberg and Skagius 1991/ have reviewed and compiled specifically diffusion data for engineered buffer and some data for the Finnsjön granite and other crystalline rocks in Sweden were also included. Recommended values of effective diffusivity, pore diffusivity and diffusion porosity were given for the rocks at this site. Their recommended values for effective diffusivity in granitic rocks is $1 \cdot 10^{-13}$ m^2 s^{-1}, and for diffusion porosity, 0.5%. /Ohlsson and Neretnieks 1997/ have also made similar recommendations in connection with SKB's SR 97 project. Their recommended values of effective diffusivities for different species ranges from $(0.3–1) \cdot 10^{-13}$ m^2 s^{-1} at high ionic strengths of the pore water and up to $9 \cdot 10^{-13}$ m^2 s^{-1} for Cs^+ at low ionic strengths of the pore water.

/Bradbury and Green 1985/ have studied the diffusion of iodide and Tc-99 (pertechnetate ion) in granites obtained from different regions of the UK. The rock samples included Ossian granite, Scottish Lowland granite, Skene Complex granite, and Cornish Carnmenellis granite. The method used was through-diffusion. The results of their measurements are included in Table A in Appendix I of this report for the respective species.

/Bradbury and Green 1985, 1986/ have also measured the diffusivity of iodide through a fissure surface of Cornish Carnmenellis granite and of a Canadian granite. For samples containing fissure coatings the diffusivity and porosity measured showed significant increases over the equavalent value for unaltered granitic rock. The increase in diffusivity ranged from ~ 20 to ~ 200 times, while the increases in porosity were between ~ 4 and ~ 100 times. /Skagius and Neretnieks 1986/ have also shown the same order of magnitude increase in diffusivity for fissure coatings in Swedish granitic rocks. These results are also shown in Table A in Appendix I of this report.

/Kaukonen et al. 1995, 1996, 1997/ have studied diffusion and sorption in Kivetty granite and Olkiluoto mica gneiss and pegmatite in Finland, of radioactive tracers ^{3}H, ^{22}Na, ^{36}Cl, ^{99}Tc, ^{235}Np and ^{237}Np in their chemical forms of HTO, Na^+, Cl^-, TcO_4^- (or in the more strongly sorbing form of Tc^{4+} under anaerobic condition of the experiment) and NpO_2^+ respectively. The solutions used were 10 times diluted Allard water /Allard et al. 1983/, which has similar compositions as the groundwater at the Kivetty site, and the saline Okiluoto groundwaters. The ionic strengths were 0.00085 M for ten-time-diluted Allard water, and 0.26–0.31 M for the Okiluoto groundwaters.

/Skagius 1986, Skagius and Neretnieks 1982, 1983, 1985, 1986, 1988/, and /Skagius et al. 1982/ have studied diffusion of various tracers in crystalline rock samples. By laboratory through-diffusion method, the diffusivity of non-sorbing tracers of tritiated water (HTO), Cr-EDTA, uranin (Na-fluorescein) and iodide were determined for rock samples of granite from Stripa and Finsjön, Sweden; gneiss from Karlshamn, Sweden and gabbro from Vipängen, Sweden. The sample thickness was 5 mm for the tests using HTO, and 10 mm for the tests using Cr-EDTA, uranin and iodide. For the tests using THO, both distilled water and the Allard "synthetic" water were used. For the tests using Cr-EDTA, uranin and iodide, the rock samples were first saturated with distilled water. For the tests using iodide, a concentration of 0.1 mol L^{-1} (in some tests 1.0 mol L^{-1}) of sodium nitrate ($NaNO_3$) was used. The ionic strength of the pore water during the experiment could be high (up to 1 M). The ion exclusion effect (see Section 4.2.1.) will thus be minimal. The ionic strength for the Allard water is 0.0085 M.

In granites the effective diffusivities were determined to be $(7–13) \cdot 10^{-14}$ m^2 s^{-1} for iodide and $(1.3–1.8) \cdot 10^{-13}$ m^2 s^{-1} for tritiated water. For Cr-EDTA and uranin they were $0.7 \cdot 10^{-14}$ and $0.3 \cdot 10^{-14}$ m^2 s^{-1}, respectively. More data are compiled in Table A of different species in Appendix I of this report.

The porosities of the rock samples have been measured by three different methods: (1) water saturation method, (2) tracer leaching method, and (3) the porosity determined by the intercept of the concentration-time plot at longer times for non-sorbing tracers (see "The through-diffusion method" in Section 3.2.1). For HTO and iodide, the values of the porosities obtained by the three different methods agree with each other. For Cr-EDTA and uranin, the porosity determined by the third method is lower. This was interpreted as the result of geometric hindrance of accessibility of the micropores by larger molecules (size exclusion). The porosities measured by the three different methods are listed in Table C in Appendix I of this report. The porosities measured by using the water-saturation method are also included in the table (Table A) together with the diffusivities for different species.

The results obtained by /Skagius 1986/ indicated that the porosities are higher in granites than in gneisses, and the diffusivities of all of the non-sorbing tracers used in the tests are higher in granites than in gneisses as well. This shows that, when assigning values of porosity and diffusivity in a specific site, the differences of the rock types must be considered. The results also showed that the fissure-coating materials usually have large porosities and large diffusivities.

The data on diffusivities of the sorbing tracers, Cs and Sr, have also been obtained by /Skagius 1986/ using methods of in-diffusion into crushed particles of the size 0.1 to 5.0 mm, of in-diffusion into rock pieces of the size with a diameter of 41–42 mm and a thickness of 5, 10, 32 and 40 mm. For smaller thickness of the rock pieces, the diffusivities were calculated from the variation of concentration in limited immersing solution. For larger thickness of the rock pieces, the rock samples were ground and concentration profiles inside the rock samples were obtained. The results of effective diffusivities for Cs and Sr are shown in Tables A5 and A6 in Appendix I.

Some of the formation factors shown in Tables A5 and A6 for Cs and Sr are even larger than can be accounted for by the total porosity of the respective rocks. This is believed to be caused by a transport mechanism we call "surface diffusion", in which the sorbed species in the diffuse double layer are assumed to be mobile within the sorbed phase. This is a well-known mechanism in industrial ion exchange resins. The quantification of the surface diffusion depends on the clarification and quantification of the sorption processes, and is usually difficult. Pure data fittings of the in-diffusion concentration profiles revealed that the effective diffusivity and the surface diffusivity may be of the same order of magnitude, $1 \cdot 10^{-14}$ to $1 \cdot 10^{-13}$ m^2 s^{-1}. Anyway the mechanism of surface sorption will enhance the total effect of matrix diffusion and neglecting it will position the results on a more conservative side from the viewpoint of performance assessment studies. The sorption data of the species will be reported in another report /Crawford et al. 2006/ and will not be considered here.

To simulate the in situ rock stress, diffusion experiments have also been performed under mechanical stress by /Skagius 1986/. The results showed that the diffusivity in rock samples at 300–350 bars (30–35 MPa) stress was reduced to 20–70% of the value under atmospheric pressure.

The diffusivities have also been measured by /Brandbury and Green 1986/ under confining pressures up to 16 MPa. The results indicate that at the highest pressures, there was a reduction by a factor of 2 and 2.5 of the rock formation factors compared with those obtained at ambient pressure.

/Birgersson 1988, Birgersson and Neretnieks 1988, 1990/ have made field tests of tracer diffusion in the relatively homogeneous granitic rock in the Stripa mine in Sweden at a depth of 360 m. Tracers of iodine, Cr-EDTA and uranin were circulated in a pressure of 0.9 MPa from a borehole directly into the rock matrix. This was equal to the hydraulic pressure in the packed-off sections in the borehole determined before the experiment started. The disturbance to the rock of the drilling of the borehole was minimised by drilling a small hole in the bottom of a large hole. Concentrations of the tracers that penetrated into the rock matrix were determined by overcoring of the injection hole, with several coring boreholes lying in a line and surrounding the injection hole, and subsequent leaching of the rock samples and analysing of the tracers. With their arrangement of the cores a penetration of the tracers in different directions can be investigated for a maximum penetration depth of 40 cm from the injection hole over a length of nearly 3 meters. A rock volume of more than 5 m^3 was thus covered. In this volume more than 2,500 samples were taken and analysed for tracers. Considerable heterogeneity was found over distances of tens of centimetres.

An advective-diffusive transport model was used to fit the concentration profiles to obtain pore diffusivity (D_p) of the samples. As the hydraulic conductivity is extremely low ($1 \cdot 10^{-13}$ m s^{-1}), the tracers were mainly transported into the rock matrix by diffusion. The transport porosity (ε^+) and the total pore porosity (ε_p) have not been distinguished in the model and the pore diffusivity obtained is actually $\varepsilon^+ D_p / \varepsilon_p$. Therefore data for effective diffusivity can be obtained by multiplying their values of pore diffusivity with the values of the total porosity they have measured by water saturation method. Some of the measured values were included in Table A in Appendix I of this report for respective tracers. The highest concentrations of tracers nearest the injection hole were about 1/3 of that of the original tracer solution if based on the laboratory measured porosity of the samples. This suggests that the porosity increased by about a factor 3 when the rock was de-stressed upon excavation. The porosity of the rock samples determined by the water saturation method was about 0.3%. This suggests that the in situ matrix porosity was about 0.1%.

The work of /Birgersson 1988, Birgersson and Neretnieks 1988, 1990/ clearly showed that all the three tracers used in the test migrated into the zone of undisturbed rock, and in some cases there was no obvious decrease in tracer concentrations for at least some 40 cm. It also indicated that the differences of the diffusivity could be as large as an order of magnitude between samples separated by just a few tens of centimetres. Variation of the diffusivity was large even in a relatively homogeneous rock matrix.

The composition of the groundwater at the test depth of the Stripa site has not been included in the original work. The groundwater compositions are, however, available in the literature /e.g. Nordstrom et al. 1985, Table 4.1/. The ionic strength of the water at depths between 420–460 m is about 6.0 mM.

In Table A, the data of the formation factors measured in laboratory by diffusion methods are presented for respective species, together with the rock type, the water type used, the porosity measured by, e.g. water-saturation method and the effective diffusivity. In Table B, the laboratory diffusion data of formation factors are arranged with different rock types to assist statistical analyses of the formation factor distribution of different rocks. In Table C, some data of rock porosities measured by different methods are presented.

Diffusion data have also been obtained by the through-diffusion method using He gas as a tracer /e.g. Hartikainen et al. 1995ab, 1996, 1997/. These data do not contradict the other findings.

In their studies of the effects of ionic strength of the pore water on the surface diffusion and anion exclusion, Ohlsson and co-workers /e.g. Ohlsson 2000, Ohlsson and Neretnieks 1998, 1999, 2001, Ohlsson et al. 2001/ have obtained data of matrix diffusion for Äspö diorite (in Central Sweden) and other crystalline rocks. Their data will be discussed in Section 4.2.1.

3.2 Modelling

3.2.1 Modelling used to support the data

A summary of theory of matrix diffusion

In this section, we give a brief description of the theoretical background of matrix diffusion. More detailed accounts on the theory can be found in the literature /e.g. Skagius 1986/. For more generic theory of flow and transport in porous media and fractured rock, the reader is referred to the following review articles and monographs /Bear 1972, Dullien 1992, Petruzzelli and Helfferich 1993, Bear et al. 1993, Sahimi 1995, Zheng and Bennett 2002/. Recent thorough overviews are found in /Bodin et al. 2003ab/ and /Jacob 2004/.

The mathematical theory of diffusion is based on Fick's first and second laws and mass conservation. The equation describing diffusion in a fluid is:

$$N = -D_w A \frac{dc}{dx} \qquad \text{(Equation 3.1)}$$

where

N is flow rate of diffusing species (mol s^{-1}),

D_w is diffusivity of the species in fluid (m^2 s^{-1}),

A is cross-section area of the fluid (m^2),

c is concentration of the species (mol m^{-3}),

x is distance of diffusion (m).

In a porous medium where diffusion occurs in the pore fluid the flow rate is given by

$$N_p = -D_p A\varepsilon^+ \frac{dc_p}{dx} \qquad \text{(Equation 3.2)}$$

where ε^+ is transport porosity of the porous medium and the other notations with subscript "*p*" denote the corresponding properties in the pore fluid.

The total porosity of a porous medium (ε_p) consists of a *transport* porosity (ε^+) and a *storage* porosity (ε_s).

The relation between the pore diffusivity and the diffusivity in fluid is:

$$D_p = D_w \frac{\delta_D}{\tau^2} \qquad \text{(Equation 3.3)}$$

where

δ_D is constrictivity of the pores (–),
τ^2 is tortuosity of the pores (–).

An effective diffusivity is defined as the product of the pore diffusivity and the transport porosity, i.e.

$$D_e = D_p \varepsilon^+ \qquad \text{(Equation 3.4)}$$

For sorbing species, an apparent diffusivity can be defined as:

$$D_a = \frac{D_e}{\varepsilon_p + K_d\rho} = \frac{D_e}{\alpha} \qquad \text{(Equation 3.5)}$$

where

D_a is apparent diffusivity of the sorbing species ($m^2\ s^{-1}$),
K_d is linear sorption equilibrium constant ($m^3\ kg^{-1}$),
ρ is density of the porous medium including the pore water ($kg\ m^{-3}$),
α is rock storage capacity ($= \varepsilon_p + K_d\rho$). For non-sorbing tracers, α should be theoretically equal to e_p.

The formation factor of the porous medium is defined as the ratio of the effective diffusivity to the diffusivity in bulk fluid, i.e.

$$F_f = \frac{D_p\varepsilon^+}{D_w} = \frac{\varepsilon^+\delta_D}{\tau^2} \qquad \text{(Equation 3.6)}$$

Some sorbing species diffuse also in their sorbed state (in the concentrated diffuse region of the electrical double layer). In this case, a surface diffusivity should be included in the effective diffusivity and the apparent diffusivity.

$$D_e = D_p\varepsilon^+ + K_d\rho D_s \qquad \text{(Equation 3.7)}$$

and

$$D_a = \frac{D_p\varepsilon^+ + K_d\rho D_s}{\varepsilon_p + K_d\rho} = \frac{D_p\varepsilon^+ + K_d\rho D_s}{\alpha} \qquad \text{(Equation 3.8)}$$

where D_s is surface diffusivity ($m^2\ s^{-1}$).

In sparsely fractured media like granitic rocks, the groundwater flow at the scale of interest for performance assessment of a spent fuel repository will take place through a network of fractures carrying the water from one fracture to the next. The path may thus encounter fractures with different diffusion and sorption properties. The heterogeneity of the rock must therefore also be accounted for and it is not expected that one single number can be used to characterize the diffusion properties of any one site.

A conceptual model for radionuclide transport in a fracture coupled with matrix diffusion is shown in Figure 3-1.

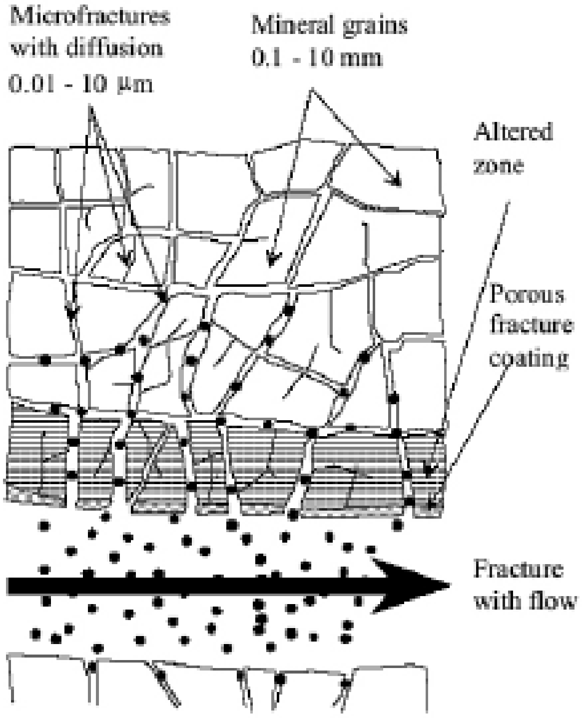


Figure 3-1. *A conceptual model for radionuclide transport in a fracture coupled with matrix diffusion /reproduced from Rasilainen 1997/.*

The through-diffusion method

In a through-diffusion experiment, a piece of rock sample of cm-lengths is placed between two containers. The containers are filled with a background solution (denoted as "water type" in the Table A in Appendix I) and the rock sample is saturated with the solution. One or more diffusing species are added into one of the two containers. The other container is usually initially devoid of the diffusing species. The variations of the concentration of the diffusing species with time in the second container are measured.

For non-sorbing tracers and sorbing tracers with a linear sorption isotherm, an analytical solution to the diffusion equation corresponding to the experimental arrangement is given in the literature /Crank 1975, p.51/:

$$\frac{Q}{Lc_1} = \frac{D_e t}{L^2} - \frac{\alpha}{6} - \frac{2\alpha}{\pi^2}\sum_{n=1}^{\infty}\frac{(-1)^n}{n^2}\exp(-\frac{D_a n^2 \pi^2 t}{L^2}) \qquad \text{(Equation 3.9)}$$

where

Q is total amount of the diffusing species accumulated in the container initially absent of the diffusing species, normalised with the cross-section area of the sample (mol m^{-2}),

L is thickness of the rock sample (m),

c_1 is initial concentration of the diffusing species in the container to which the diffusing species have been added (mol m^{-3}),

D_e is effective diffusivity of the species defined by Equation (3.4) for non-sorbing tracers and by Equation (3.7) for sorbing tracers with surface diffusion ($m^2\ s^{-1}$),

t is time elapsed (s),

D_a is apparent diffusivity defined by Equations (3.5) and (3.8) for sorbing tracers without surface diffusion and with surface diffusion, respectively ($m^2\ s^{-1}$),

α is defined as $\alpha = D_e/D_a = \varepsilon p + K_d\rho$ and is the rock storage capacity provided that there are no effects of surface diffusion.

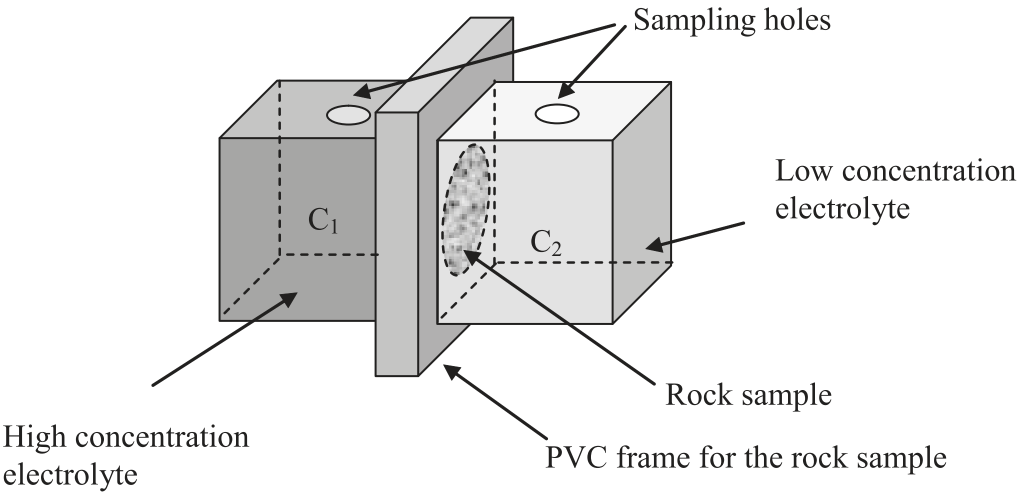


Figure 3-2. *Experimental arrangement for through-diffusion measurement.*

From the definition of α and Equation (3.5), it can be seen that, for non-sorbing tracers, α can be interpreted as the total porosity of the rock. For sorbing tracers, it is interpreted as the rock porosity plus the sorption capacity.

As t approaches infinity Equation (3.9) becomes:

$$\frac{Q}{Lc_1} = \frac{D_e t}{L^2} - \frac{\alpha}{6} \qquad \text{(Equation 3.10)}$$

This equation shows that, for relatively longer times, the accumulated concentration in the low-concentration cell will be proportional to time. In a plot of Q/Lc_1 over t the long-time part will have a linear relation, and from the slope of which the effective diffusivity, D_e, can be obtained, while the intercept will give the rock storage capacity.

For other symmetries of rock samples, e.g. a drill core cup with a cylindrical symmetry /Kautonen et al. 1997/, other analytical solutions are available for non-sorbing tracers and sorbing tracers with linear isotherms. For sorbing tracers with non-linear sorption isotherms, numerical solutions can be used to fit the experimental data. In both cases, the effective diffusivity and the rock storage capacity can be obtained.

Diffusivity and rock storage capacity can also be measured by in-diffusion method. In this method, pieces of rock samples or crushed rock samples are immersed in a limited volume of solution containing a tracer. The decrease of the tracer concentration with time can be used to fit theoretical concentration changes either analytically or numerically to obtain the sorption parameters in linear or non-linear isotherms and the effective diffusivity.

For spherical particles with linear sorption isotherm, the concentration c in the solution after time t is /Crank 1975, p. 94/:

$$\frac{c}{c_0} = 1 - \frac{M}{M_0}\left(\frac{mK_d/V}{1+mK_d/V}\right) \qquad \text{(Equation 3.11)}$$

with $$\frac{M}{M_0} = 1 - \sum_{n=1}^{\infty} \frac{6\beta(\beta+1)\exp(-D_a q_n^2 t/R^2)}{9+9\beta+q_n^2\beta^2}, \qquad \text{(Equation 3.12)}$$

$$\beta = \frac{V}{mK_d},$$

and q_n being non-zero roots of $\tan q_n - \dfrac{3q_n}{3+\beta q_n^2} = 0.$

where

c is concentration of the tracer in the limited solution (mol m^{-3}),

c_0 is initial concentration of the tracer in the solution (mol m^{-3}),

m is mass of the crushed rock sample (kg),

V is volume of the solution (m^3),

R is radius of the spherical particles (m),

K_d is sorption coefficient of the tracer (m^3 kg^{-1}),

M is total amount of solute in the spherical particles (mol),

M_0 is total amount of solute in the spherical particles at infinite time (mol).

When the steady-state is reached at longer times, the sorption coefficient K_d can be determined by setting M/M_0 in Equation (3.11) to unity. The apparent diffusivity can be determined through data fitting of the transient concentrations given by Equation (3.12) /Byegård et al. 1998/. For strongly sorbing tracers, the rock storage capacity α in Equation (3. 5) will be dominant by the sorptive storage capacity ρK_d, and the apparent diffusivity of the tracers can be determined.

For a strongly sorbing tracer, it usually takes very long time for a steady-state to be reached even for crushed particles of the size less than 1 mm /Skagius 1986/.

The conventional through-diffusion method is usually difficult to use to measure diffusion data for strongly sorbing tracers. The time for the break-through of the tracers would be extremely long even for pieces of rock samples of millimetres to a centimetre in size. In some cases, it is necessary and possible to analyse the concentration profiles of diffusing tracers inside the rock samples. /Birgersson 1988/ injected tracers directly into granitic rocks in situ and overcored the rocks around the injection hole. The concentration profiles in the overcored samples were analysed and fitted to diffusion models. When radioactive tracers are used, especially those with gamma radioactivity, it is also possible to analyse the concentration profiles inside rock samples /Kaukonen et al. 1997/.

As the penetration depth is usually short compared with the rock sample length, the semi-infinite geometry is often assumed. The concentration profile in Cartesian coordinates for a constant concentration at the inlet boundary (c_0) is:

$$\frac{c}{c_0} = \mathrm{erfc}\left(\frac{x}{2\sqrt{D_a t}}\right) \qquad \text{(Equation 3. 16)}$$

where

c is concentration at t and x from the inlet boundary (mol m^3),

x is axis along the sample thickness (m),

t is time (s).

For other symmetry, analytical solutions are also available.

When the experimentally determined concentration profiles are fitted to some theoretical profiles, only the apparent diffusivity can be obtained. The rock storage capacities have to be determined separately.

Electromigration experiments with tracers

As diffusion is such a slow process, the lengths of the rock samples used in through- diffusion experiments are generally limited to only a few centimetres. By placing an electrical potential gradient over the rock sample, the tracer flux is considerably increased /Maes et al. 1999/. A prerequisite for this is that the tracer is ionic. If the tracer is anionic, the anode is placed in the low concentration electrolyte and the cathode is placed in the high concentration electrolyte.

When connecting the electrodes to a direct current source, water is dissociated and the following electrode reactions occur:

Anode: $H_2O \Rightarrow 2H^+ + {}^1\!/_2\, O_2(g) + 2e^-$ (Equation 3.17)

Cathode: $2e^- + 2H_2O \Rightarrow 2OH^- + H_2(g)$ (Equation 3.18)

The water dissociation gives rise to pH changes in the electrolytes and there are a number of disadvantages associated with this:

1) Some ionic tracers, for example uranin, may become protonated or deprotonated if the pH changes. This will change the overall charge of the species.
2) If a rock sample is subjected to pH changes, the minerals may become altered and this may affect the transport properties of the rock.
3) The efficiency of transferring current from the electrode surfaces to the electrolytes decreases if pH changes are allowed in the electrolytes.

By intermixing the anode and the cathode electrolytes, the protons and hydroxyl ions that are formed at the electrode surfaces neutralise each other. To avoid short-circuiting in the mixing streams, drop-wise mixing is required. A consequence of such mixing is that the electrolytes holding the tracers must be separated. To meet these demands a new experimental cell was developed, the through-electromigration cell /Löfgren 2001/. The cell is shown in Figure 3-3.

In Figure 3-3 the intermixing is carried out in a separate mixing vessel. An ampere meter measures the current through the cell. Furthermore, a voltmeter that is connected to separate potential electrodes measures the potential drop over the rock sample. In the experiments described here, titanium plates were used for the current electrodes and titanium nets for the potential electrodes. The through-electromigration cell also requires filters of low porosity and low permeability between the tracer electrolyte and the anode and cathode electrolyte compartments. If the filters have approximately the same formation factor and thickness as the rock sample, the overall electromigratory tracer flux N_μ into the low concentration electrolyte is:

$$N_\mu = \mu_e (C_H - C_L) \frac{dU}{dx} \qquad \text{(Equation 3.19)}$$

where

μ_e is effective ionic mobility ($m^2\ V^{-1}\ s^{-1}$),
C_H is tracer concentration in the high concentration electrolyte ($mol\ m^{-3}$),
C_L is tracer concentration in the low concentration electrolyte ($mol\ m^{-3}$),
dU/dx is the potential gradient over the sample($V\ m^{-1}$).

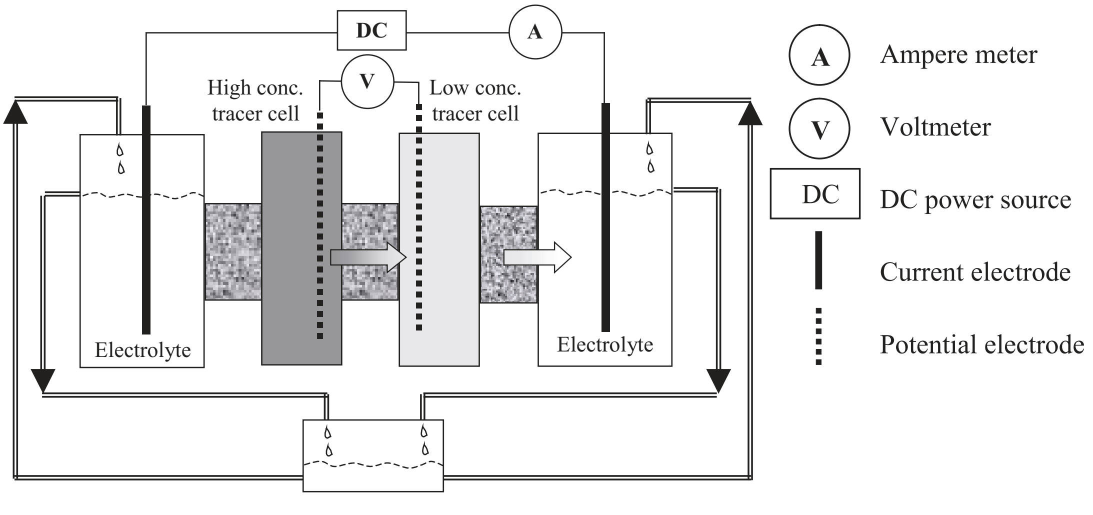


Figure 3-3. *Schematics of the through electromigration cell.*

As $C_H \gg C_L$, the tracer flux out of the low concentration electrolyte can be neglected. In Equation (3.19) diffusion and electroosmosis are neglected. The diffusive flux will be much smaller than the electromigratory flux. The effect of the electroosmotic flux has been investigated and has been found to be small enough to be neglected /Löfgren 2004/. By knowing the electromigratory flux through the cell, the concentrations and the potential gradient over the cell, the effective ionic mobility can be found. From this the effective diffusivity D_e can be obtained by using the Einstein relation:

$$D_e = \mu_e \frac{RT}{Fz} \qquad \text{(Equation 3.20)}$$

By using Equation 3.6 the formation factor can be obtained.

Electrical conductivity methods

In a through-electromigration experiment, most of the current I is carried by the background ions, for example Na^+ and Cl^-.

$$I = -F_f \frac{AF^2}{RT} \sum_i z_i^2 C_i D_{w,i} \frac{dU}{dx} \qquad \text{(Equation 3.21)}$$

where

F_f is formation factor(–),
A is cross section area of the rock sample (m^2),
F is Faraday constant ($C\ mol^{-1}$),
R is gas constant ($J\ mol^{-1}\ K^{-1}$),
T is temperature (K),
z_i is charge number (–),
$D_{w,i}$ is diffusivity of the species in free solution ($m^2\ s^{-1}$),
C_i is concentration of the species in the electrolyte ($mol\ m^{-3}$).

These ions pass through the same porous system as the tracers. Therefore, the background ions can be used instead of tracers for obtaining the formation factor. After having equilibrated the pore water of the rock sample with an electrolyte of known electrical conductivity, the electrical conductivity of the rock is measured using direct current. From Equation (3.21) it can be shown /Löfgren 2004/ that the formation factor is the ratio of the electrical conductivities of the pore water and the rock. As the electrical conductivity is the reciprocal of the resistivity:

$$F_f = \frac{\rho_w}{\rho_r} = \frac{\kappa_r}{\kappa_w} \qquad \text{(Equation 3.22)}$$

where

κ_w is pore water electrical conductivity ($S\ m^{-1}$),
κ_r is rock electrical conductivity ($S\ m^{-1}$),
ρ_w is pore water resistivity (ohm m),
ρ_r is rock resistivity (ohm m).

The experimental set-up shown in Figure 3-3 can be used in these measurements. However, no separate tracer electrolyte compartments are needed.

Instead of mixing the electrolytes, one can use alternating current. If the current changes direction a number of times per seconds, the volume of the electrolytes can be reduced considerably. The frequencies used in the laboratory are typically a few hundred Hz or less, which is low enough not to cause significant capacitance effects /Löfgren 2001/. If the electrical conductivity of the pore water is known, the formation factor is obtained by measuring the rock resistivity and using Equation (3.22). In the measurements performed, two thin sponges soaked in electrolyte were placed between the electrodes and the rock sample (Figure 3-4).

A comparison of the different laboratory methods can be found in /Löfgren and Neretnieks 2006/.

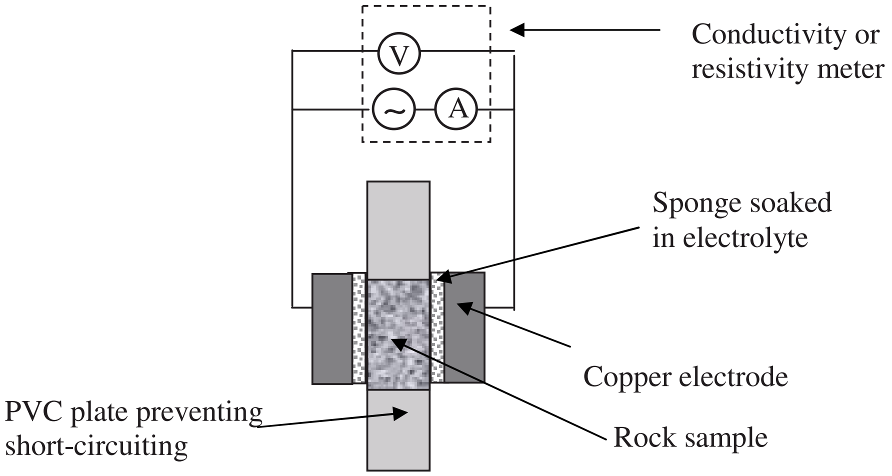


Figure 3-4. *Experimental set-up of rock resistivity measurements using alternating current.*

In situ methods

The in situ method of obtaining formation factors by electrical methods is analogue to the laboratory electrical conductivity method using alternating current. Instead of bringing the rock sample to the laboratory, a tool measuring the rock resistivity is lowered down the borehole. Figure 3-5 shows the current lines emanating from such a tool. For more information on relevant geophysical in situ tools, please consult /Löfgren and Neretnieks 2002/.

As one cannot equilibrate the pore water of the rock matrix in situ with a solution of known electrical conductivity, one has to assume that the pore water is already in equilibrium with the freely flowing groundwater at a corresponding depth. Figure 3-6 shows the Posiva difference flow meter. A drawdown is used at the surface. By packing off specific fractures, groundwater can be withdrawn from these fractures and its electrical conductivity can be measured.

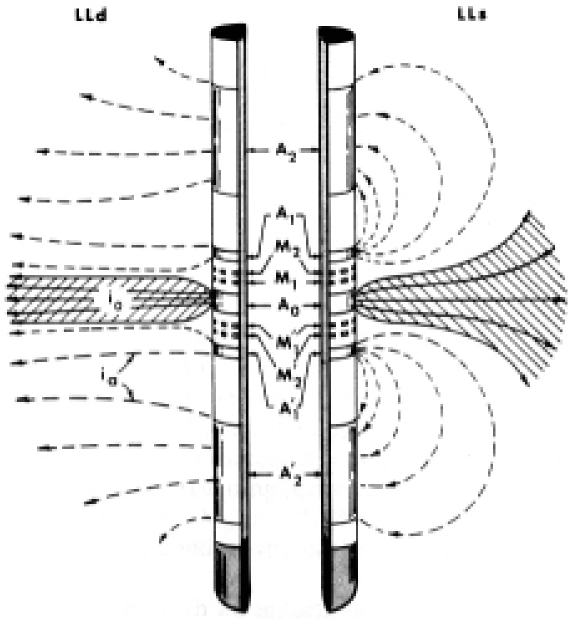


Fig. 3.8. — Principle of the Dual Laterolog.
(*Schlumberger document*)

Figure 3-5. *Schematics of an in situ rock resistivity tool.*

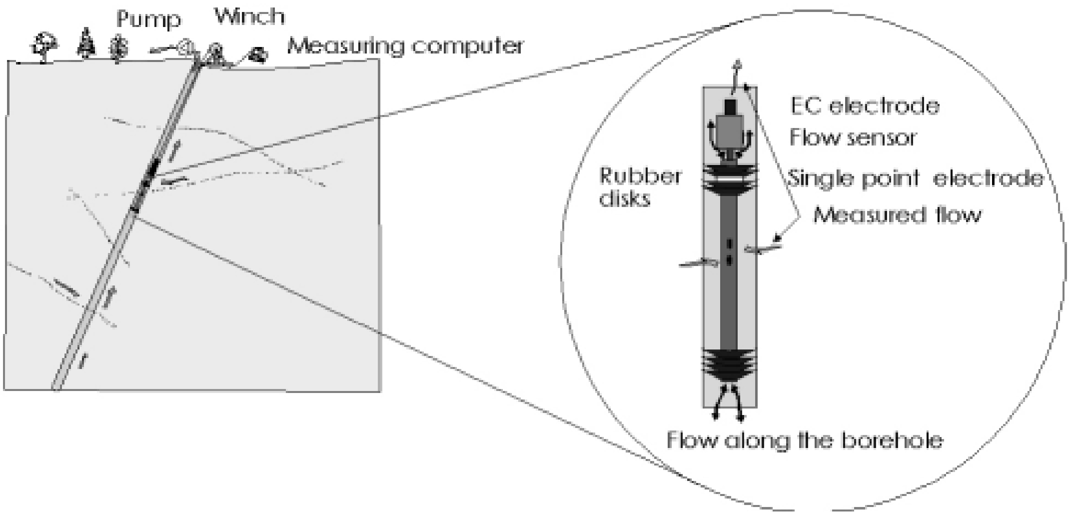


Figure 3-6. Schematics of the Posiva difference flow meter /taken from Rouhiainen and Pöllänen 2003/.

For more details on obtaining in situ formation factors by electrical methods, please consult /Löfgren and Neretnieks 2005ab/.

Other remarks

There are some assumptions in the above treatment that may not always be fulfilled. Some of the more important are discussed below. A more detailed discussion of these issues can be found in Section 4.2.1.

1. The pore system is essentially the same throughout the sample. This is not fulfilled for short samples where normally dead-end pores may have opened to the sample surface. Also the disturbances induced by sample preparation are more dominant near the surface. Measurements with different sample lengths are used to check this effect. Often less than one centimetre disturbance from the surface can be seen. This is not a serious problem for electrical conductivity measurements where long samples can be readily used. For through-diffusion measurements this effect has some influence as rarely longer samples than one or a few centimetre are used. For instationary measurements with (strongly) sorbing tracers the problem is serious because the tracers will encounter essentially only the disturbed region.
2. Surface diffusion and ion exclusion effects can have a strong influence on the rate of transport through a sample in low salinity waters. This can be avoided by using highly saline water to impregnate the samples for laboratory testing and formation factor determinations. The effects themselves can be assessed by impregnating the pores of the samples with low salinity (distilled) water and comparing the transport of the species of interest with the high salinity results. This can be done in the laboratory. The problem of knowing the pore water salinity in the matrix around a deep borehole has as yet not been solved. We rely primarily on the assumption that the matrix pore waters are essentially the same as those that can be extracted from nearby fractures in the same boreholes. Supporting evidence is gained from measuring the electrical conductivity of the drill cores from the same location with the original pore water composition and with saline water impregnated cores. If no large difference is obtained it indicates that at least not very non-saline waters were present in the matrix. The presence of non-saline water in the pore would lead to over-estimation of the formation factor. In situ formation factors are not evaluated for the upper 100 to 200 m where the waters in the fractures were of lower salinity.

3. The assumption that electrical conductivity can be used to determine diffusion properties is well established for bulk water. It has been questioned whether it is true also for the narrow pores between and within the mineral crystals of the rocks. Many of the minerals have fixed charges in addition to the surface charges that develop when the surfaces are hydrolysed by the water. Some minerals are electrically conducting and could transport current in addition to that by the ions in the water. A number of different types of test have been performed to check these concerns. It has been found that very similar values of the formation factor are found on the same samples when using 1) through-diffusion, 2) electrical conductivity in an AC field, 3) electrical conductivity in a DC field, and 4) through-migration of ions subject to a DC electric field. For a detailed discussion on these experiments and their conclusions see /Löfgren 2004/. The conclusion is that electrical conductivity in an AC field gives correct information on the formation factor.
4. Possible clogging of pores due to chemical precipitation. This could happen if water with a different composition invaded the fractures and precipitates formed. This does not seem to have happened more than perhaps locally over the age of the Swedish rock as seen by the down-hole electrical measurements because the majority of measurements in the deep holes at the investigated sites were found to be conductive. Further, most mineral alterations found in samples give more porous systems.

We conclude that the down-hole measurement method used to determine the formation factor is robust and gives data that are correct for use in PA modelling.

4 Evaluation of sources affecting uncertainty

4.1 Overview

4.1.1 Experts view

The concepts and theoretical background for the determination of rock porosity and matrix diffusion data of crystalline rocks are well founded. The various diffusion methods, especially the through-diffusion method, are conceptually and theoretically straightforward. The porosity and diffusivity are both rock properties and should in principle be the same whichever tracers one uses to determine them.

In practice, however, due to the relatively slow rate of mass transport in the rock matrix, one is usually subject to various limitations in determining the porosity and diffusivity. With direct diffusion measurements, the length of rock samples can often be no longer than a few centimetres. When samples are taken from drill cores to the laboratory, the original lithostatic stress and hydrostatic pressure are released. Drilling and preparation of the samples inevitably introduce damaged zones at the surface of the samples.

The newly developed electrical conductivity measurement has been demonstrated to be able to measure rock formation factors in situ using either DC current or low-frequency AC current. Certain issues need to be addressed concerning the reliability of this method. Some of them have been discussed at the end of Section 3.2.1.

Even though matrix diffusion is essentially a physical phenomenon, the numerical values of the diffusivity can still vary when different tracers are used to measure them. The relative sizes of the species may have an effect. Larger species may not be able to migrate through micropores smaller than their sizes. Charges on mineral surfaces developed due to isomorphic substitution inside the crystal structure of the mineral often result in a diffuse double layer in the pore water adjacent to the mineral surfaces. This diffuse double layer may have a strong impact on the diffusion of charged species in the pores of the rocks.

More detailed accounts of the uncertainties will be given in the next section.

4.2 Uncertainty related to different conditions

4.2.1 Conditions affecting matrix diffusion and porosity

Some diffusion data determined both by through-diffusion and in-diffusion methods in laboratory are summarised in Table A in Appendix I. The formation factors obtained by different researchers all over the world for different crystalline rocks are compiled in Table B. The data are also summarised in Figures 4-1 to 4-3.

It can be seen from the tables and figures that the data have large variations, over several orders of magnitude. The other data of rock formation factors measured by in situ electrical method are presented in figures in Chapter 5. They also show orders-of-magnitude variations. These large variations are direct reflection of the heterogeneity of natural rock formations. It should be noted that these large variations are centimetre to decimetre scale variations, determined by the sample size for the laboratory data, and by the sizes of the tool and its measurement section for the in situ data. Over larger scales, as are usually used in flow and transport models in performance assessment, the variations may be evened out to a much smallar degree, as is discussed in Section 2.2.

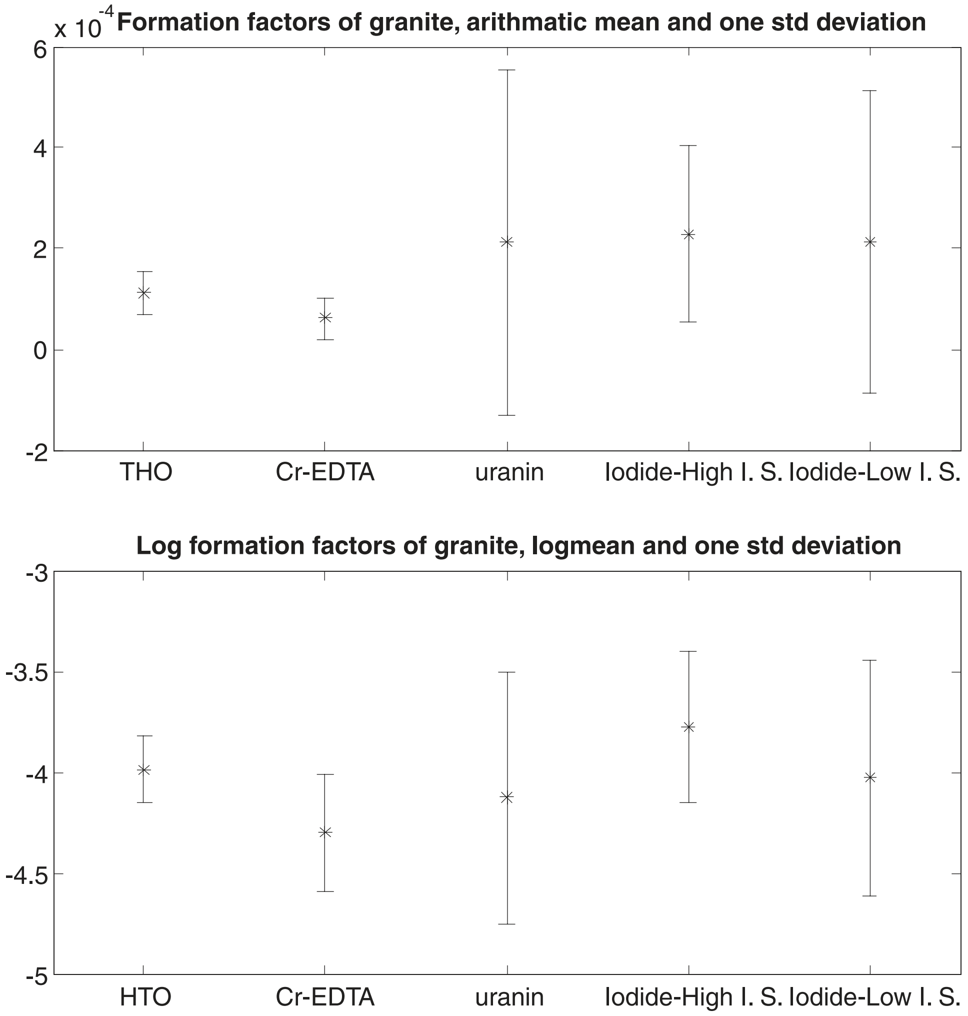


Figure 4-1. *Formation factors of granite determined by using different non-sorbing tracers in laboratory diffusion measurement. The tracers are tritiated water (THO), Cr-EDTA, uranin, iodide in solution of high ionic strength (0.1M) and iodide in solution of low ionic strength (0.003–0.007 M). (upper: arithmetic mean with one standard deviation; lower: logarithmic mean of base 10 with one standard deviation) (Data from Table A in Appendix I).*

Within the large range of variations, certain uncertainties related to the data can still be identified. For the laboratory data measured by through-diffusion and in-diffusion methods, there will be the following uncertainties:

- Uncertainties related to rock stress.
- Uncertainties related to excavation damages and sample preparation.
- Uncertainties related to rock sample size.
- Uncertainties related to subjective sampling of the rock samples.
- Uncertainties related to properties of different tracers.

There are also other uncertainties related to specific tracers. The different uncertainties will be discussed in the following subsections. Some have already been touched upon in the previous sections in the discussion of model assumptions underlying the interpretation of measurements.

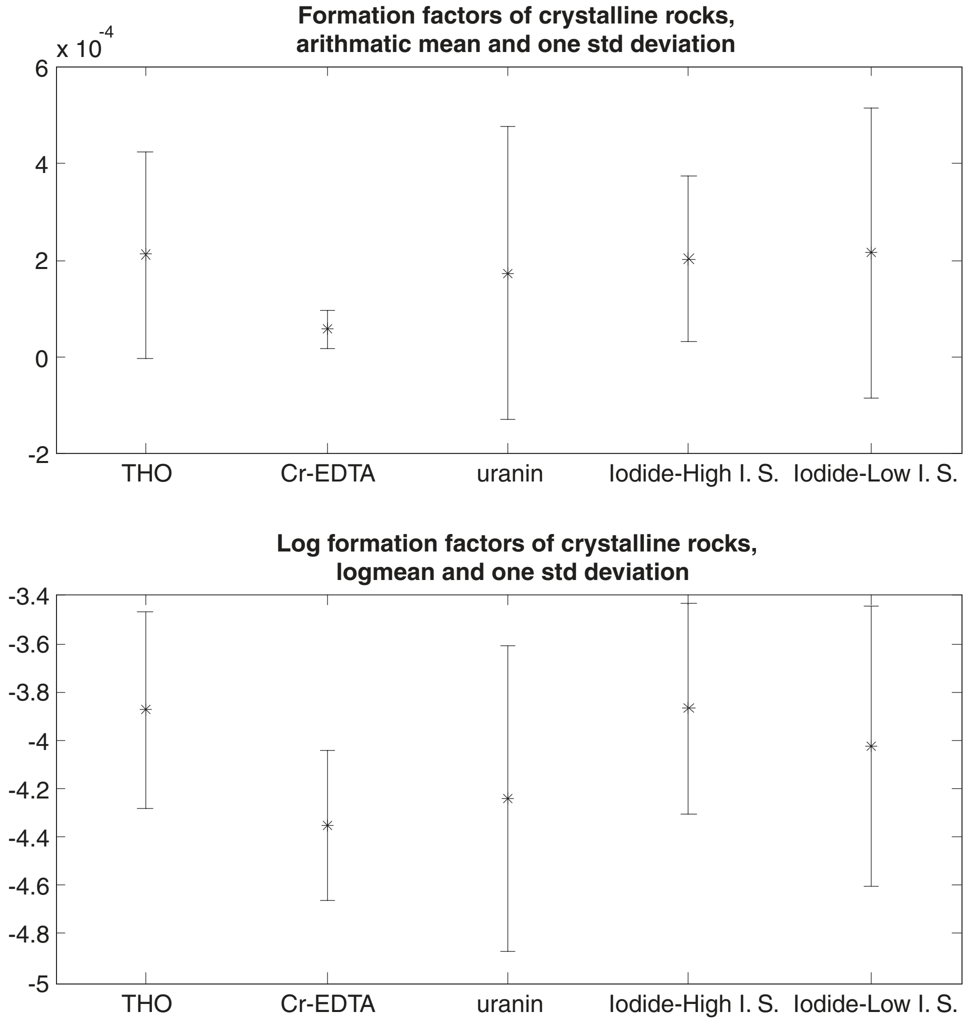


Figure 4-2. *Formation factors of different crystalline rocks (including granite) determined by using different non-sorbing tracers in laboratory diffusion measurement. The tracers are tritiated water (THO), Cr-EDTA, uranin, iodide in solution of high ionic strength (0.1M) and iodide in solution of low ionic strength (0.003–0.007 M). (upper: arithmetic mean with one standard deviation; lower: logarithmic mean of base 10 with one standard deviation) (Data from Table A in Appendix I).*

Uncertainties related to rock stress

At repository depth the rock stress is usually high. The hydraulic pressure of the groundwater will be roughly 5 MPa, and the rock static stress (both horizontal and vertical) will be 20 MPa or larger /SKB 1999/. When the rock samples are taken to the laboratory, they are subjected only to ambient pressure. The de-stressing of the rock sample may result in opening of some micropores and thus the diffusivity may increase.

/Skagius and Neretnieks 1986/ attempted to simulate in the laboratory the stress existing in situ due to the overburden. This was done both in through-diffusion and in electrical conductivity experiments. Granite and gneiss rock samples 10 cm thick from different locations in Sweden were examined. The whole experimental cell was placed in a pressure vessel where pressures up to 35 MPa could be obtained. It was found that the diffusivity of the rock samples was reduced to 20–70% under pressure. Similar results were obtained using granite samples from the United Kingdom by /Bradbury and Green 1986/. According to /Miller et al. 1994/ the reduction in effective diffusivity found by Skagius and Neretnieks should be treated as a minimum value as

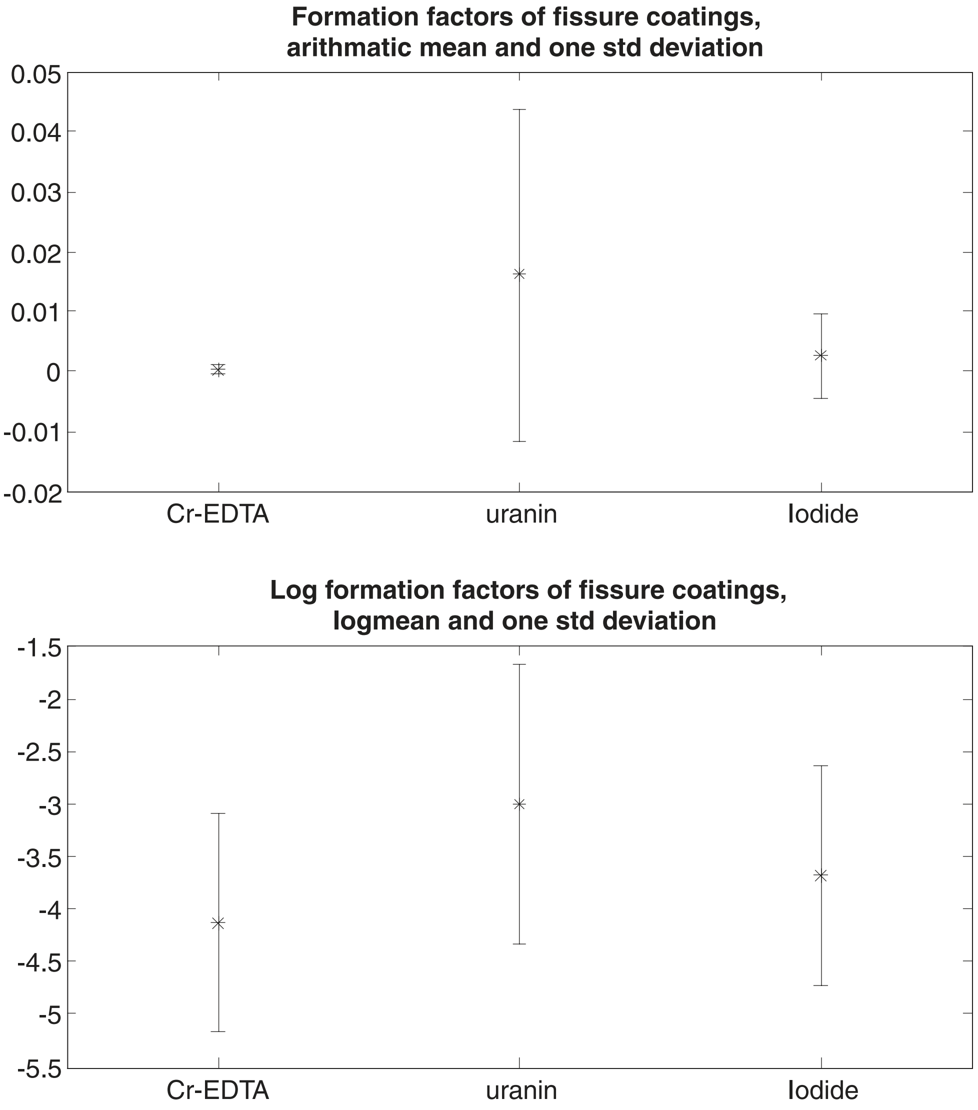


Figure 4-3. *Formation factors of fissure coatings (including a layer of fissure coating on granite) determined by using different non-sorbing tracers in laboratory diffusion measurement. The tracers are Cr-EDTA, uranin, and iodide (upper: arithmetic mean with one standard deviation; lower: logarithmic mean of base 10 with one standard deviation) (Data from Table A in Appendix I).*

the study was conceptually simplistic, assuming that re-stressing core samples with simulated overburden pressures would represent undisturbed in situ conditions. De-stressing, sample handling and re-stressing will always induce new pores and fractures which will not be closed by reversing the processes.

The effect of de-stressing of rock samples could thus be larger than the value obtained by /Skagius and Neretnieks 1986/. The in situ electrical method of borehole logging discussed later in this section reveals that the effect could be as large as one order of magnitude or more.

Uncertainties related to excavation damages and sample preparation

/Autio et al. 1999/ examined the excavation damage induced when drilling experimental full-scale deposition holes in the Research Tunnel at Olkiluoto in Finland. It was found that the porosity of the excavation damaged zone (EDZ) was increased many times and that it extended about 10 mm into the rock samples. Figure 4-4 shows the obtained porosity profiles where the porosity is normalised to the average porosity of the undisturbed rock, which was 0.14%.

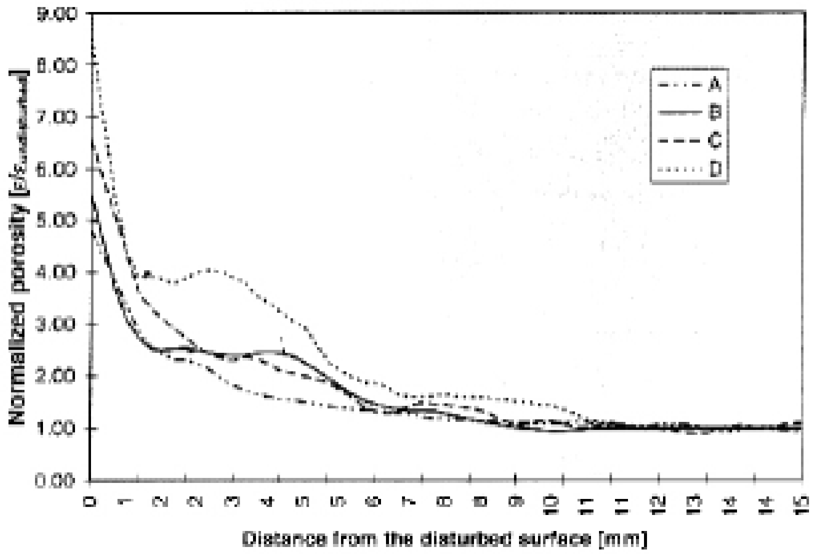


Figure 4-4. *Porosity profiles of excavation disturbed zone /from Audio et al. 1999/.*

In order to examine to what extent such an EDZ could influence through-diffusion and electrical conductivity measurements suggested for the Swedish site investigation program a very simplistic model was made. The profiles obtained by /Autio et al. 1999/ were simplified into a case where the core was divided into an excavation-damaged zone of 4 sections of different porosities and the undisturbed rock (Figure 4-5).

It was assumed that an absolute rather than a relative porosity was induced by the excavation. By subtracting the porosity of the undisturbed rock the excavation induced porosity was obtained. By assuming a constant tortuosity to constrictivity ratio of 20 throughout the EDZ, excavations induced formation factors for the different sectionscould be obtained. The products of the respective section area and formation factor were divided by the total sample area and summed. This gives an average excavation induced formation factor of about $3.7 \cdot 10^{-5}$. This is in the same order of magnitude as the total formation factor measured in many through-diffusion and electrical conductivity experiments. It is admitted that it may be unfounded to use the same excavation damage for the core drilled boreholes as for the full-scale deposition holes and that the reasoning above is overly simplified. It is recommended to increase the knowledge of the disturbed zone caused by the core drilling in the site characterisation.

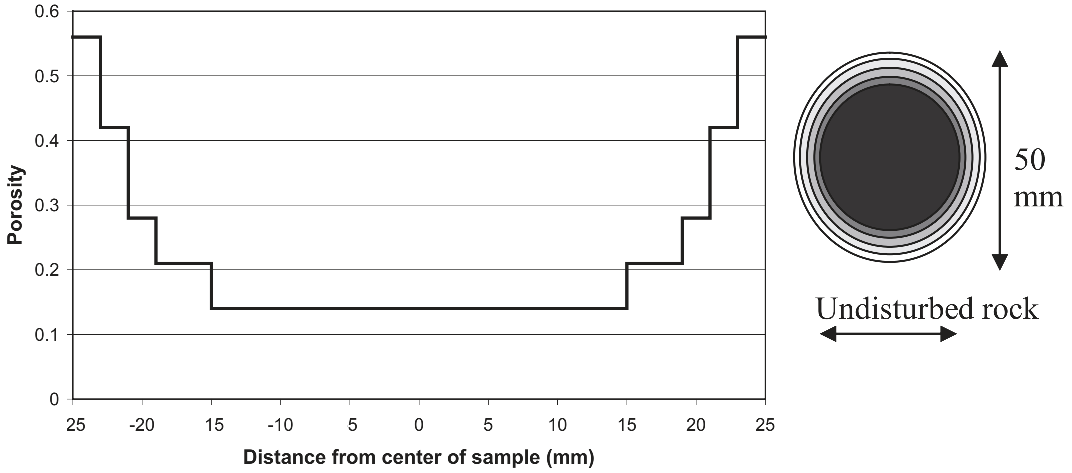


Figure 4-5. *Simplified porosity profiles of excavation disturbed zone.*

Uncertainties related to rock sample size

Conceptually, the diffusion accessible porosity can be divided into transport porosity and dead-end porosity. Diffusion length up to 5–6 mm was suggested for the dead-end pores /Bradbury et al. 1982/. If using short samples (~ 1 cm) a significant number of the dead-end pores may be connected and this would lead to overestimations of the effective diffusivity. If using even shorter samples of less than a few millimetres the effect of the tortuosity and constrictivity will decrease and the obtained formation factor will approach the total porosity. /Valkiainen et al. 1996/ investigated how the obtained effective diffusivity varied with sample length in Finnish granite. A decrease in the importance of dead-end pores, and thus also a decrease in connectivity, was observed up to sample lengths of 4 cm. From the results it was concluded that 2 cm samples would represent a larger rock mass well enough. Similar decreases of effective diffusivities of HTO and $^{22}Na^{+}$ in Äspö diorite have also been observed by /Byegård et al. 1998/. However, in the investigation of /Ohlsson 2000/ a relatively constant diffusivity was obtained for all of the lengths from 1–15 cm that were investigated.

Due to the very long time required for the steady-state to be reached in laboratory diffusion experiments using strongly sorbing tracers, very thin and even crushed rock samples have to be used. The rock samples may thus be not representative of the status of the rock at repository depth. Crushing of rock samples and preparation of very thin rock samples by sawing may introduce new surface areas and open some dead-end pores to transport pores. Also the sorption properties of the experimental tracers on the crushed particles may be significantly different from those on large samples of rock pieces /Crawford et al. 2006/. The derivation of the diffusivities will be complicated by uncertainties in determining the rock storage capacity. For this reason and others, the data of formation factor compiled in Table B are mainly obtained from laboratory through-diffusion measurements using non-sorbing tracers.

It seems that size of the rock samples used in laboratory experiment may influence the diffusivity measurement in some cases but not in others. The issue may depend on different rock type, different tracers used and it is difficult to quantify the pertaining uncertainties. The example given by /Byegård et al. 1998/ could imply that the effect is 2–3 folds going from less than 1 cm to 4 cm of rock samples.

Uncertainties related to subjective sampling of the rock samples

The pieces of rock samples used in laboratory diffusion experiment are obtained from drill cores of different boreholes. For practical reasons, the samples are usually taken from a very short section of the drill core from one borehole. Sometimes they are just taken consecutively from a section of the core. Often sections of the rock are selected subjectively to contain "good" rock that can be easily worked with to prepare pieces of samples for later test usage. The number of rock samples taken from a borehole is usually limited. The results thus obtained may not be statistically representative of the rocks intersected by the borehole at a certain site.

The Stripa in situ diffusion experiments discussed earlier and in situ electrical conductivity measurement reveal that large variations of the rock formation factor may occur along a drilled borehole. These large variations may not be readily observed when just a limited number of samples obtained from a short section of the borehole is tested in laboratory.

Uncertainties related to properties of different tracers

The species of primary concern in the performance assessment of a spent fuel repository are fission products and actinides, for example ^{137}Cs, ^{90}Sr, ^{129}I, ^{99}Tc, and different isotopes of U, Np, Pu and Am. For practical reasons, like readily available methods for chemical analysis, other species are also used (and more often) in the laboratory through- and in-diffusion methods for measuring the diffusion properties.

The non-sorbing species Cr-EDTA and uranin (Na-fluorescein) have relatively large molecular sizes compared with inorganic ionic species. Smaller pores in the rock samples may not be available for the diffusive transport of them when they are readily available for other species of smaller molecular sizes. The rock formation factors measured by using these and other large species are usually lower than values measured using other species, as was shown in /Skagius 1986/. The deviations are difficult to quantify because of the large natural variations of the formation factors but most of the times they may well be less than one order of magnitude.

The minerals and the surfaces of most rocks including crystalline rocks are charged. For crystalline rocks the charges are usually negative. In contact with pore water, the negative charge on the surface may result in a diffuse double layer in the pore water adjacent to the surface. Anions in the pore water will be excluded from accessing the region of the pore near the mineral surface due to electrostatic interaction. This phenomenon is termed *anion exclusion* /Valkiainen et al. 1995, Olin et al. 1997, Ohlsson 2000, Ohlsson and Neretnieks 1998, 1999, 2001, Ohlsson et al. 2001/. The thickness of the diffuse double layer depends on the ionic strength of the pore water. The thickness increases with decreasing ionic strength. When the ionic strength of the pore water is low, the formation factors measured by using anions like I^- and TcO_4^- may be smaller than those measured when the water has a high ionic strength. This effect is difficult to quantify because, at present, there is still a lack of understanding of the mechanisms of the electrostatic interaction between the ions and the diffuse double layer within the pore water.

Another phenomenon closely related to the charges of the diffusing species is *surface diffusion*. It was observed that, by using some sorbing cations in diffusion measurement, the effective diffusivities obtained are larger than it should be when the batch sorption data and the porosity of the rock are accounted for (See Section 3.1.1). This is usually explained as that, in the sorbed phase, the species are still mobile and can diffuse within the sorbed phase. This effect is more obvious when the ionic strengths of the pore water are low and the amount of the species in the sorbed phase becomes comparable with the amount in the pore water and thus dominates the total diffusive transport through the pore. This is an even less understood process and it would be impossible at present to quantify the effects for different cations. Surface diffusion is only expected for alkali and alkaline earth elements that are primarily sorbed by ion exchange and not by surface complexation mechanisms.

4.3 Conceptual uncertainties

We have not found that there are any conceptual uncertainties that could have a serious influence on the proposed data on the formation factor and porosity for use in PA modelling.

4.4 Data Uncertainty, spatial and temporal variation

4.4.1 Spatial, temporal and site specific variation

Spatial variation of the formation factor is evidenced in the downhole measurements /Löfgren and Neretnieks 2005ab/.

Temporal variation: variation of matrix diffusion and porosity is not expected to have a major influence.

Site specific variations are obtained directly as a large number of deep boreholes are measured with high resolution.

4.4.2 Lack of knowledge, errors etc

The unknown pore water composition directly induces an uncertainty in the evaluation of the formation factor. This effect can be important if the pore water actually has a low salinity when it is assumed that it has a high salinity based on water extracted from nearby flowing fractures. Partly this can be remedied by measurements of the cores at the same locations. No deeper analysis of this has been made. Some correlation to rock types is indicated subjectively. We suggest that formation factors vary considerably within distances of a metre or less. See /Löfgren 2001/.

4.5 Correlations

4.5.1 Correlations among matrix diffusion and porosity data

Porosity and matrix diffusion is positively correlated but variations are very large /Löfgren, 2001/. There is a correlation to be expected between rock stress and porosity as well as rock stress and matrix diffusion. It has not been possible to quantify the first as there are no in situ measurements of porosity.

5 Quantification of data and uncertainties

5.1 Methods and approaches for uncertainty estimation

5.1.1 General approach

The formation factor in Swedish crystalline rock is reported to range over many orders of magnitude. This should be compared with the uncertainty of a few factors induced in the results when measuring the entity with various laboratory or in situ methods. Unless there is a substantial systematic error in the measurements, experimental uncertainties will have a limited significance on the formation factor distributions presented in this report.

The systematic errors arising from the methodologies described in Chapter 3 normally do not exceed a factor of two. This is valid both for methodologies based on diffusion and electromigration. As neither diffusion nor electromigration experiments, in general, are evaluated using proper equations for multicomponent systems, it is hard to assess whether diffusion or electromigration methods gives the "least" error. This relates both to systematic and other errors.

It is also hard to assess whether laboratory or in situ methods gives the "least" error. This relates both to systematic and other errors. Laboratory methods suffer from the fact that the rock is altered when taken to the laboratory. In situ methods suffer from the fact that the experimental conditions are less well known.

As the natural variation of the formation factor is assessed to be much greater than the experimental uncertainties, the focus of the uncertainty estimation in this report is on the natural variation of the entity. It is assumed that the experimental data, which the formation factor distributions presented in this report are based upon, are "correctly measured".

Temporal variations are assessed to be insignificant. The spatial distribution of the formation factor appears to correlate with the spatial distribution of different rock types, as well as with other conditions stated in Chapter 2. The general approach of estimating uncertainties is therefore to vary different conditions and to discuss how the resulting formation factor distributions are affected.

The general approach for estimating uncertainties of obtained porosities is analogous to that of formation factors, discussed above. However, porosities appear to range over only a few orders of magnitude and that the experimental uncertainties appear to be smaller.

The variation for both the formation factor and the porosity will be smaller when averaged over a block having a size larger than the correlation length of the heterogeneity. This is important to keep in mind when using the results presented in this chapter in performance assessments. Exactly how this should be done is not known at present. This issue is discussed in Chapter 2.

5.2 Conditions for which parameter values are to be supplied

5.2.1 Conditions for site specific and generic data

SKB is currently pursuing site investigations for a deep repository in the municipalities of Östhammar and Oskarshamn. Therefore, analyses of site-specific data from the site investigation areas in these two municipalities are presented as the main results in this report.

The analysed data are taken from the subareas Forsmark in Östhammar and Simpevarp and Laxemar in Oskarshamn. The formation factor data obtained in Forsmark are taken from the boreholes KFM01A and KFM02A. The porosity data obtained in Forsmark are taken from the boreholes KFM01A, KFM02A, KFM03A, KFM04A, and KFM05A. The formation factors and porosities obtained in Simpevarp are taken from the boreholes KSH01A and KSH02.The formation factors and porosities obtained in Laxemar are taken from the borehole KLX02.

For a description of the sites and boreholes, from where the data are taken, please consult Chapter 3 of this report. The formation factor data are obtained both in the laboratory and in situ. The laboratory results have either been obtained by diffusion or electromigration methods. The in situ data have been obtained by electromigration methods. The porosity data have been obtained in the laboratory, mostly by immersion techniques.

When compiling site-specific data, all data available at the conditions shown in Table 5-1 have been used.

A full sensitivity analysis, when varying all conditions simultaneously, is outside the scope of this report. However, the impact on the formation factor and/or porosity when varying one condition at a time is discussed. The conditions that are varied are described in Section 5.2.2. Of the conditions shown in Table 5-1, variations of the temperature were deemed not to have a great impact on the resulting formation factor profiles.

In addition to the site-specific data, generic data are presented for comparison in this report. All generic data presented have been obtained by traditional through-diffusion experiments. The data presented are mainly obtained on Swedish rocks, but some international data are presented for comparison. Formation factor distributions are presented for crystalline rocks as a group as well as for granite alone.

Table 5-1. Conditions for site-specific data.

	Formation factor Laboratory	In situ	Porosity Laboratory
Rock type	All rock types present included	All rock types present included	All rock types present included
Rock type heterogeneity	All rock type heterogeneities present included. Sample size ~10^{-4} m^3	All rock type heterogeneities present included. Sample size ~1 m^3	All rock type heterogeneities present included. Sample size ~10^{-4} m^3
Fracture heterogeneity	Only non-fractured or sparsely fractured rock used	All fracture heterogeneities present included	Only non-fractured or sparsely fractured rock used
Groundwater chemistry	All groundwater chemistry used in the laboratory included	All groundwater chemistry present in situ below transition included	Not influenced by groundwater chemistry
Temperature	All room temperatures used in the laboratory included	All temperatures present in situ included	Not influenced by temperature
Degree of alteration	All degrees of alteration present included	Only rock more than half a meter away from any fractures. Therefore, mostly undisturbed rock	All degrees of alteration present included
Rock stress	Atmospheric pressure included	All rock stresses present in situ included	Atmospheric pressure included

Table 5-2. Conditions for generic data.

	Formation factor Crystalline rocks	Granite	Porosity Crystalline rocks
Rock type	All data found on crystalline rocks included	All data found on granite included	All rock types present included
Rock type heterogeneity	All rock type heterogeneities present included. Sample size ~10^{-4} m^3	All rock type heterogeneities present included. Sample size ~10^{-4} m^3	All rock type heterogeneities present included. Sample size ~10^{-4} m^3
Fracture heterogeneity	Only non-fractured or sparsely fractured rock used	Only non-fractured or sparsely fractured rock used	Only non-fractured or sparsely fractured rock used
Groundwater chemistry	All groundwater chemistry used in the laboratory included. Only non-sorbing tracers included.	All groundwater chemistry used in the laboratory included. Only non-sorbing tracers included.	Not influenced by groundwater chemistry
Temperature	All room temperatures used in the laboratory included	All room temperatures used in the laboratory included	Not influenced by temperature
Degree of alteration	All degrees of alteration present included	All degrees of alteration present included	All degrees of alteration present included
Rock stress	Atmospheric pressure included	Atmospheric pressure included	Atmospheric pressure included

5.2.2 Conditions impacting the formation factor and porosity

Rock type

The rock type has been identified as an important condition both in Chapter 2 and Chapter 4 of this report. Earlier publications suggest that both the formation factor and the porosity may vary over orders of magnitude from rock type to rock type /e.g. Skagius 1986, Harvey 1996, Löfgren 2004/.

It is not within the scope of this report to present results, e.g. formation factor distributions, for each single rock type identified within the site investigation program. However, to show that the diffusive properties of different rock types may differ, the in situ formation factor of two different rock types within the Forsmark site investigation area is investigated.

Such investigations can be performed for each rock type identified within the site investigation program. However, before doing this, it is important to co-ordinate such an exercise with the modellers of the performance assessment. It is likely that only a few main rock types need to be investigated. This report aims to show what statistical analysis can be made. Based on this, the modellers should be better equipped to provide a list of the analyses that need to be made in order to obtain proper input data for the performance assessment.

Rock type heterogeneity

The degree of rock type heterogeneity may influence the results and uncertainties in two different ways. Firstly, if assuming that the measured data represents the diffusive properties of the rock, the variance of the formation factor and porosity distributions obtained is likely to be larger if the site is lithologically heterogeneous than homogenous. As described in Chapter 3, the lithology of the Simpevarp subarea is more heterogeneous compared to that of the Forsmark subarea. To show the effect of the heterogeneity, the formation factor distribution from Simpevarp is compared to that of Forsmark.

Secondly, in a heterogeneous rock, the obtained distributions will differ depending on the sample size. The sample size in the laboratory is often on the order of 10^{-4} m^3, while the sample size for electrical conductivity methods in situ is on the order of 10^0 m^3. If the scale of the heterogeneity is small, one can expect that the variances of the distributions obtained in the laboratory are larger than of those obtained in situ. The reason is that some averaging will occur in the larger samples. The more heterogeneous a site is, the more the laboratory variance may differ from the in situ variance. To show this, the laboratory and in situ formation factor distributions are compared for Forsmark and Simpevarp respectively.

Fracture heterogeneity

The effect of fracture heterogeneity on advective flow is discussed in Chapter 2. When it comes to the diffusive properties of rock, the question is to what extent fractures should be included in the rock matrix porosity. It seems reasonable, from a radionuclide retention modelling perspective, not to count fractures where advective flow occurs as part of the rock matrix porosity. The reason is that the rock matrix porosity is an entity influencing the storage capacity of the rock, and thus a retention parameter in the models. Retention such as surface sorption occurs in the fractures. However, in the SA we do not take credit for this.

In the laboratory, non-fractured or sparsely fractured rock samples are, in general, used. This may produce an over-conservative result. In situ, the formation factor of the intact rock matrix can be obtained. This formation factor is called the “rock matrix formation factor”, F_f^{rm} (–). In addition the formation factor of rock containing hydraulically non-conductive fractures (having a water flow below the detection limit of the in situ flow logging tool) can be obtained. This formation factor is called the “fractured rock formation factor” F_f^{fr} (–). To show how the distributions of the two different in situ formation factors may differ, results from Forsmark and Simpevarp are discussed.

Groundwater chemistry

The effective diffusivity of species diffusing through the rock matrix may to some extent depend on the groundwater chemistry as discussed in Section 2.1.1.

If the pore water is non-saline, a large portion of cations diffusing or electromigrating through the rock may do so in the diffuse double layer. If using electrical methods, surface conduction resulting from such electromigration may disturb the results significantly. Therefore in situ formation factors have only been obtained in regions where the groundwater is fairly saline, having an electrical conductivity above 0.5 S m^{-1}. This is normally the case below the transition from fresh-meteoric waters to brackish-marine waters in the upper 200 m of the bedrock /Löfgren and Neretnieks 2005ab/.

Degree of alteration

The degree of alteration has been identified as an important condition both in Chapter 2 and Chapter 4. Earlier publications revealed that the diffusive properties of rock may vary depending on the degree of alteration, where the more altered rock has the larger porosity /e.g. Byegård et al. 2002/. The altered rock is often found adjacent to hydraulically conductive fractures and may potentially be important for radionuclide retention. At present, the frequency of which altered rock occurs around hydraulically conductive fractures is not well known, although it is under investigation.

This report shows some differences in porosities between strongly altered and weekly altered or undisturbed rocks surrounding the borehole KFM02A in Forsmark. At present the diffusive properties of altered rocks are much less well known than the diffusive properties of undisturbed rock. Therefore, this discussion will be on a more qualitative base and will reflect the need for further investigation of the diffusive properties of altered rocks.

Rock stress

The rock stress has been identified as an important condition in both Chapter 2 and Chapter 4 of this report. Earlier publications showed that the formation factors obtained in the laboratory may overestimate the in situ formation factor several times /e.g. Skagius 1986/. The same is true for the porosity /e.g. Möri et al. 2003/. The deviation between in situ data and laboratory data is likely to differ from site to site and from depth to depth. This report reflects upon the need to use depth specific (or rock stress specific) data in the performance assessment. In doing this, an example from the borehole KFM02A in Forsmark is used. Data obtained in the laboratory is compared with data obtained in situ in a number of different depth intervals of the 1,000 m deep borehole.

5.3 Data and uncertainty estimates

5.3.1 Site specific and generic formation factors

Forsmark

At present, much of the knowledge of site-specific formation factors within the Forsmark subarea is based on the investigations of the boreholes KFM01A and KFM02A /Löfgren and Neretnieks 2005b/ performed within the site investigation program by SKB. A short description of the investigations can be found in Chapter 3.

Formation factors have been obtained both in the laboratory and in situ. Figure 5-1 shows the distribution of the 79 formation factors obtained in the laboratory on drill core samples. In obtaining the distribution shown in Figure 5-1, results from all rock samples taken to the laboratory were used, irrespective of conditions, such as e.g. rock type.

As can be seen in Figure 5-1 the formation factors obtained in the laboratory are log-normally distributed, except for two outliers. For guidance on how to read the distribution and the normal-score plot in Figure 5-1 and similar figures presented in this report, please consult Appendix II. In essence, if the grey diamonds can be fitted with a straight line the data is log-normally distributed. The mean value and standard deviation of the laboratory data from the Forsmark subarea are summarised in Table 5-3.

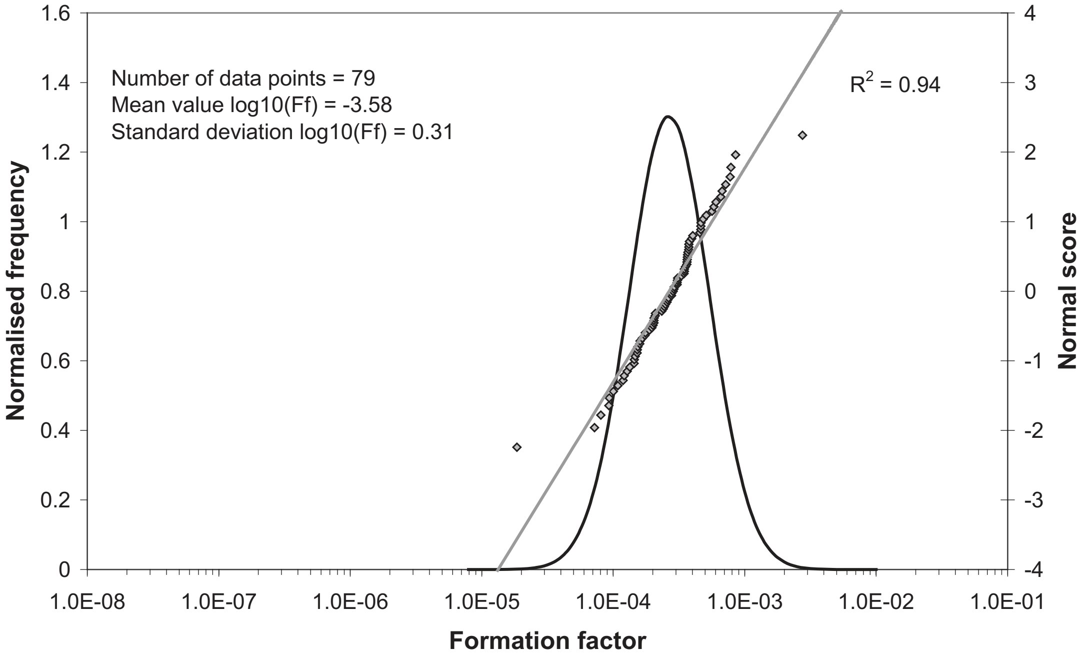


Figure 5-1. *Distribution of laboratory formation factors in Forsmark.*

Figure 5-2 shows the distribution of the 18,330 rock matrix formation factors obtained in situ within the Forsmark subarea. In obtaining the distribution shown in Figure 5-2, in situ results from all depths were used, irrespective of conditions, such as rock type or rock stress.

The normal-score plot in Figure 5-2 gives the visual impression that formation factors in the upper region deviates heavily from the log-normal distribution. However, this is somewhat misleading as only 0.9% of the obtained formation factors exceeds $1 \cdot 10^{-4}$.

The formation factors below $1 \cdot 10^{-5}$ deviate from the log-normal distribution. This deviation is seen in many of the in situ formation factor distributions presented in this report. As much as 17% of the obtained formation factors in Figure 5-2 have a value below $1 \cdot 10^{-5}$.

There are two reasonable explanations for the fact that less formation factors are obtained in the lower region than expected. The first one is the upper measuring ranges of the rock resistivity tools used in situ, which according to the manufacturers are 50,000–100,000 ohm m. This would result in a lower measuring range of the in situ formation factor between $3 \cdot 10^{-6}$ and $2 \cdot 10^{-5}$, if taking into account that the electrical conductivities of the groundwater in the measured region vary between 0.5 S m^{-1}–3 S m^{-1}.

A second reason may be that the scale of the samples in situ, about 1 m^3, is larger than the scale of the heterogeneity. Thus rock with a higher formation factor will influence more on the result than rock with a lower formation factor. This would make it less likely to obtain formation factors in the lower region.

The mean value and standard deviation of the in situ data from the Forsmark subarea are summarised in Table 5-3.

Table 5-3. Site-specific formation factor distributions for Forsmark.

	Number of data points	Mean value $\log_{10}(F_f)$	Standard deviation $\log_{10}(F_f)$	Arithmetic mean value F_f
Laboratory F_f	79	–3.58	0.31	$3.40 \cdot 10^{-4}$
In situ F_f^{rm}	18,330	–4.74	0.25	$3.78 \cdot 10^{-5}$

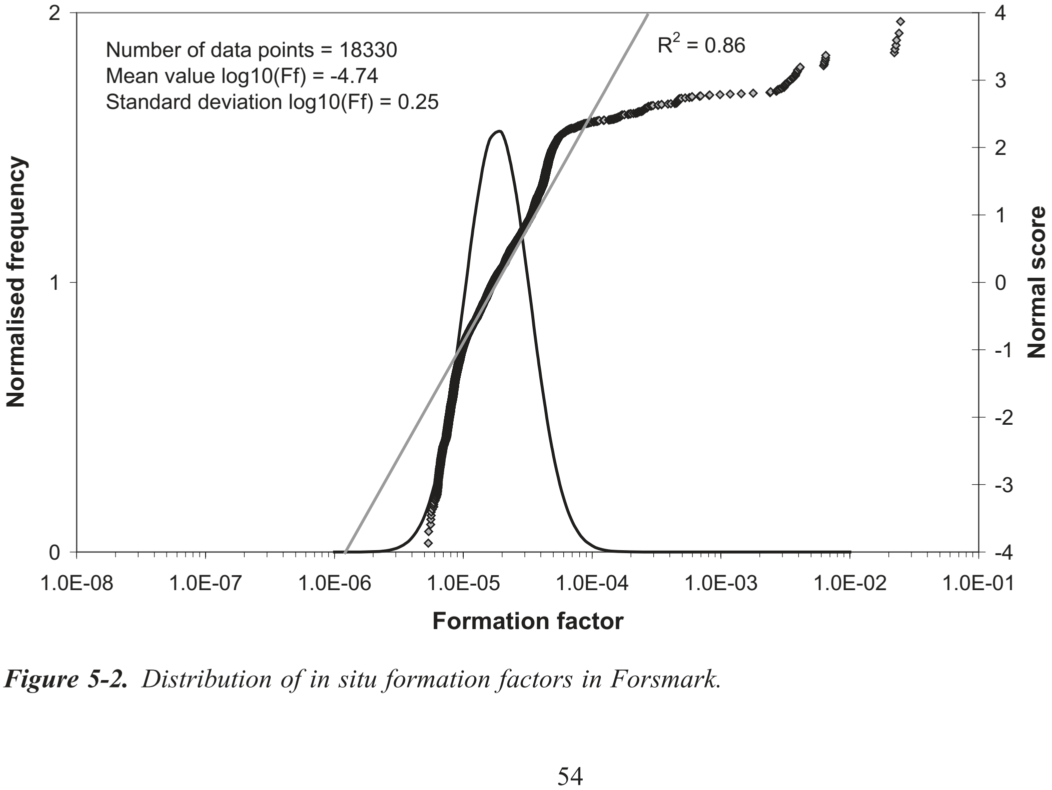


Figure 5-2. Distribution of in situ formation factors in Forsmark.

Simpevarp

At present, much of the knowledge of site-specific formation factors within the Simpevarp subarea is based on the investigations of the boreholes KSH01A and KSH02 /Löfgren and Neretnieks 2005a/ performed within the site investigation program by SKB. A short description of the investigations can be found in Chapter 3.

Formation factors have been obtained both in the laboratory and in situ. Figure 5-3 shows the distribution of the 83 formation factors obtained in the laboratory on drill core samples. In obtaining the distribution shown in Figure 5-3, results from all rock samples taken to the laboratory were used, irrespective of conditions, such as e.g. rock type.

As can be seen in Figure 5-3, the formation factors obtain in the laboratory are fairly log-normally distributed. There seems to be a deviation in the upper formation factor region. According to the log-normal distribution, a few more samples with larger formation factors are expected than was found. A reason for this may be that drill core samples with visible cracks running through them were excluded from the laboratory investigation. The mean value and standard deviation of the laboratory data from the Simpevarp subarea are summarised in Table 5-4.

Figure 5-4 shows the distribution of the 1,866 rock matrix formation factors obtained in situ within the Simpevarp subarea. In obtaining the distribution shown in Figure 5-4, in situ results from all depth were used, irrespective of conditions, such as rock type or rock stress.

The normal-score plot in Figure 5-4 shows that the formation factors below $5{\cdot}10^{-6}$ deviate from the distribution. The same explanations are offered as for the Forsmark in situ data discussed above. 10% of the obtained formation factors in Figure 5-4 have a value below $5{\cdot}10^{-6}$. The mean value and standard deviation of the in situ data from the Simpevarp subarea are summarised in Table 5-4.

Table 5-4. Site-specific formation factor distributions for Simpevarp.

	Number of data points	Mean value $\log_{10}(F_f)$	Standard deviation $\log_{10}(F_f)$	Arithmetic mean value F_f
Laboratory F_f	83	–4.52	0.91	$1.28{\cdot}10^{-4}$
In situ F_f^{rm}	1,866	–4.70	0.45	$4.22{\cdot}10^{-5}$

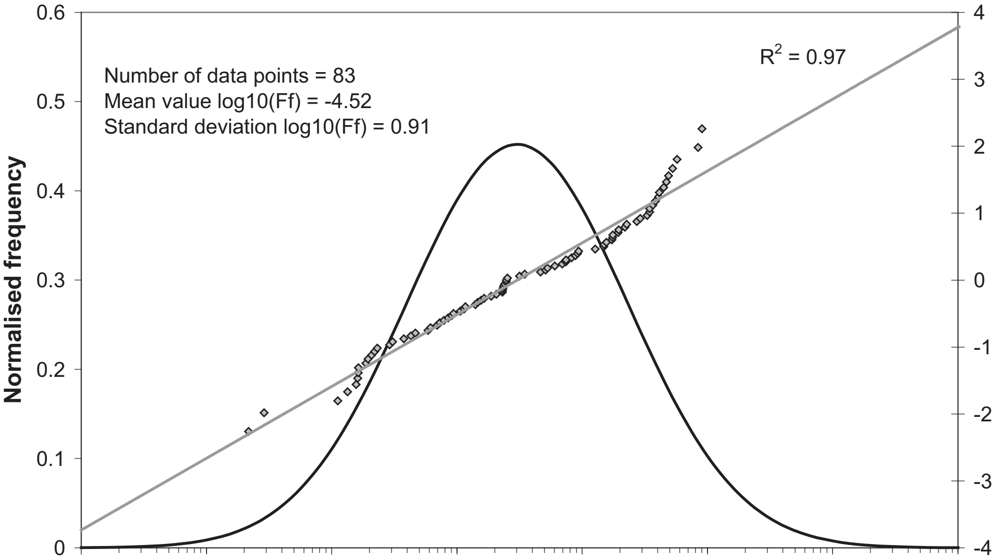


Figure 5-3. *Distribution of laboratory formation factors in Simpevarp.*

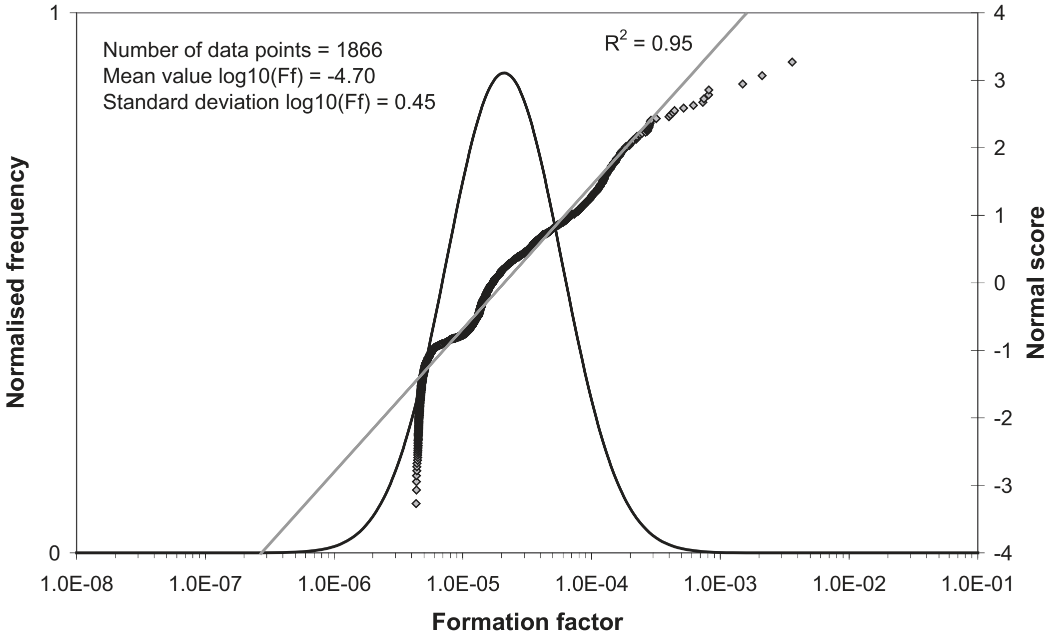


Figure 5-4. Distribution of in situ formation factors in Simpevarp.

Laxemar

At present, much of the knowledge of site-specific formation factors within the Laxemar subarea is based on the investigations in two sections of the borehole KLX02 /Löfgren 2001/ performed by SKB. The sections between the borehole lengths 350–400 m and 720–800 m were investigated. A short description of the investigations can be found in Chapter 3.

Formation factors have been obtained both in the laboratory and in situ. Figure 5-5 shows the distribution of the 80 formation factors obtained in the laboratory on drill core samples. In obtaining the distribution shown in Figure 5-5, results from all rock samples taken to the laboratory were used, irrespective of conditions, such as e.g. rock type.

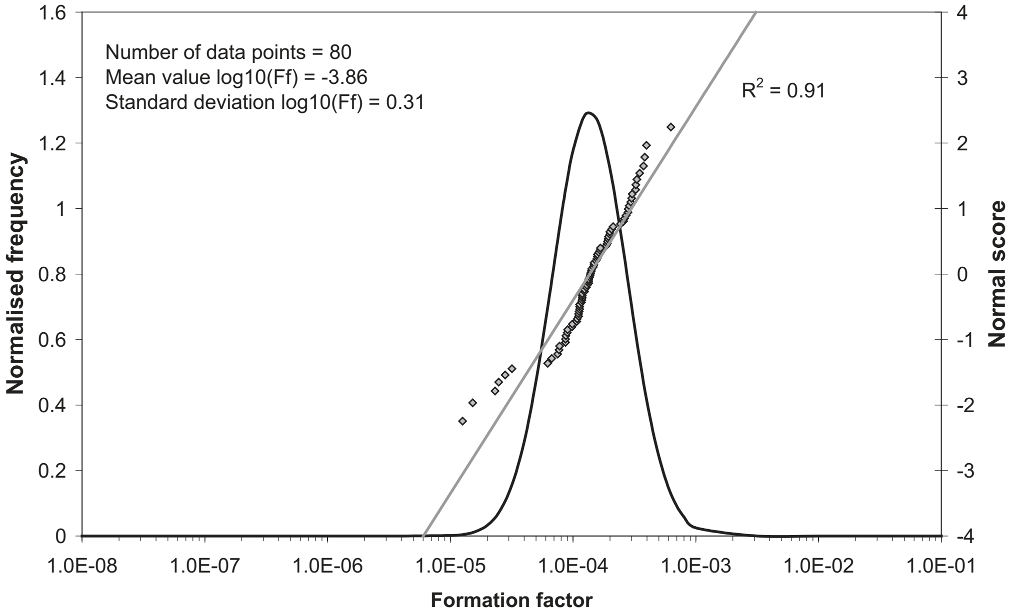


Figure 5-5. Distribution of laboratory formation factors in Laxemar.

As can be seen in Figure 5-5, the formation factors obtain in the laboratory are fairly log-normally distributed. The mean value and standard deviation of the laboratory data from the Laxemar subarea are summarised in Table 5-5.

Figure 5-6 shows the distribution of the 347 rock matrix formation factors obtained in situ within the Laxemar subarea. In obtaining the distribution shown in Figure 5-6, in situ results from all depths investigated were used, irrespective of conditions, such as rock type or rock stress.

The mean value and standard deviation of the laboratory data from the Laxemar subarea are summarised in Table 5-5.

Generic formation factors

All site-specific formation factors shown in Section 5.3.1 have been obtained by electromigration methods by the same authors. Therefore, there may be a need to strengthen the conclusion that the formation factor is fairly well log-normally distributed with typical mean values and standard deviations of $\log_{10}(F_f)$ for certain rock types. Therefore, generic formation factors obtained on Swedish crystalline rock in the laboratory were analysed. These formation factors have been obtained by traditional through-diffusion methods. The literature survey of the published data is, however, far from exhaustive.

Table 5-5. Site-specific formation factor distributions for Laxemar.

	Number of data points	Mean value $\log_{10}(F_f)$	Standard deviation $\log_{10}(F_f)$	Arithmetic mean value F_f
Laboratory F_f	80	–3.86	0.31	$1.68 \cdot 10^{-4}$
In situ F_f^{rm}	347	–4.60	0.23	$2.85 \cdot 10^{-5}$

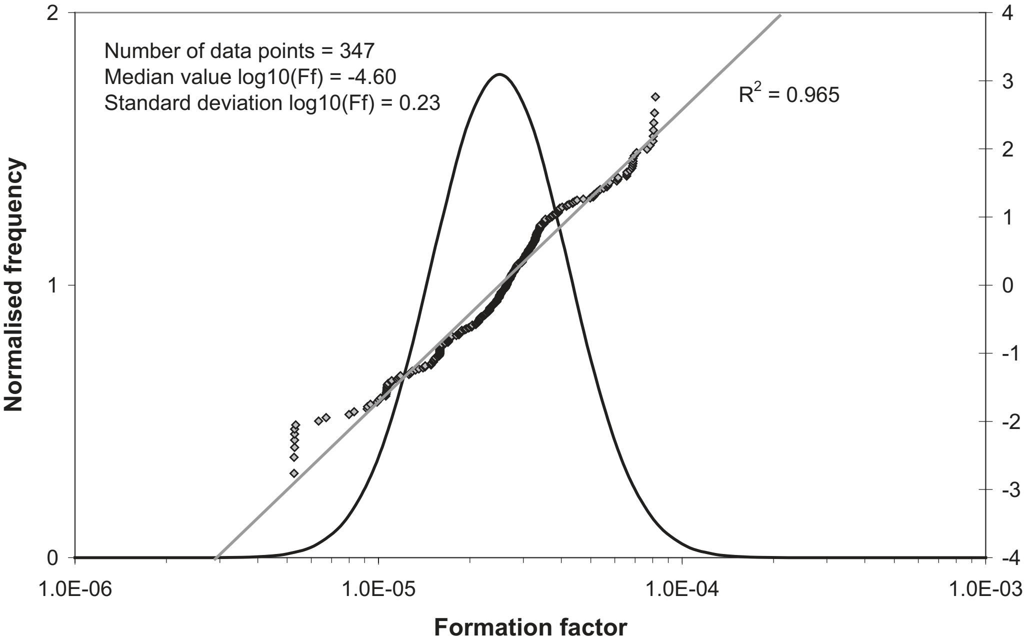


Figure 5-6. Distribution of in situ formation factors in Laxemar.

93 formation factors, obtained in the laboratory of Swedish granite by through-diffusion methods, could be found in the literature /Skagius 1986, Birgersson 1988, Ittner et al. 1988, 1990, Löfgren 2004/. Figure 5-7 shows that these formation factors are fairly well log-normally distributed.

The formation factors shown in Figure 5-7 are compiled in Table B1 in Appendix I.

130 formation factors obtained in the laboratory on Swedish crystalline rocks, including granite, by through-diffusion methods could be found in the literature /Skagius 1986, Birgersson 1988, Ittner et al. 1988, 1990, Ohlsson 2000, Löfgren 2004/. Figure 5-8 shows that these formation factors are fairly well log-normally distributed.

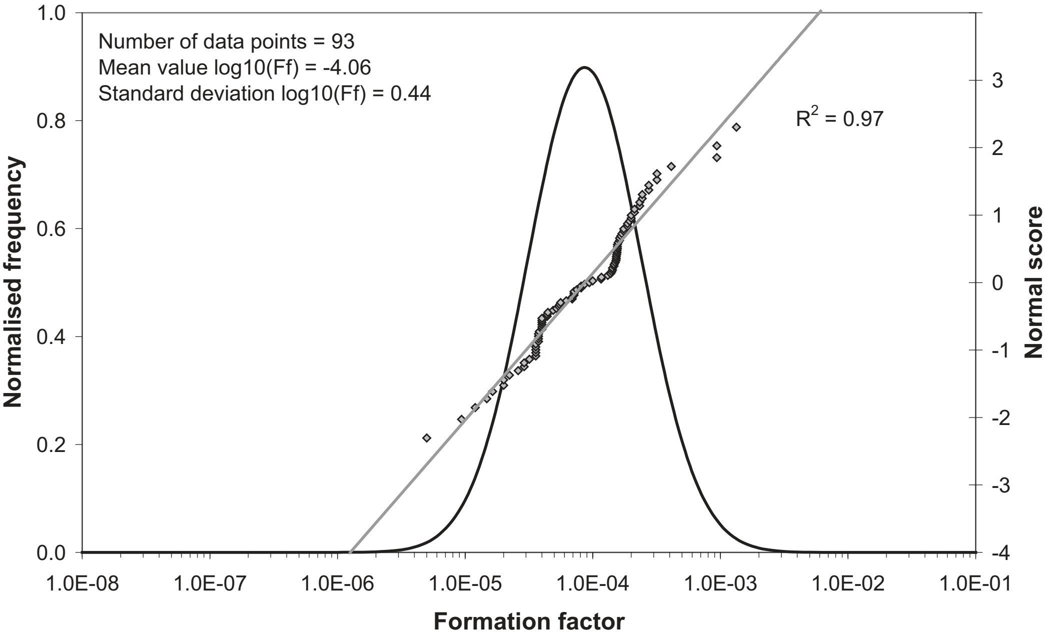


Figure 5-7. *Swedish generic formation factors, granite.*

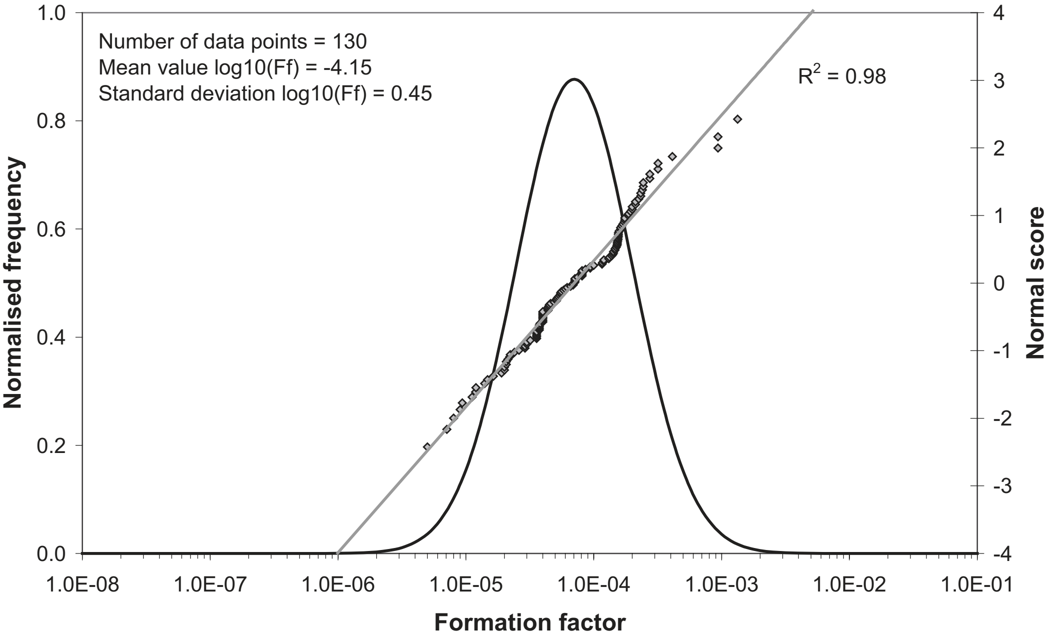


Figure 5-8. *Swedish generic formation factors, crystalline rocks.*

The formation factors shown in Figure 5-8 are compiled in Table B2 in Appendix I. The use of the distribution in Figure 5-8 is recommended when evaluating laboratory experiments on Swedish crystalline rock samples taken outside of the site investigation areas.

Finally, to give an even heavier weight to the site-specific distributions, an analysis of the generic formation factors obtained on international crystalline rocks in the laboratory with traditional through-diffusion methods was performed. 100 formation factors were found /Bradbury and Green 1985, Eriksen 1985, Lehikoinien et al. 1992, Kaukonen et al. 1993, Sato et al. 1996, Kaukonen et al. 1997, Yamaguchi and Nakayama 1998, Vilks et al. 2003/, and their distribution is shown in Figure 5-9.

The formation factors shown in Figure 5-9 are compiled in Table B3 in Appendix I. The distribution shown in Figure 5-9 is comparable with the other laboratory formation factor distributions shown in this chapter.

The mean value and standard deviation of the generic laboratory data are summarised in Table 5-5.

Table 5-6: Generic laboratory formation factor distributions.

	Number of data points	Mean value $\log_{10}(F_f)$	Standard deviation $\log_{10}(F_f)$	Arithmetic mean value F_f
Sweden, granite	80	–4.06	0.44	$1.43 \cdot 10^{-4}$
Sweden, crystalline rock	130	–4.15	0.45	$1.19 \cdot 10^{-4}$
International, crystalline rock	100	–3.72	0.47	$3.07 \cdot 10^{-4}$

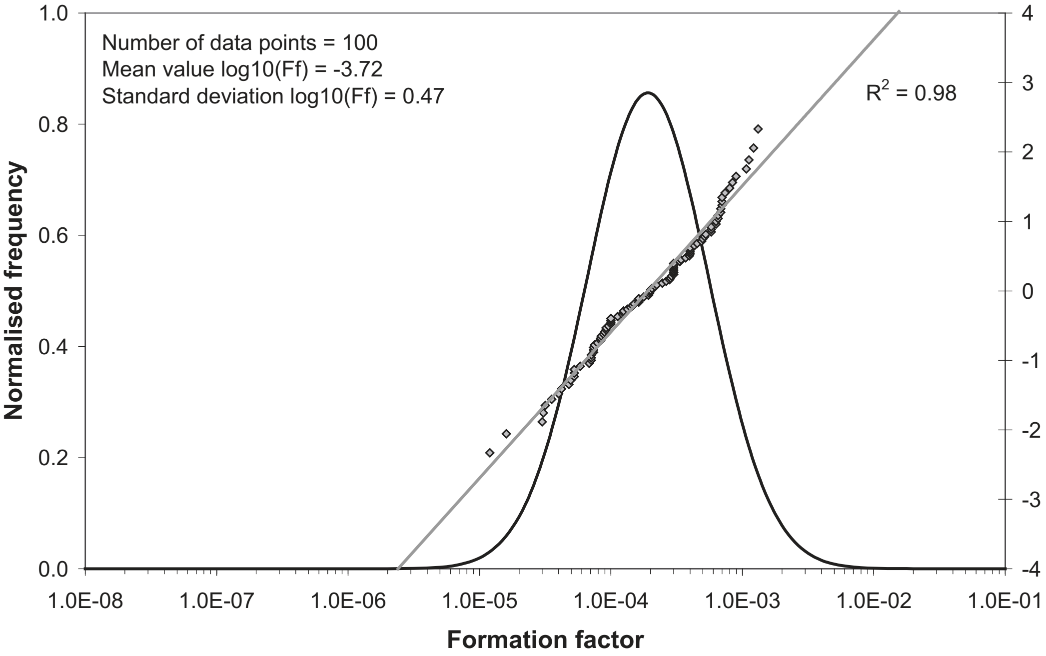


Figure 5-9. International generic formation factors, crystalline rocks.

5.3.2 Site-specific and generic porosities

Forsmark

At present, much of the knowledge of site-specific porosities within the Forsmark subarea is based on the laboratory investigations on the drill cores of the boreholes KFM01A, KFM02A, KFM03A, KFM04A, and KFM05A. These investigations have been performed within the site investigation program of SKB. A short description of the investigations can be found in Chapter 3.

In total, 172 porosity data have been obtained in the laboratory. Out of the 172 drill core samples, 14 samples were strongly altered, having porosities between 9 and 19%. As this is extremely unusual, the results from these samples are discussed in the "Degree of alteration" part of Section 5.3.4. The porosities of these samples are excluded from the porosity distribution shown in Figure 5-10. Except for this, porosities from all rock samples taken to the laboratory were used, irrespective of conditions, such as e.g. rock type.

The mean value and standard deviation of the laboratory porosity data from Forsmark are summarised in Table 5-7.

At present, it is not fully understood how to translate porosities found in the laboratory to in situ porosities. Based on experiments at the Grimsel Test Site in Switzerland on granodiorite, /Möri et al. 2003/ suggested that "conventional laboratory determinations on rock samples overestimate the matrix porosity by a factor of 2 to 2.5" as compared to in situ porosities. There are primarily two sources of the additional porosity created when bringing the rock sample to the laboratory. Firstly the porosity is expected to increase throughout the rock sample as the rock stress is released. Secondly, an additional porosity will be mechanically induced in the drilling process of the borehole and in the sample preparation process /Autio et al. 1999/. For these reasons it is recommended to use the smaller values of site-specific porosities whenever available in the performance assessment than those obtained in the laboratory.

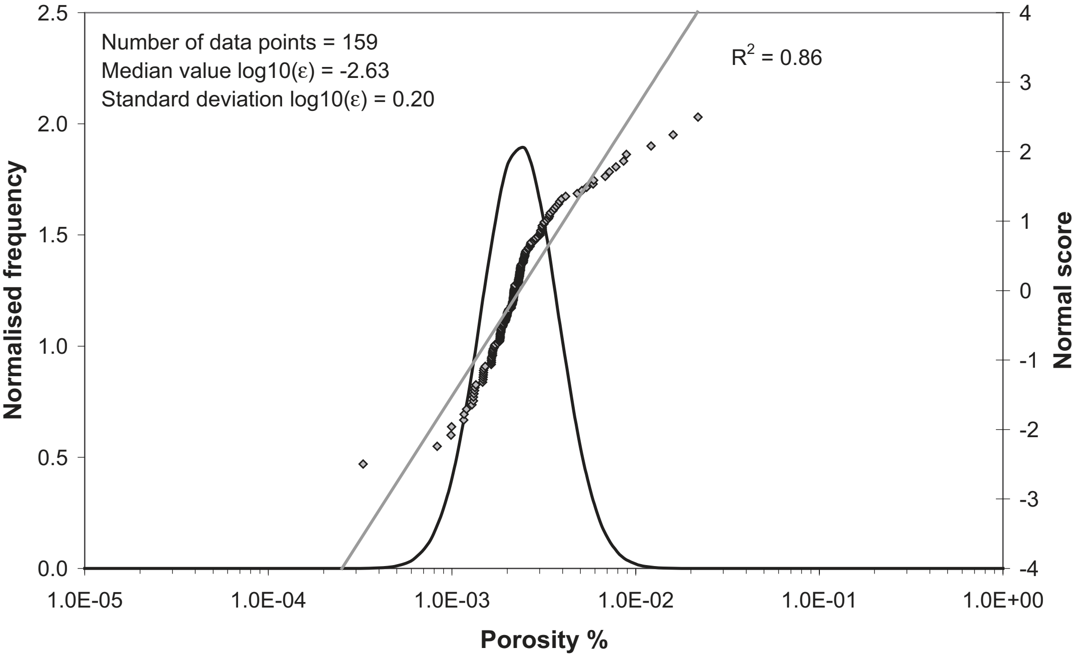


***Figure 5-10.** Distribution of laboratory porosities in Forsmark.*

Simpevarp

At present, much of the knowledge of site-specific porosities within the Simpevarp subarea is based on the laboratory investigations on the drill cores of the boreholes KSH01A and KSH02. These investigations have been performed within the site investigation program of SKB. A short description of the investigations can be found in Chapter 3.

In total, 81 porosities have been obtained in the laboratory. In obtaining the distribution shown in Figure 5-11, results from all rock samples taken to the laboratory were used, irrespective of conditions, such as e.g. rock type.

The mean value and standard deviation of the laboratory porosity data from Simpevarp are summarised in Table 5-11. As discussed above, it is recommended to use smaller site-specific porosities in the performance assessment than obtained in the laboratory.

Laxemar

At present, much of the knowledge of site-specific porosities within the Laxemar subarea is based on the investigations in certain sections of the boreholes KLX02 /Löfgren 2001/. The sections investigated were located between the borehole lengths 350–400 m and 720–800 m. A short description of the investigations can be found in Chapter 3.

In total, 76 porosities have been obtained in the laboratory /Löfgren 2001/. However, 20 of these porosities were obtained when investigating a local rock type change and were not evenly distributed over the drill core sections investigated. Therefore, these porosities were excluded from the distribution shown in Figure 5-12. Except for this, porosities from all rock samples taken to the laboratory were used, irrespective of conditions, such as e.g. rock type.

The mean value and standard deviation of the laboratory porosity data from Laxemar are summarised in Table 5-12.

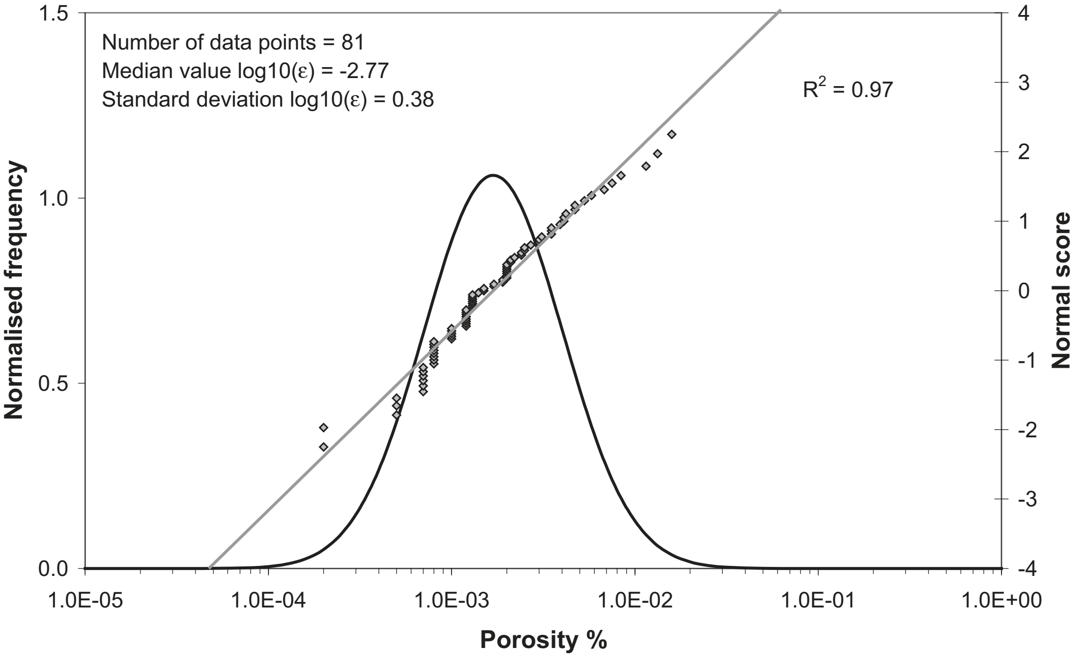


Figure 5-11. Distribution of laboratory porosities in Simpevarp.

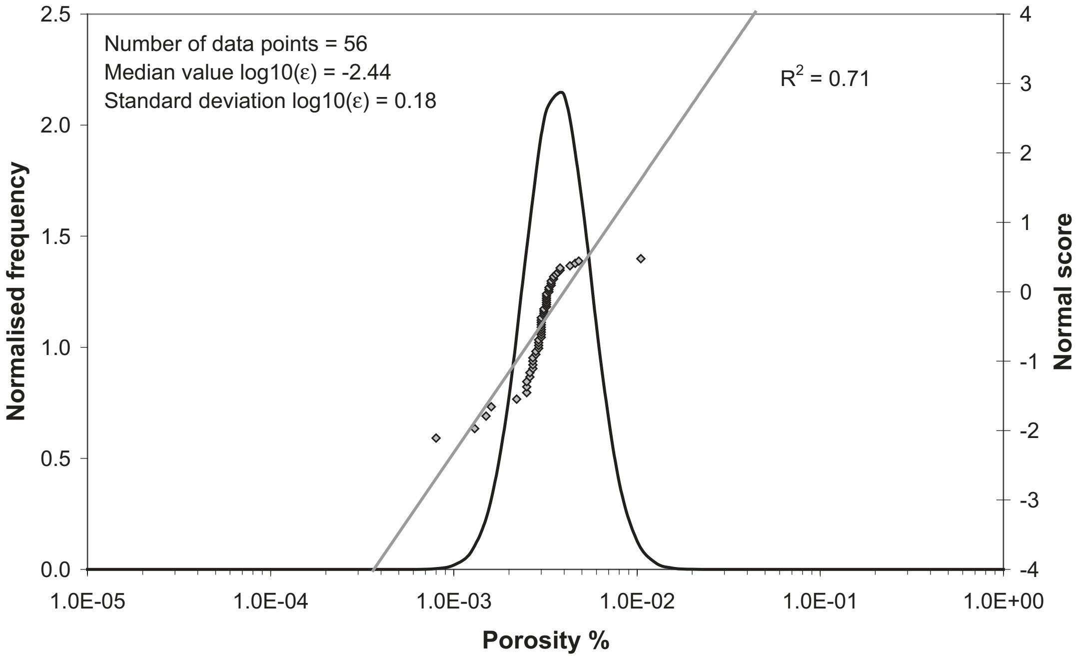


Figure 5-12. *Distribution of laboratory porosities in Laxemar.*

Table 5-7. Site-specific laboratory porosity distributions.

	Number of data points	Mean value $\log_{10}(\varepsilon)$	Standard deviation $\log_{10}(\varepsilon)$	Arithmetic mean value ε
Forsmark	159	–2.63	0.20	$2.75\cdot10^{-3}$
Simpevarp	81	–2.77	0.38	$2.48\cdot10^{-3}$
Laxemar	56	–2.44	0.18	$3.14\cdot10^{-3}$

Generic porosities

The porosity of crystalline rocks has been measured for many decades with quite straightforward methods. The porosity of both Swedish and international granitic rock is commonly reported to be a few per mille. In the Swedish SR-97 safety assessment the porosity value of 0.5% was used. This value may be somewhat high for the Swedish granitic rock, especially for the in situ values. Porosities for both Swedish and international crystalline rocks obtained by different authors are compiled in Tables A and C in Appendix I. These porosities are comparable with the site-specific porosities.

Normal or log-normal porosity distribution

In previous publications, based on fewer data points, it has been suggested that the porosity data could be fitted to a normal distribution /e.g. Löfgren 2004/. For comparison, all the data previous plotted in Figures 5-10 to 5-12 for the lognormal distribution only are summarised and re-plotted for both the log-normal and normal distributions in Figure 5-13 and Figure 5-14 respectively.

From the good fit in Figure 5-13 of the lognormal-scale plot and the poor fit in Figure 5-14 of the normal-scale plot, it can be seen that porosity data for the Swedish crystalline rocks preferentially should be fitted to the log-normal distribution rather than to the normal distribution.

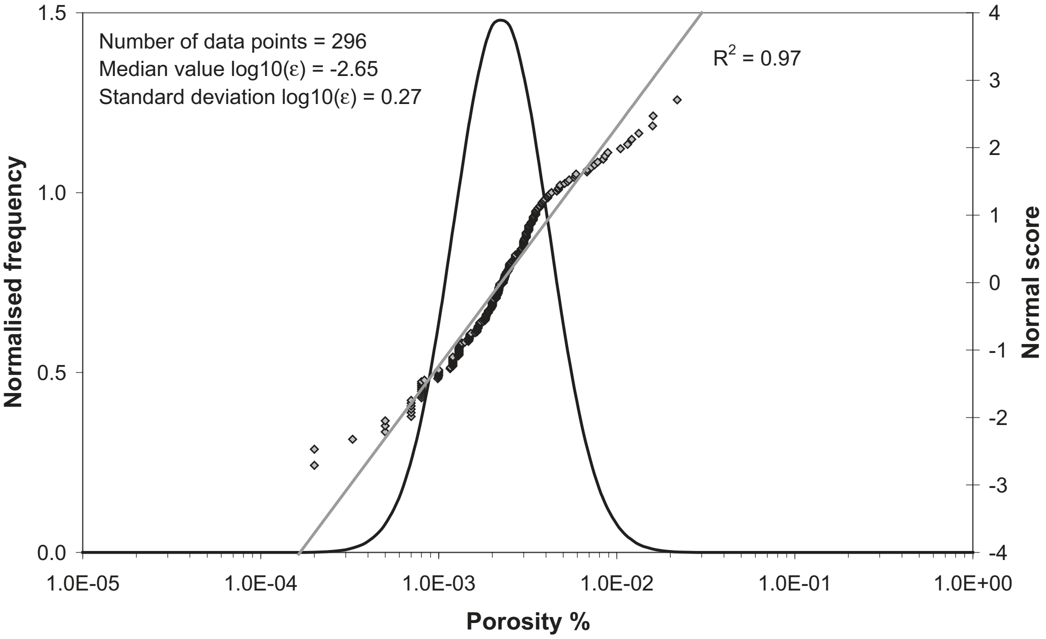


Figure 5-13. Log-normal distribution of site-specific porosities.

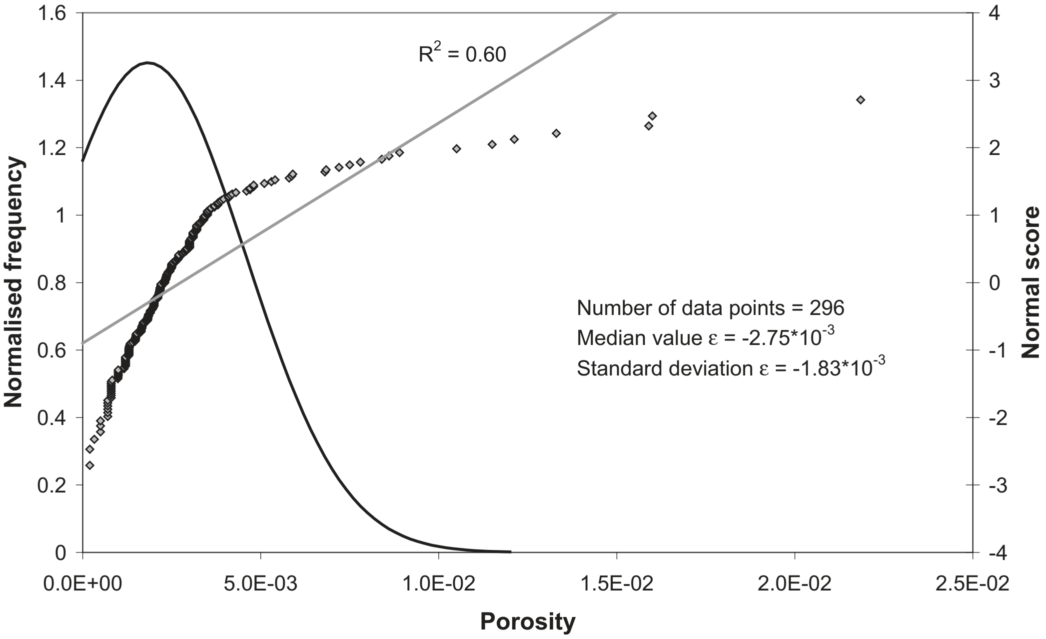


Figure 5-14. Normal distribution of site-specific porosities.

5.3.3 Variation of conditions

Variation of rock type

In order to show how formation factor distributions for different rock types may differ, the in situ data from boreholes KFM01A and KFM02A within the Forsmark subarea are examined. The main rock type is compared to a rock type of minor occurrence. Data from the full lengths of the boreholes are, in general, used in this exercise. The aim of this is to show that rock type specific distributions can be obtained for rock types specified by, for example, the modellers of performance assessments.

The main rock type is described as "Granite to granodiorite, metamorphic, medium-grained" and numbered as 101,057 according to the SKB rock type description system. In borehole KFM02A there is a region between 240 m and 310 m where this rock type is strongly altered and described as vuggy metagranite /Carlsten et al. 2004b/. Data from this section has been excluded from the distribution shown in Figure 5-15 and are instead used in the discussion on the degree of alteration below.

As the rock type of minor occurrence, amphibolite (numbered 102017) was chosen and its formation factor distribution is shown in Figure 5-16.

As can be seen the diffusive properties of these two rock types do not differ significantly. The arithmetic mean value of the formation factor of rock type 101057 is $2.10 \cdot 10^{-5}$ and of rock type 102017 is $2.46 \cdot 10^{-5}$. One may or may not draw the conclusion that these two rock types can be lumped together, with respect to their diffusive properties, in a performance assessment. The formation factor distributions of other rock types may have differed more or less.

Variation of rock type heterogeneity

As discussed previously, the formation factor distributions of lithologically heterogeneous sites are expected to have a higher variation than those of lithologically homogenous sites. This can be seen if comparing in situ results in Figure 5-2 from the Forsmark subarea and Figure 5-4 from the Simpevarp subarea. The mean values of the distributions are almost identical for the two sites, while the standard deviation is larger for the Simpevarp site.

In the performance assessment there may be a need to divide the site into different sub-domains, where the rock has diffusive properties within certain ranges. There may be a need to divide lithologically heterogeneous sites into more sub-domains than lithologically homogenous sites.

As discussed in Chapter 2, the variance of the formation factor distribution will depend on the scale of the heterogeneity in combination with the scale of the sample. If using small samples, it is likely that the variance is larger than if using larger samples. The distribution in Figure 5-3 from Simpevarp is obtained on small samples in the laboratory. The variance of this distribution is larger than the variance in Figure 5-4 from Simpevarp, obtained on larger samples in situ.

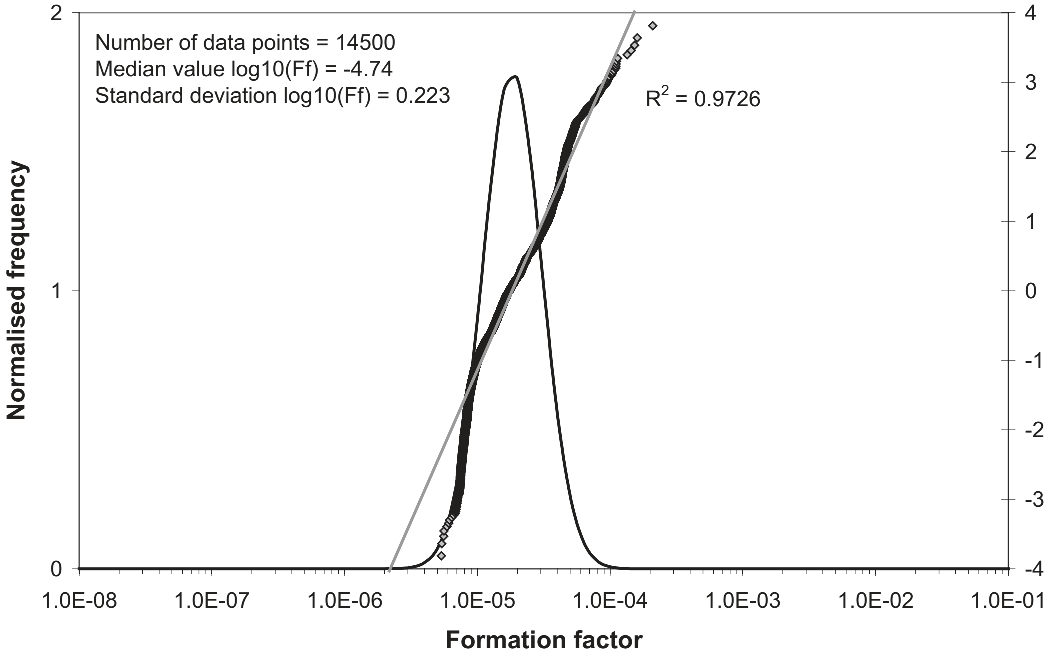


Figure 5-15. Distribution of in situ formation factors for rock type 101057.

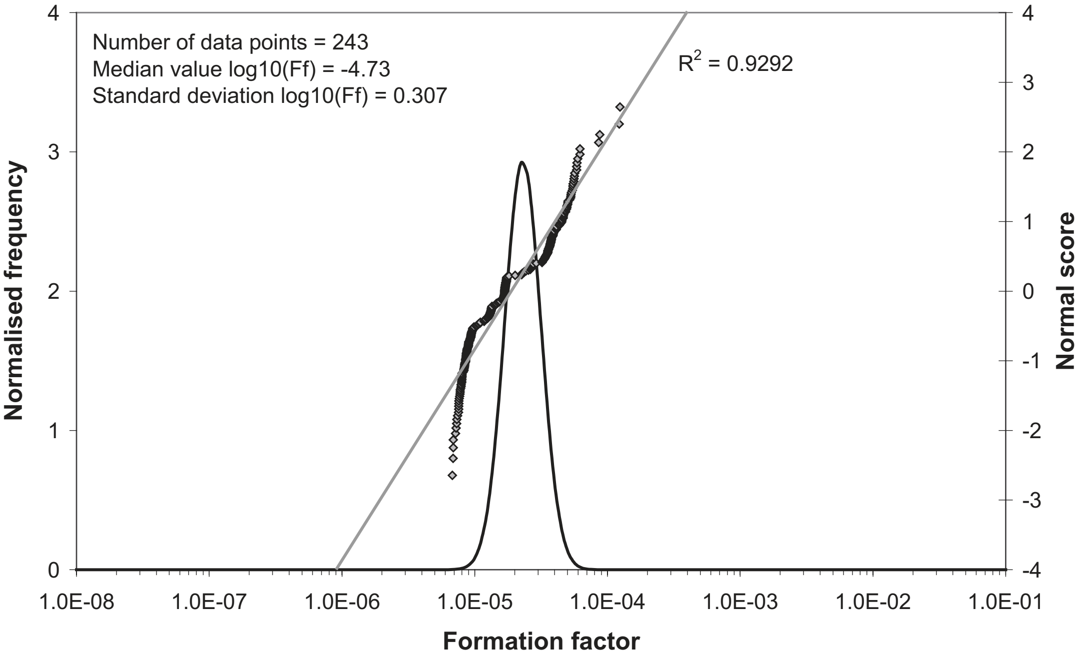


Figure 5-16. *Distribution of in situ formation factors for rock type 102017.*

As discussed in Section 2.2.1 it is presently not known exactly how to properly transform formation factor distributions obtained on one sample scale to another. Clearly a thorough investigation of how the heterogeneity and migration in three dimensions will affect such a transformation is needed. In Equation (2.12) it is shown how to perform the transformation in one dimension. In the following example it is assumed that Equation (2.12) applies. As we do not understand exactly the basis for such an assumption, the discussion below can only be taken as qualitative.

$$s_{large} = \sqrt{\frac{L_{small}}{L_{large}}} \cdot s_{small} \qquad \text{(Equation 2.12)}$$

Figure 5-3 shows the standard deviation of the laboratory formation factors from Simpevarp. The characteristic sample length in the laboratory is 0.03 m /Löfgren and Neretnieks 2005a/. If using these numbers in Equation (2.12), what would then the standard deviation for other characteristic sample lengths be?

To focus on the reasoning, the resulting curve was compared with the standard deviation of the in situ formation factors from Simpevarp. Here the characteristic sample length was taken to be 0.1 m, based on the vertical resolution of the in situ rock resistivity tool. The standard deviations and characteristic sample lengths are shown in Table 5-8.

The values in lines 2 and 3 would be appropriate for sorbing nuclides and PA time scales. For non-sorbing nuclides line 4 would be more appropriate.

Table 5-8. Input data to Figure 5-17.

Type of sample	Characteristic sample length and penetration depths (m)	Standard deviation $\log_{10}(F_f)$
Laboratory	0.03	0.91
In situ	0.1	0.45
FARF31	30	?

The values in Table 5-8 are plotted in Figure 5-17. The resulting curve based on the two first points and the reasoning around Equations 2.11 and 2.12 is also shown

As can be seen, the standard deviations for the laboratory formation factors could (perhaps somehow fortunately) be representative to standard deviations for the in situ formation factors applicable for sorbing species over PA time scales. The measured distributions with their means and standard deviations can thus be used directly in PA nuclide transport calculations to obtain local values along a streamtube for sorbing species.

For averaging along a flowpath that extends 500 m the standard deviation of the mean for the flowpath will be $\sqrt{500/0.1} = 70.7$ times smaller than that for the local values based on a sample 0.1 m in size. This implies that for most PA purposes the variation expressed as standard deviation of D_e along the flowpaths is negligible.

For a non-sorbing species the standard deviation can be expected to be even smaller because then also the averaging in the direction into the rock averages D_e even more.

It is interesting to note that even in lithologically heterogeneous sites, the variance of the formation factor may be small on a performance assessment scale. Again it should be noted that the above reasoning is only quantitative.

Variation of fracture heterogeneity

To investigate whether diffusion into open but hydraulically non-conductive fractures is of interest for retention modelling, all in situ data from borehole KFM02A in Forsmark was investigated. The aim of this discussion is to show that it may be over-conservative to only account for diffusion in the intact rock matrix in retention modelling.

The rock matrix formation factor obtained in situ F_f^{rm} is compared with the fractured rock formation factor obtained in situ F_f^{fr}. As a deviation from the log-normal distribution is introduced when including open but hydraulically non-conductive fractures into the rock porosity, formation factor histograms are shown instead of normal-scale plots in Figures 5-18 and 5-19.

Figure 5-18 shows that the rock matrix formation factor is fairly well distributed according to the log-normal distribution.

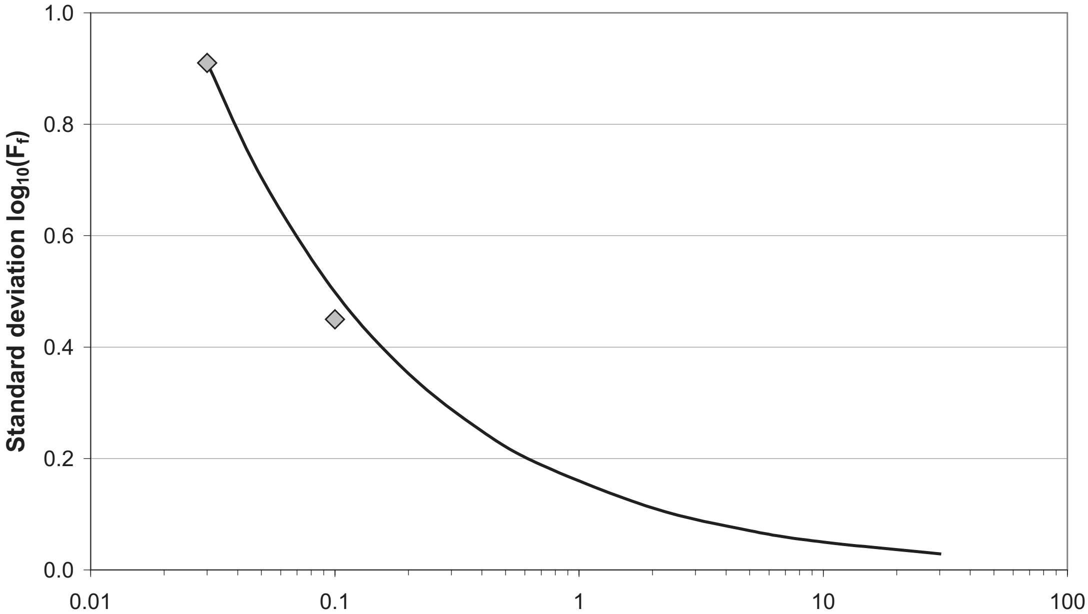


Figure 5-17. Possible standard deviations at different sample scales.

In the distribution for the fractured rock formation factor, shown in Figure 5-19, there is an increase in number of formation factors in the upper region. This increase is related to the open but hydraulically non-conductive fractures intersecting the borehole.

The arithmetic mean value is $7.23 \cdot 10^{-5}$ for the rock matrix formation factor and $1.90 \cdot 10^{-4}$ for the fractured rock formation factor. Judging from this, diffusion into open but hydraulically non-conductive fractures may be as important for retention as diffusion into the rock matrix, at least for non-sorbing species. A prerequisite for this is that the fractures are distributed throughout the rock volume. The retention capacity due to diffusion into open but hydraulically non-conductive fractures is expected to be larger in heavily fractured rock than in sparsely fractured rock.

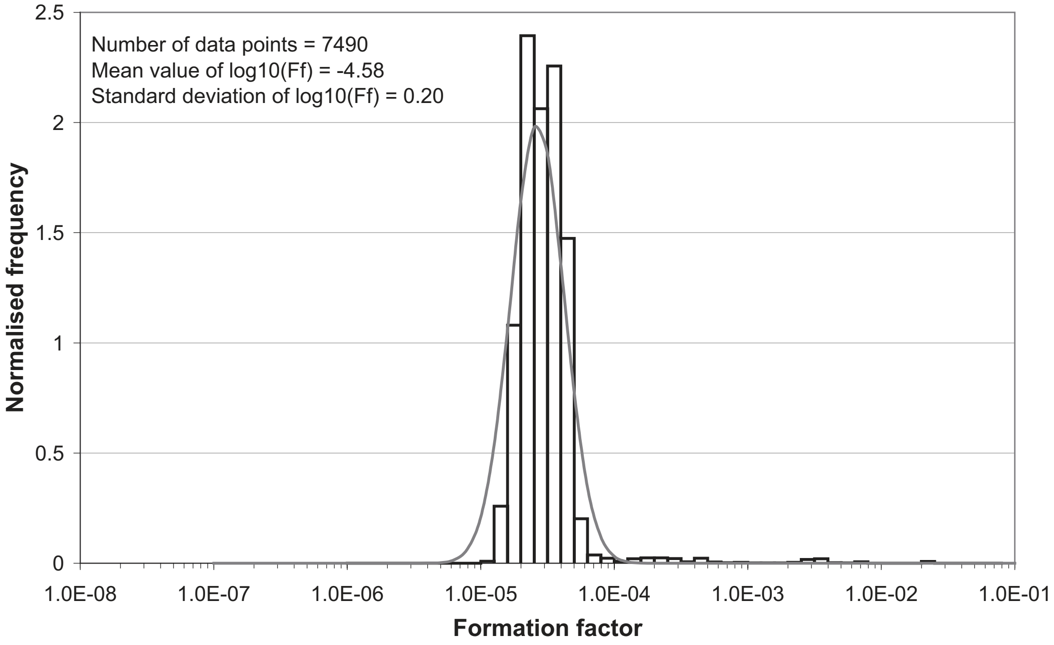


***Figure 5-18.** Distribution of rock matrix formation factors in KFM02A.*

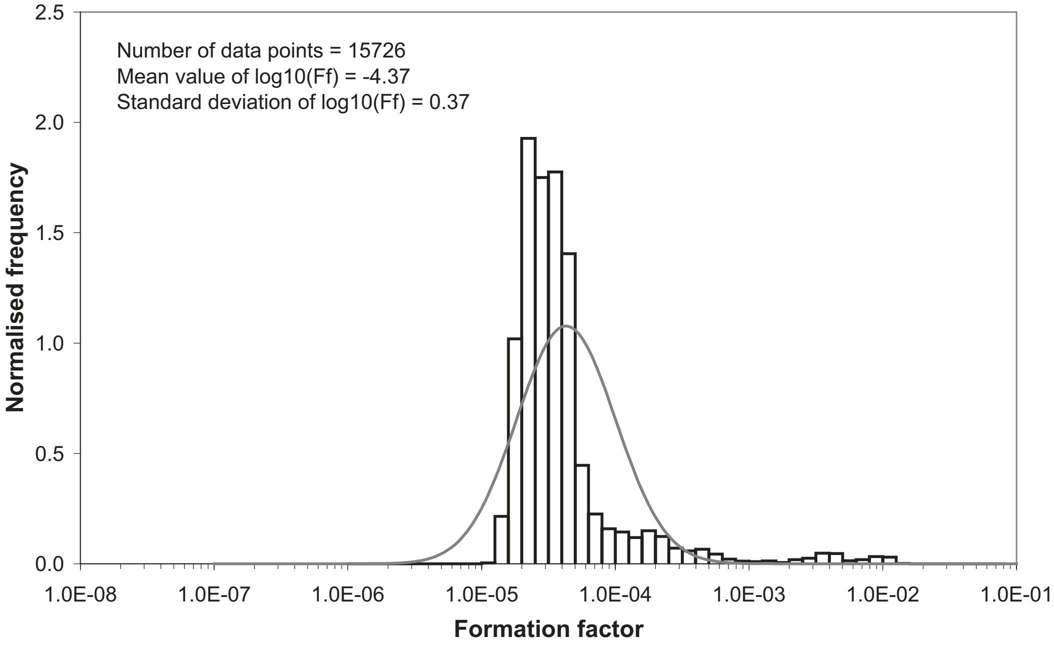


***Figure 5-19.** Distribution of fractured rock formation factors in KFM02A.*

Variation of degree of alteration

The diffusive properties of altered rock adjacent to hydraulically conductive fractures have at present not been systematically investigated, even though such investigations are currently undertaken. Therefore this discussion should only be taken qualitatively.

Before introducing a retention capacity of an alteration zone into a performance assessment, one has to show to what extent the rock adjacent to hydraulically conductive fractures is altered. This concerns both the thickness of the alteration zone and the statistical likelihood that the rock constituting the flow wetted surface is altered.

The alteration zone is commonly referred to as having higher porosity and formation factor than the undisturbed rock /e.g. Byegård et al. 2002/. Formation factors and porosities of fissure coating material is compiled in Table B4 in Appendix I. Figure 5-20 shows the formation factor distribution of the strongly altered zone at 240–310 m depth in KFM02A in Forsmark where the rock is described as vuggy metagranite /Carlsten et al. 2004a/.

This rock is a part of the rock type 101057 (granite to granodiorite, metamorphic, medium-grained) and Figure 5-20 could be compared with Figure 5-15, which shows the formation factor distribution of the non-altered rock of this rock type. As can be seen the formation factors are much higher in the altered rock compared to the unaltered rock. The arithmetic mean value of F_f^{rm} for the vuggy metagranite is as high as $2.46 \cdot 10^{-3}$.

This discussion only aims to point out that the retention capacity of altered rock may be of interest to include in a performance assessment. However, more information on the alteration zone is needed, This includes information of porosity, internal surface and diffusivity. Practically always the rock nearest the fracture has a somewhat increased porosity. This implies that it will be more readily accessed by a diffusing species. At the same time there is only a marginal decrease in the mass of minerals per volume of rock so that a negligible reduction in sorption capacity can be expected unless there is a substantial change in sorbing surface or if the minerals have become less sorbing.

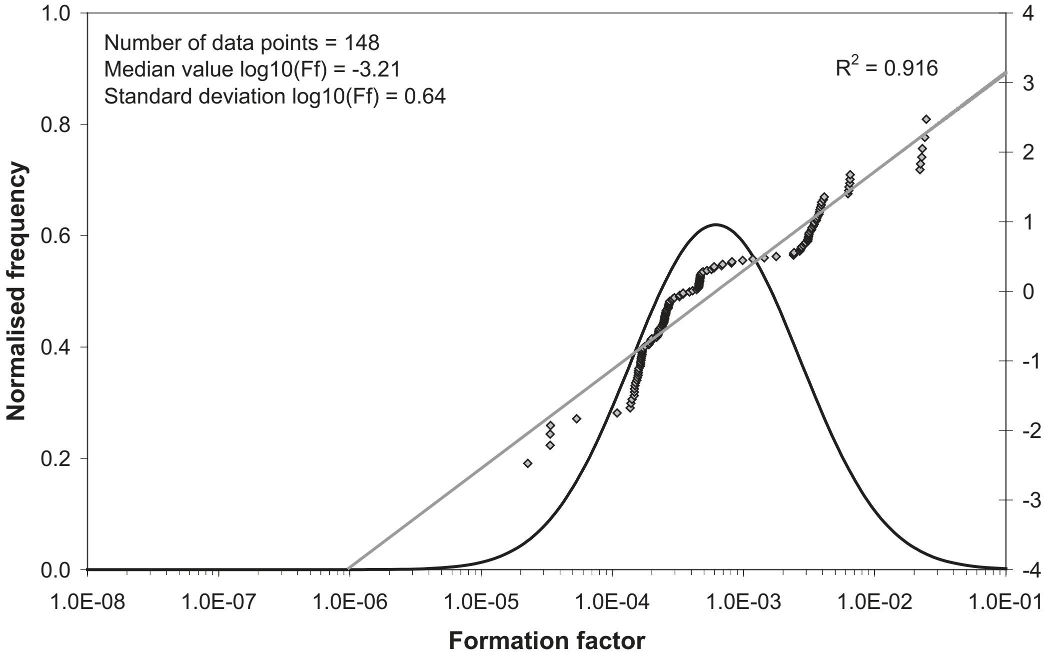


Figure 5-20. *Distribution of rock formation factors in strongly altered zone in KFM02A.*

Variation of rock stress

In earlier publications it has been reported that it is likely that samples brought to the laboratory have an increased formation factor as well as porosity /e.g. Skagius 1986, Möri et al. 2003/. This is partly due to the fact that stress is released and partly that the samples are mechanically damaged in the sample preparation.

To investigate how the rock stress affects the formation factor in situ, data from borehole KFM01A in Forsmark was investigated. Only rock matrix formation factors of the rock type 101057 (granite to granodiorite, metamorphic, medium-grained) were used. The borehole was divided into different sections as shown in Table 5-9. Data from each section were fitted to log-normal distributions. The distribution parameters are shown in Table 5-9.

In Table 5-9, the distribution parameters of the laboratory formation factors, obtained on the rock type 101057 from the whole borehole, are included for comparison. The mean values and standard deviations of the distribution in Table 5-9 are shown in Figure 5-21 as a function of borehole length.

Table 5-9. Distribution parameters at different depths.

Section	Number of data points	Mean value $\text{Log}_{10}(F_f)$	Standard deviation $\text{Log}_{10}(F_f)$
Laboratory Whole borehole	32	–3.58	0.25
150–250 m	419	–4.54	0.13
250–350 m	294	–4.57	0.11
350–450 m	638	–4.59	0.10
450–550 m	219	–4.58	0.23
550–650 m	502	–4.68	0.087
650–750 m	1,083	–4.52	0.10
750–850 m	1,259	–4.40	0.051
850–950 m	693	–4.52	0.082
950–1,000 m	627	–4.55	0.081

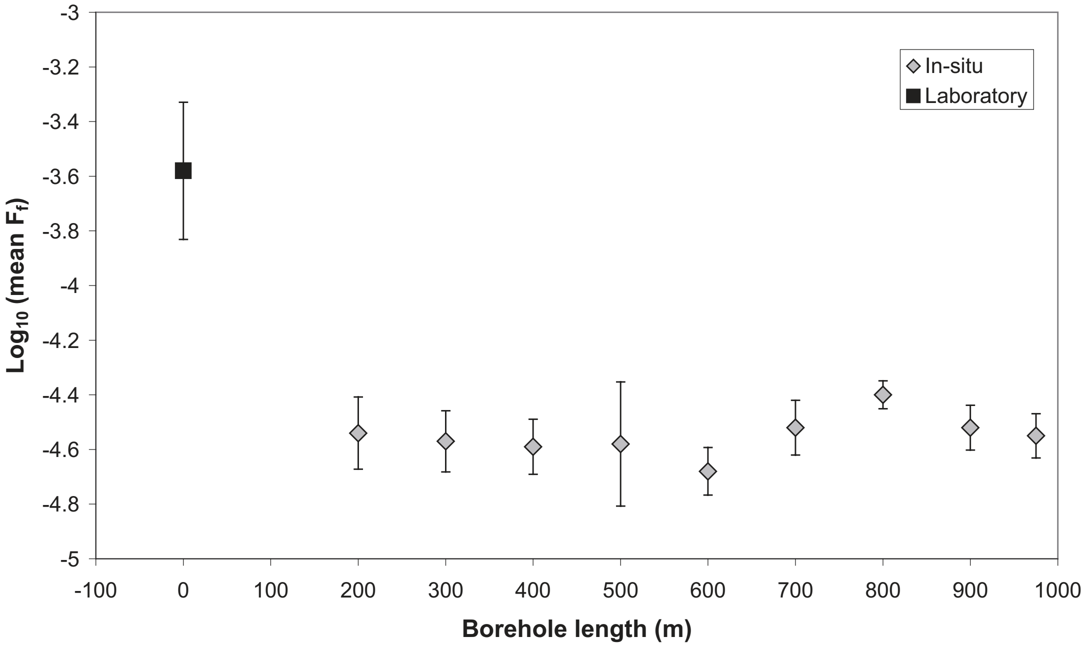


Figure 5-21. *Mean value and standard deviation of* $\text{Log}_{10}(F_f)$ *at different depth.*

From Figure 5-21 one may draw the conclusion that the rock stress does not significantly affect the diffusive properties of the rock, since there is not so large variation of the formation factor along the borehole length. The increased formation factors in laboratory samples would then be due to the mechanical damage induced in the sample preparation. A combination of the two interpretations may also be used.

As the laboratory formation factors are as much as one order of magnitude larger than the in situ formation factors, it is suggested to use in situ formation factors in the performance assessment. The error induced when bringing the samples to the laboratory is suggested to be larger than the error induced in the in situ measurements due to lack of controlling the experimental conditions.

5.3.4 Subjective uncertainty assessment for formation factors and effective diffusivities

The focus of this section is on the uncertainties of the in situ data. The in situ measurement technique is new. It is the first time such measurements have been made to determine in situ formation factors. We have tried to find any weaknesses in the technique by twisting and turning arguments and have encouraged a number of knowledgeable colleagues to find weaknesses in the method and the underlying theories. So far no serious objections have been raised.

A number of arguments have been raised by colleagues that electrical conductivity methods might not give data useful for determining matrix diffusion data. This has lead us to investigate these arguments and to explore counter-arguments. One, such major argument against the electrical conductivity method is that alternating current is used. Therefore, the methods may not show that ions actually pass through the rock matrix, which is the basis for translating electrical conductivity to diffusivity. To investigate this argument we developed a new technique, the through-electromigration method, by which tracer ions were demonstrated to move through samples when subjected to a direct current electric field. The migration rate corresponded to that predicted by the alternating current measurements. This matter is discussed in detail elsewhere /Löfgren and Neretnieks 2006/. This and other test have led us to believe that the in situ method is conceptually sound and gives results that should be treated with the same dignity as results from traditional methods. Below, a subjective uncertainty assessment is given for the obtained formation factors and effective diffusivities.

Uncertainties in measured in situ values

The uncertainties associated with the rock resistivity measurements in situ are small, but not insignificant, compared to other uncertainties. By examining rock resistivity logs from different downhole tools in the same borehole, it is assessed that the uncertainties of the rock resistivity measurements are on the average less than a factor of two. However, if greater care is taken in the calibration of the downhole tool prior to the measurement, this uncertainty would be smaller and insignificant compared to other uncertainties. It is reasonable to demand that the contractors providing the downhole tool or downhole logging should provide a good calibration of the tools. If part of the rock is very resistive, its resistivity may be larger than the measuring range of the downhole tool. Therefore, it is recommended to choose the downhole tool with care.

The uncertainties associated with the measurements of the groundwater electrical conductivity is assessed to be insignificant compared to other uncertainties. As the uncertainties associated with the in situ measurements are small they are handled under the heading “various uncertainties”.

Uncertainties in assessed pore water electrical conductivity

Translating the electrical conductivity to a formation factor involves an assumption that the pore water electrical conductivity is essentially the same as that of the groundwater extracted from nearby fractures. Some confirmation of this is gained from laboratory measurements of electrical conductivities on cores from the same depth in the borehole. Furthermore, the water composition from fractures over long distances in the holes, especially in depths below a few hundred meters changes slowly. Judging from the groundwater electrical conductivity vs. depth profiles from the site investigations in Forsmark, Simpevarp, and Laxemar, the electrical conductivity of the groundwater changes less than 1 S m^{-1} per 100 meter. This subjective uncertainty is assessed to, on the average, be less than a factor of 2.

Uncertainties in translating formation factors to effective diffusivities

Translating the formation factor to effective diffusivity involves data on diffusivity in the bulk water. For a small number of the nuclides the diffusivity in the bulk water is fairly well known. For other species it is taken to be 10^{-9} m^2 s^{-1} /Ohlsson and Neretnieks 1997/. This uncertainty is subjectively set to be less than a factor 2.

Translating the formation factor to effective diffusivity also involves using a proper multicomponent mass transfer model. At present binary mass transfer models are used. This uncertainty is subjectively set to be less than a factor 2.

Natural variability of the formation factor

The more heterogeneous the rock is, both with regard to rock types and fractures, the larger the natural variability of the formation factor is expected to be. The variability of the rock matrix formation factors obtained in situ is summarised in Tables 5-3 for results obtained from Forsmark, Table 5-4 from Simpevarp, and Table 5-5 from Laxemar. Expressed as standard deviations in $\log_{10}$ scale they are: 0.25, 0.45, and 0.23 respectively. For use in the performance assessment modelling the variability expressed as σ will be reduced by a factor of about 30 for a flowpath length of 100 m because the flowing water will randomly encounter sections with different formation factors (see Sections 2.2.1 and 5.3.3). The reduction in σ can be larger if the averaging is done over the whole length of the streamtube from the canister to the ground surface.

When averaging, it is assumed that the flow paths do not preferentially go through special rock types but encounters rock types at random. This assumption is supported by observations in the site investigations. However, the rock surrounding the flow paths may be preferentially altered, which is discussed below. It is assessed that uncertainties due to the natural variation of the formation factor will have a negligible effect on the performance assessment, as compared to other uncertainties.

Uncertainties in pore connectivity

The pore connectivity, which would affect the maximum penetration depth of the radionuclides into the rock matrix, has been identified as an important entity for a performance assessment. In the site investigations it has been shown that current emitted from the rock resistivity downhole tool can be passed through the rock on a meter scale at least. Therefore, we suggest that the porous system of the rock matrix is connected on all scales relevant for the performance assessment. The uncertainty in pore connectivity is negligible compared to other uncertainties.

Uncertainties due to altered rock nearest the fracture surface

This uncertainty cannot be well quantified because the frequency and extent of such alterations along the flowpaths are not known. Also the diffusion and sorption properties of the altered rock are not well quantified. The altered rock is, however, more porous than undisturbed rock in most cases and would increase the matrix diffusivity. For constant water chemistry over time, this underestimates the uptake into the matrix and thus the retardation of nuclides. Pore clogging due to precipitation is not expected under normal chemical conditions. The presence of massive amounts of concrete could possibly alter the situation but this is not planned. This uncertainty may cause an underestimation of the formation factor by a subjectively assessed factor of 2 or possibly more if the rock is extensively altered.

Uncertainties due to rock stress and release of rock stress

This uncertainty may be large if basing the formation factors used in the performance assessment on results obtained in the laboratory. This is partly due to the release in rock stress but also partly due to a mechanical disturbance of the rock induced in the sample preparation. It is subjectively assessed that formation factors obtained in the laboratory are overestimated by one order of magnitude.

- The uncertainties associated with rock stress are assessed to be negligible compared to other uncertainties if in situ measurements are used. The disturbance in the stress field due to the presence of the hole is small and is expected to be only a few centimetres and the current emitted from the rock resistivity downhole tool penetrates more than a metre into rock.

Other uncertainties that emanate from observation on laboratory and other investigations.

We have not found any observations in the large amount of different laboratory or the small number of field investigations that detract from the above conclusions and suggestions.

5.3.5 Subjective uncertainty assessment for rock matrix porosities

Uncertainties in measurement methods

Uncertainties in measurement methods are subjectively assessed to be less than a factor of 1.2 for the weighing method as well as the less commonly used breakthrough time method.

Natural variability of the measured porosities in the laboratory

Table 5-6 summarises measured data for Swedish granites and crystalline rocks as well as for data on crystalline rocks from other countries. The standard deviation in log space varies between 0.44 and 0.47. This is approximately equivalent to a variation with a factor of 3 up and down.

Uncertainties due to rock stress and release of rock stress

The samples used in the laboratory are taken out of their natural environment and de-stressed. Furthermore the samples are mechanically damaged in the sample preparation. Indications from Swedish, Canadian, and Swiss in situ measurements suggest that the in situ porosities at a depth of 300 to 400 metres are about a factor of three smaller than suggested by laboratory measurement (see Section 3.1.1). A factor of 5 is subjectively deemed to be a reasonable upper value due to this effect.

5.4 Recommended values for PA

We recommend for performance assessment purposes, that the arithmetic mean of the in situ measured formation factors should be used in a base case. For an entire flow path, the uncertainty in those data is on the average at most a factor of three for the decrease and possibly more for the increase, because we have not accounted for the possible presence of more diffusive altered rock adjacent to hydraulically conductive fractures. The recommended central formation factor values for the subareas Forsmark, Simpevarp, and Laxemar are listed in Table 5-10 together with high and low estimations.

We recommend for performance assessment purposes that the arithmetic mean of the laboratory measured porosities should be used as a high estimation of the in situ porosity. The reason is that the arithmetic mean is mathematically larger than the geometric mean and the laboratory measured values are often larger than the in situ values due to e.g. de-stress effect. For the central value in the base case, we recommend that the high value should be divided by a factor of three. For the low value, we recommend that the high value should be divided by a factor of five. The recommended in situ porosities for the subareas Forsmark, Simpevarp, and Laxemar are listed in Table 5-10.

To obtain effective diffusivities for specific species, it is recommended to multiply formation factors in Table 5-10 with the diffusivities in free solution shown in Table 5-11.

Table 5-10. Recommended value for base case in PA.

	Formation factor $F_f \cdot 10^5$ **Central value**	**Low**	**High**	**Porosity %** **Central value**	**Low**	**High**
Forsmark	3.8	1.3	12	0.1	0.06	0.3
Simpevarp	4.2	1.4	13	0.08	0.05	0.25
Laxemar	2.9	1.0	9	0.1	0.06	0.3

Table 5-11. Recommended value for diffusivities in free solution.

Element/specie	**Diffusivity in free solution $D_w \cdot 10^9$ (m^2/s)**	**Possible ion exclusion/surface diffusion effects in low salinity waters**
HTO	2.4	
Ag	1.7	
Br	2.0	Ion exclusion
C/CO_3	1.2	Ion exclusion
Cd	0.72	
Cl	2.0	Ion exclusion
Co	0.7	
Cs	2.1	Surface diffusion
I	0.83	Ion exclusion
Na	1.3	Surface diffusion
Ni	0.68	
Ra	0.89	Surface diffusion
Sr	0.79	Surface diffusion
Th	0.15	
All other nuclides	1.0	

From /Ohlsson and Neretnieks 1997/.

6 Concluding remarks

The in situ measurements of electrical conductivity have made it possible to obtain data for the rock under its (practically) virgin stress condition. This eliminates many uncertainties that would be introduced if laboratory data were to be converted to in situ field conditions. The porosity of the matrix has a smaller impact on PA results and should be of even less concern.

The recommended data will not change the PA results much compared to SR-97 results. Possibly there will be a higher confidence that the matrix diffusion data are fairly well bounded.

The major uncertainty regarding matrix diffusion effects is due to the uncertainties in assessing the FWS (Flow-Wetted Surface), i.e. the flowing water contacts.

In the companion report on sorption data /Crawford et al. 2006/ and their uncertainties it is shown that these uncertainties are larger than those for matrix diffusion and porosity data.

The impact of uncertainties of D_e, K_d and FWS are seen in Equation (2.7). An uncertainty in D_e and K_d influences the nuclide travel time in a linear way whereas the uncertainty in FWS influences by the square.

7 References

Allard B, Larson S A, Tullborg E-L, Wikberg P, 1983. Chemistry of deep groundwaters from granitic bedrock. KBS TR-83-59, Swedish Nuclear Fuel Supply Co/Division KBS, Stockholm, Sweden.

Andersson J, 1999. Data and data uncertainties. Compilation of data and data uncertainties for radionuclide transport calculations. SKB TR-99-09, Svensk Kärnbränslehantering AB.

Autio J, Kirkkomäti T, Siitari-Kauppi M, Timonen J, Laajalahti M, Aaltonen T, Maaranen J, 1999. Äspö Hard Rock Laboratory. Use of the ^{14}C-PMMA and He-gas methods to characterise excavation disturbance in crystalline rock. SKB IPR-99-18, Svensk Kärnbränslehantering AB.

Bear J, 1972. Dynamics of Fluids in Porous Media. Dover Publications, Inc.

Bear J, Tsang C-F, de Marsily G (ed), 1993. Flow and Contaminant Transport in Fractured Rock. Academic Press, Inc.

Bergman T, Rudmark L, Wahlgren C-H, Johansson R, Isaksson H, Stanfors R, 2000. Förstudie Oskarshamn. Kompletterande geologiska studier. SKB R-00-45, Svensk Kärnbränslehantering AB. (In Swedish).

Birgersson L, 1988. Diffusion in the matrix of granitic rock. Field test in the Stripa Mine. Licentiate Thesis, Dept. of Chemical Engineering, Royal Institute of Technology, Stockholm, Sweden.

Birgersson L, Neretnieks I, 1988. Diffusion in the matrix of granitic rock – field test in the Stripa mine. Mat. Res. Soc. Symp. Proc., 112, 189–198.

Birgersson L, Neretnieks I, 1990. Diffusion in the matrix of granitic rock: field test in the Stripa mine. Water Resour. Res., 26, 2833–2842.

Bodin J, Delay F, de Marsily G, 2003a. Solute transport in a single fracture with negligible matrix permeability: 1. Fundamental mechanisms. Hydrogeology J., 11, 418–433.

Bodin J, Delay F, de Marsily G, 2003b. Solute transport in a single fracture with negligible matrix permeability: 2. mathematical formation. Hydrogeology J., 11, 434–454.

Brandberg F, Skagius K, 1991. Porosity, sorption and diffusivity data compiled for the SKB 91 study. SKB TR-91-16, Svensk Kärnbränslehantering AB.

Bradbury M H, Lever D, Kinsley D, 1982. Aqueous phase diffusion in crystalline rock. Scientific Basis for Nuclear Waste Management V, Vol. 11, Berlin (Germany), Ed: W. Lutze.

Bradbury M H, Green A, 1985. Measurement of important parameters determining aqueous phase diffusion rates through crystalline rock matrix. J. Hydrol., 82, 39–55.

Bradbury M H, Green A, 1986. Investigations into the factors influencing long range matrix diffusion rates and pore spece accessibility at depth in granite. J. Hydrol., 89, 123–139.

Byegård J, Johansson H, Skålberg M, 1998. The interaction of sorbing and non-sorbing tracers with different Äspö rock types. Sorption and diffusion experiments in the laboratory scale. SKB TR-98-18, Svensk Kärnbränslehantering AB.

Byegård J, Skålberg M, Widestrand H, Tullborg E-L, 2002. Effects of heterogeneous porosity on retention: Results from the TRUE laboratory experiments. Radionuclide retention in geologic media, workshop proceedings. Nuclear Energy Agency. ISBN 92-64-19695-1.

Carlson L, Platz D, 1985. Permeability, porosity and retardation characteristics of granite samples from the Boettstein borehole – a laboratory study. Nagra Technical Report NTB 84-24, National Cooperative for the Disposal of Radioactive Waste, Baden, Switzerland.

Carlsten S, Petersson J, Stephens M, Mattsson H, Gustafsson J, 2004a. Geological single-hole interpretation of KFM02A and HFM04-05 (DS2). Site Investigation Report. SKB P-04-117, Svensk Kärnbränslehantering AB.

Carlsten S, Petersson J, Stephens M, Thunehed H, Gustafsson J, 2004b. Geological single-hole interpretation of KFM03B, KFM03A and HFM06–08 (DS3). Forsmark site investigation. SKB P-04-118, Svensk Kärnbränslehantering AB.

Cliffe K A, Morris S T, Porter J D, 1999. Assessment Model Validity Document; NAMMU: A program for calculating groundwater flow and transport through porous media. SKB R-99-51, Svensk Kärnbränslehantering AB.

Cramer J J, Melnyk T W, Miller H G, 1997. In situ diffusion in grantite: results from scoping experiments. COG-96-656-1, AECL-11756, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada.

Crank J, 1975. The Mathematics of Diffusion, 2nd Ed. Oxford University Press.

Crawford J, Neretnieks I, Malmström M, 2006. Data and uncertaintiy assessment for radionuclide K_d partitioning coefficients in granitic rock for use in SR-Can calculations. SKB R-06-75, Svensk Kärnbränslehantering AB.

Curtis P, Elfström M, Stanfors R, 2003. Compilation of structural geological data covering the Simpevarp peninsula, Ävrö and Hålö. Oskarshamn site investigation. SKB P-03-07, Svensk Kärnbränslehantering AB.

Dullien F A L, 1992. Porous Media: Fluid Transport and Pore Structure, 2nd Ed. Academic Press, Inc.

Elert M, Gylling B, Lindgren M, 2004. Assessment model validity document FARF31. SKB R-04-51, Svensk Kärnbränslehantering AB.

Eriksen, 1985. Migration in single fissures. SKB Progress Report AR-85-07, Svensk Kärnbränslehantering AB.

Gylling B, 1997. Development and applications of the channel network model for simulations of flow and solute transport in fractured rock. Doctoral Thesis, Dept. of Chemical Engineering and Technology, Royal Institute of Technology, Stockholm, Sweden. ISBN 91-7170-190-7.

Hartikainen J, Hartikainen K, Pietarila H, Timonen J, 1995a. Permeability and diffusivity measurements with the He-gas method of disturbed zone in rock samples cored from the full-scale experimental deposition holes in the VTO Research Tunnel. Nuclear Waste Commission of Finnish Power Companies. Report YTJ-95-16, Helsinki, Finland.

Hartikainen K, Hautojärvi A, Pietarila H, Timonen J, 1995b. Diffusion measurements on crystalline rock matrix. Mat. Res. Soc. Symp. Proc., 353, 435–440.

Hartikainen J, Hartikainen K, Hautojärvi A, Kuoppamäki K, Timonen J, 1996. Helium gas method for rock characteristics and matrix diffusion. Posiva Oy. Report POSIVA-96-22, Helsinki, Finland.

Hartikainen K, Hartikainen J, 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 Oy. Report POSIVA-97-13, Helsinki, Finland.

Hartley L J, Holton D, Hoch A R, 2002. NAPSAC (release 4.3). Technical summary document, SERCO/ERRA-N/TSD02v1.

Hartley L J, Holton D, 2003. CONNECTFLOW (release 2.0). Technical summary document, SERCO/ERRA-C/TSD02v1.

Hartley L, Cox I, Holton D, Hunter F, Joyce S, Gylling B, Lindgren M, 2004. Groundwater flow and radionuclide transport modelling using CONNECTFLOW in support of the SR-Can assessment. SKB R-04-61, Svensk Kärnbränslehantering AB.

Harvey K B, 1996. Measurements of diffusive properties of intact rock. AECL-11439, COG-95-456-I. Atomic Energy of Canada Limited, Canada.

Hedin A, 2002. Safety Assessment of a Spent Nuclear Fuel Repository: Sensitivity Analyses for Prioritisation of Research. Proceedings of the 6th International Conference on Probabilistic Safety Assessment and Management*, PSAM6*. Elsevier Science Ltd.

Hedin A, 2003. Probabilistic dose calculations and sensitivity analyses using analytic models. Reliability Engineering and System Safety, 79, 195–204.

Hora S, 2002. Expert opinion in SR 97 and the SKI/SSI joint review of SR 97. SSI report 2002:20, Swedish Radiation Protection Authority, Stockholm, Sweden.

Hora S, Jensen M, 2002. Expert judgement elicitation. SSI report 2002:19, Swedish Radiation Protection Authority, Stockholm, Sweden.

Idemitsu K, Furuya H, Hara T, Inagaki Y, 1992. Migration of cesium, strontium and cobalt in water-saturated inada granite. J. Nucl. Sci. Technol., 29, 454–460.

Ittner T, Torstenfelt B, Allard B, 1988. Migration of the fission products strontium, technetium, iodine, cesium and the actinides neptunium, plutonium, americium in granitic rock. SKB TR-88-02, Svensk Kärnbränslehantering AB.

Ittner T, Torstenfelt B, Allard B, 1990. Diffusion of strontium, iodine and cesium in granitic rock. Radiochim. Acta, 49, 101–106.

Jacob A, 2004. Matrix diffusion for performance assessment – Experimental evidence, modelling assumptions and open issues. Nagra Technical Report 04-07, National Cooperative for the Disposal of Radioactive Waste, Wettingen, Switzerland; PSI Bericht Nr. 04-08, Paul Scherrer Institut,Willigen, Switzerland.

JNC, 2000. H12: Project to establish the scientific and technical basis for HLW disposal in Japan, Supporting report 3, Safety assessement of the geological disposal system. JNC TN1410, Japan Nuclear Cycle Development Institute, Tokai-Mura, Japan.

Kaukonen V, Hakanen M, Lindberg A, 1993. Sorption of Cs, U, Np and Pu and diffusion of water, Cs and Np in basic plutonic rocks and vulcanite. Dept. of Radiochemistry, Unversity of Helsinki, Finland.

Kaukonen V, Puukko E, Hakanen M, Lindberg A, 1995. Diffusion of neptunium in Olkiluoto mica gneiss and pegmatite and in Kivetty granite. Work Report TURVA-95-05, Nuclear Waste Commission of Finnish Power Companies, Helsinki, Finland.

Kaukonen V, Hakanen M, Lindberg A, 1996. Diffusion and sorption of HTO, Np, Na and Cl in rocks of Kivetty and Olkiluoto and in their main minerals. TURVA-96-03, Posiva Oy, Helsinki, Finland.

Kaukonen V, Hakanen M, Lindberg A, 1997. Diffusion and sorption of HTO, Np, Na and Cl in rocks and minerals of Kivetty and Olkiluoto. Posiva-97-07, Posiva Oy, Helsinki, Finland.

Lehikoinen J, Muurinen A, Olin M, Uusheimo K, Valkiainen M, 1992. Diffusivity and porosity studies in rock matrix. The effect of salinity. Technical Research Centre of Finland. Espoo, Finland.

Löfgren M, 2001. Formation Factor Logging in Igneous Rock by Electrical Methods. Licentiate Thesis, Dept. of Chemical Engineering and Technology, Royal Institute of Technology, Stockholm, Sweden. ISBN: 91-7283-207-X.

Löfgren M, Ohlsson Y, Neretnieks I, 2001. Formation Factor Determinations by In situ Resistivity Logging. Mat. Res. Soc. Symp. Proc., 663.

Löfgren M, Neretnieks I, 2002. Formation factor logging in situ by electrical methods. Background and methodology. SKB TR-02-27, Svensk Kärnbränslehantering AB.

Löfgren M, Neretnieks I, 2003. Formation factor logging by electrical methods. Comparison of formation factor logs obtained in situ and in the laboratory. J. Contam. Hydrol., 61, 107–115.

Löfgren M, 2004. Diffusive properties of granitic rock as measured by in situ electrical methods. Doctoral Thesis, Dept. of Chemical Engineering and Technology, Royal Institute of Technology, Stockholm, Sweden. ISBN 91-7283-935-X.

Löfgren M, Neretnieks I, 2005a. Formation factor logging in situ by electrical methods in KSH01A and KSH02. Measurements and evaluation of methodology. Site Investigation Report. SKB P-05-27, Svensk Kärnbränslehantering AB.

Löfgren M, Neretnieks I, 2005b. Formation factor logging in situ by electrical methods in KFM01A and KFM02A. Measurements and evaluation of methodology. Site Investigation Report. SKB P-05-29, Svensk Kärnbränslehantering AB.

Löfgren M, Neretnieks I, 2006. Through-electromigration: A new method of investigating pore connectivity and obtaining formation factors. J. Contam. Hydrol., 87, 237–252.

Maes N, Moors H, Dierckx A, De Cannière P, Put M, 1999. The assessment of electromigration as a new technique to study diffusion of radionuclides in clayey soils. J. of Contaminant Hydrology, 36, 231–247.

Miller W, Alexander R, Chapman N, McKinley I, Smellie J, 1994. Natural Analogue Studies in the Geological Disposal of Radioactive Waste. Studies in Environmental Science 57. Elsevier.

Morris S T, Cliffe K A, 1994. Verification of HYDRASTAR: Analysis of hydraulic conductivity fields and dispersion. SKB TR-94-21, Svensk Kärnbränslehantering AB.

Möri A, Mazurek M, Adler M, Schild M, Siegesmund S, Vollbrecht A, Ota K, Ando T, Alexander W R, Smith P A, Haag P, Bühler Ch, 2003. The Nagra-JNC in situ study of safety relevant radionuclide retardation in fractured crystalline rock, IV: The in situ study of matrix porosity in the vicinity of a water conducting fracture. Nagra Technical Report 00-08, National Cooperative for the Disposal of Radioactive Waste, Wettingen, Switzerland.

Neretnieks I, 1980. Diffusion in the rock matrix: an important factor in radionuclide retardation? J. Geophys. Res., 85, 4379–4397.

Neretnieks I, 2006. Fast method for simulation of radionuclide chain migration in dual porosity fractured rocks. J. Contam. Hydrol., 88, 269–288.

Nordstrom D K, Andrews J N, Carlsson L, Fontes J-C, Fritz P, Moser H, Olsson T, 1985. Hydrogeological and hydrogeochemical investigations in boreholes – Final report of the phase I geochemical investigations of the Stripa groundwaters. SKB Stripa Project 85-06, Svensk Kärnbränslehantering AB.

Norman S, Kjellbert N, 1990. FARF31 – A far field radionuclide migration code for use with the PROPER package. SKB TR-90-01, Svensk Kärnbränslehantering AB.

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

Ohlsson Y, Neretnieks I, 1997. Diffusion data in granite. Recommended values. SKB TR-97-20, Svensk Kärnbränslehantering AB.

Ohlsson Y, Neretnieks I, 1998. Some evidence for surface ion mobility in rock. J. Contam. Hydrol., 35, 91–100.

Ohlsson Y, Neretnieks I, 1999. A study of rock matrix diffusion properties by electrical conductivity measurements. Mat. Res. Soc. Symp. Proc., 556.

Ohlsson Y, 2000. Studies of Ionic Diffusion in Crystalline Rock. Doctoral Thesis, Dept. of Chemical Engineering and Technology, Royal Institute of Technology, Stockholm, Sweden.

Ohlsson Y, Neretnieks I, 2001. Matrix diffusion measurements through-diffusion versus electrical conductivity measurements. Mat. Res. Soc. Symp. Proc., 663.

Ohlsson Y, Löfgren M, Neretnieks I, 2001. Rock matrix diffusivity determinations by in situ electrical conductivity measurements. J. Contam. Hydrol., 47, 117–125.

Olin M, Valkiainen M, Aalto H, 1997. Matrix diffusion in crystalline rocks: coupling of anion exclusion, surface diffusion and surface complexation. Posiva-96-25, Posiva Oy, Helsinki, Finland.

Ota K, Möri A, Alexander W R, Frieg B, Schild M, 2003. Influence of the mode of matrix porosity determined on matrix diffusion calculations. J. of Contam. Hydrol., 61, 131–145.

Petruzzelli D, Helfferich F G (ed.), 1993. Migration and Fate of Pollutants in Soils and Subsoils. NATO ASI Series: Series G: Ecological Sciences, Vol. 32. Springer-Verlag.

Rasilainen K, 1997. Matrix diffusion model – In situ tests using natural analogues. Technical Research Centre of Finland, Espoo, VTT Publications 331.

RETROCK, 2004. Treatment of geosphere retention phenomena in safety assessments. Scientific basis for retention processes and their implementation in safety assessment models (WP2). SKB R-04-48, Svensk Kärnbränslehantering AB.

Sahimi M, 1995. Flow and Transport in Porous Media and Fractured Rock – from Classical Methods to Modern Appraoches. VCH Verlagsgesellschaft mbH, Weinheim, Germany.

Sato H, Shibutani T, Yui M, 1996. Experimental and modelling studies on diffusion of Cs, Ni and Sm in granodiorite, basalt and mudstone. J. of Contaminant Hydrology, 26, 119–133.

Skagius K, Neretnieks I, 1982. Diffusion in crystalline rocks. Scientific Basis for Nuclear Waste Management V, Vol. 11. Berlin (Germany), Ed. W. Lutze.

Skagius K, Svedberg G, Neretnieks I, 1982. A study of strontium and cesium sorption on granite. Nuclear Technology, 59, 302–313.

Skagius K, Neretnieks I, 1983. Diffusion measurements in crystalline rocks. SKB TR-83-15, Svensk Kärnbränslehantering AB.

Skagius K, Neretnieks I, 1985. Diffusivity measurements and electrical resistivity measurements in rock samples under mechanical stress. SKB TR-85-25, Svensk Kärnbränslehantering AB.

Skagius K, 1986. Diffusion of Dissolved Species in the Matrix of Some Swedish Crystalline Rocks. Doctoral Thesis, Dept. of Chemical Engineering, Royal Institute of Technology, Stockholm, Sweden.

Skagius K, Neretnieks I, 1986. Porosities and diffusivities of some nonsorbing species in crystalline rocks. Water Resour. Res., 22, 389–397.

Skagius K, Neretnieks I, 1988. Measurement of Cesium and Strontium in biotite gneiss. Water Resour. Res., 24, 75–84.

SKB, 1999. SR 97 – Post-closure safety. SKB TR-99-06, Svensk Kärnbränslehantering AB.

SKB, 2006. Site descriptive modelling, Forsmark stage 2.1, Feedback for completion of the site investigation including input from safety assessment and repository engineering. SKB R-06-38, Svensk Kärnbränslehantering AB.

SKB, 2006a. Long-term safety for KBS-3 repositories at Forsmark and Laxemar – a first evalution. Main report of the SR-Can project. SKB TR-06-09, Svensk Kärnbränslehantering AB.

SKB, 2006b. Geosphere process report for the safety assessment SR-Can. SKB TR-06-19, Svensk Kärnbränslehantering AB.

SKI, 1996. SITE-94, deep repository performance assessment project. SKI Report 96:36, Swedish Nuclear Power Inspectorate, Stockholm, Sweden.

SKI and SSI, 2001. SKI's and SSI's joint review of SKB's safety assessment report, SR 97, Summary. SKI Report 01:3, Swedish Nuclear Power Inspectorate, Stockholm, Sweden; SSI Report 2001:02, Swedish Radiation Protection Authority, Stockholm, Sweden.

Smith P A, 1990. Modelling of a diffusion-sorption experiment by linear and nonlinear sorption isotherms. Nucl. Technol., 92, 363–373.

Svensson U, Kuylenstierna H-L, Ferry M, 2004. DarcyTools, version 2.1 – Concepts, methods, equations and demo simulations. SKB R-04-19, Svensk Kärnbränslehantering AB.

Thunehed H, Keisu M, 2004. Interpretation of borehole geophysical measurements in KFM05A, HFM14, HFM15 and HFM19. Forsmark site investigation. SKB P-04-154, Svensk Kärnbränslehantering AB.

Thunehed H, 2005. Forsmark site investigation: Resistivity measurements on samples from KFM01A and KFM02A. SKB P-05-26, Svensk Kärnbränslehantering AB.

Vahlund F, Hermansson H, 2004. A direct numerical approach to solving the transport equations for radionuclide transport in fractured rock. SKB R-04-50, Svensk Kärnbränslehantering AB.

Valkiainen M, 1992. Diffusion in the rock matrix – a review of laboratory tests and field studies. YTJ-92-04, Nuclear Waste Commission of Finnish Power Companies, Helsinki, Finland.

Valkiainen M, Aalto H, Lindberg A, Olin M, Siitari-Kauppi M, 1995. Diffusion in the matrix of rocks from Olkiluoto – The effect of anion exclusion. YJT-95-20, Nuclear Waste Commission of Finnish Power Companies, Helsinki, Finland.

Valkiainen M, Aalto H, Lehikoinen J, Uusheimo, 1996. The effect of thickness in through-diffusion experiments. Technical Research Center of Finland. VTT Research notes 1788, ISBN 951-38-4983-X.

Vilks P, Cramer J J, Jensen M, Miller N H, Miller H G, Stanchell F W, 2003. In situ diffusion experiment in granite: phase I. J. Contam. Hydrol., 61, 191–202.

Vilks P, Miller N H, Stanchell F W, 2004. Phase II in situ diffusion experiment. Report No: 06819-REP-01200-10128-R00. Ontario Power Generation, Nuclear Waste Management Division, Toronto, Ontario, Canada.

Wilmot R D, Galson D A, 2000. Expert judgement in performance assessment. SKI report 2000:04, Swedish Nuclear Power Inspectorate, Stockholm, Sweden.

Wilmot R D, Galson D A, Hora S C, 2000. Expert judgements in performance assessments. Report of an SKI/SSI Seminar. SKI report 2000:35, Swedish Nuclear Power Inspectorate, Stockholm, Sweden.

Yamaguchi T, Sakamoto Y, Senoo M, 1993. Consideration on effective diffusivity of strontium in granite. J. Nucl. Sci. Tech., 30, 796–803.

Yamaguchi T, Sakamoto Y, Nakayama S, Vandergraaf T T, 1997. Effective diffusivity of the uranyl ion in a granite from Inada, Ibaraki, Japan. J. Contam. Hydrol., 26, 109–117.

Yamaguchi T, Nakayama S, 1998. Diffusivity of U, Pu and Am carbonate complexes in a granite from Inada, Ibaraki, Japan studied by through diffusion. J. Contam. Hydrol., 35(1–3), 55–65.

Zheng C M, Bennett G D, 2002. Applied Contaminant Transport Modelling. Wiley-Interscience.

Appendix I

Table A1. Literature data of porosity, bulk diffusivity, effective diffusivity, rock storage capacity and formation factor for HTO.

Rock type	Water type and ionic strength (M)	ε (%)	$D_w(m^2\ s^{-1})$	$D_e(m^2\ s^{-1})$	α (%)	F_f	Ref
			2.40E–09				/Skagius and Neretnieks 1986/
	Distilled water		1.70E–09				/Kaukonen et al. 1997/
	Olkiluoto, 0.26–0.31		1.90E–09				
Granite, Finnsjön, Sweden	Allard, 0.0085	0.18–0.21		1.80E–13		7.50E–05	/Skagius 1986/
	Distilled water			1.30E–13		5.42E–05	
Granite, Kivetty, Finland	Ten times Dilluted Allard, 0.00085	0.05–0.16		3.90E–13	3.3	1.63E–04	/Kaukonen et al. 1997/
				3.60E–13	0.81	1.50E–04	
				1.70E–13	5.5	7.08E–05	
				1.70E–13	0.11	7.08E–05	
				2.20E–13	3.1	9.17E–05	
				1.40E–13	0.03	5.83E–05	
				1.70E–13	6.4	7.08E–05	
				1.80E–13	0.94	7.50E–05	
				2.70E–13	6.1	1.13E–04	
				2.40E–13	1.4	1.00E–04	
				2.40E–13	2.1	1.00E–04	
				3.90E–13	0.94	1.63E–04	
		0.1		5.00E–13		2.08E–04	/Kaukonen et al. 1997/
				4.70E–13		1.96E–04	
				2.10E–13		8.75E–05	
				2.00E–13		8.33E–05	
				3.00E–13		1.25E–04	
				2.00E–13		8.33E–05	
				2.30E–13		9.58E–05	
				2.20E–13		9.17E–05	
				3.20E–13		1.33E–04	
				3.00E–13		1.25E–04	
				3.50E–13		1.46E–04	
				3.90E–13		1.63E–04	
Arithmetic mean				**2.67E–13**		**1.11E–04**	
Standard Deviation				**1.02E–13**		**4.24E–05**	

Rock type	Water type and ionic strength (M)	ε (%)	D_w(m^2 s^{-1})	D_e(m^2 s^{-1})	α (%)	F_f	Ref
Pegmatite, Olkiluoto, Finland	Olkiluoto, 0.26–0.31	0.13–0.39		8.60E–13	0.88	4.53E–04	/Kaukonen et al. 1997/
				9.30E–13	1.1	4.89E–04	
				1.10E–12	0.73	5.79E–04	
				6.50E–13	0.82	3.42E–04	
				9.80E–13	0.1	5.16E–04	
				1.40E–12	0.45	7.37E–04	
				1.30E–12	0.83	6.84E–04	
Pegmatite, Olkiluoto, Finland	Olkiluoto, 0.26–0.31	0.1		8.20E–13		4.32E–04	
				1.20E–12		6.32E–04	
				1.60E–12		8.42E–04	
				5.00E–13		2.63E–04	
				1.10E–12		5.79E–04	
				1.10E–12		5.79E–04	
				1.20E–12		6.32E–04	
Arithmetic mean				**1.05E–12**		**5.54E–04**	
Standard Deviation				**2.82E–13**		**1.54E–04**	
Mica gneiss, Olkiluoto, Finland	Olkiluoto, 0.26–0.31	0.05–0.06		1.40E–13	2.2	7.37E–05	/Kaukonen et al. 1997/
				9.80E–14	6.6	5.16E–05	
				1.40E–13	0.45	7.37E–05	
				7.70E–14	7	4.05E–05	
				1.20E–13	0.43	6.32E–05	
				2.00E–13	0.45	1.05E–04	
				5.60E–14	0.24	2.95E–05	
				1.50E–13	0.37	7.89E–05	
				1.30E–13	3.8	6.84E–05	
				1.50E–13	1.2	7.89E–05	
				5.30E–13	1	2.79E–04	
				1.00E–12	2.4	5.26E–04	
		0.05		1.40E–13		7.37E–05	
				1.00E–13		5.26E–05	
				1.70E–13		8.95E–05	
				1.00E–13		5.26E–05	
				1.60E–13		8.42E–05	
				1.90E–13		1.00E–04	
				7.50E–13		3.95E–04	
				1.30E–12		6.84E–04	
Gneiss, Hästholmen. Finland	NaCl, 0.0044			9.60E–14		4.00E–05	/Lehikoinien et al. 1992/
	NaCl, 0.7			8.50E–14		3.54E–05	/Lehikoinien et al. 1992/
	NaCl, 1.0			7.60E–14		3.17E–05	/Lehikoinien et al. 1992/
Arithmetic mean				**2.59E–13**		**1.35E–04**	
Standard Deviation				**3.24E–13**		**1.71E–04**	

Rock type	Water type and ionic strength (M)	ε (%)	D_w(m^2 s^{-1})	D_e(m^2 s^{-1})	α (%)	F_f	Ref
Plutonic, Mäntsäälä, Finland	Allard, 0.0085	0.2		9.40E–14	0.17	4.95E–05	/Kaukonen et al. 1993/
				5.40E–13	0.36	2.84E–04	
Plutonic, Syyry, Finland	Syyry	0.26–0.43		5.80E–14	0.17	3.05E–05	
				5.70E–14	0.11	3.00E–05	
				9.10E–14	0.23	4.79E–05	
Vulcanite	Allard, 0.0085	0.27		6.35E–13	0.31	3.34E–04	
				2.68E–13	0.25	1.41E–04	
				3.67E–13	0.31	1.93E–04	
Arithmetic mean				**2.64E–13**		**1.39E–04**	
Standard Deviation				**2.29E–13**		**1.21E–04**	

Table A2. Literature data of porosity, bulk diffusivity, effective diffusivity, rock storage capacity and formation factor for Cr-EDTA.

Rock type	Water type and ionic strength (M)	ε (%)	D_w(m^2 s^{-1})	D_e(m^2 s^{-1})	α (%)	F_f	Ref
			4.20E–10				/Skagius 1986/
Granite, Finsjön, Sweden	Distilled water	0.21–0.22		6.90E–15		1.64E–05	/Skagius 1986/
				6.20E–15	0.04	1.48E–05	
Granite, Gideå, Sweden	Distilled water	0.18–0.19		1.60E–14	0.04	3.80E–05	
		0.17–0.18		2.30E–14	0.08	5.50E–05	
Magmatite granite, Svartboberget, Sweden	Distilled water	0.28–0.31		4.90E–14	0.29	1.17E–04	
		0.22–0.26		2.90E–14	0.13	6.90E–05	
Granite, Stripa, Sweden	Stripa groundwater, 0.006	0.3		1.50E–14		3.57E–05	/Birgersson 1988/
		0.3		6.00E–14		1.43E–04	
		0.3		3.00E–14		7.14E–05	
		0.3		3.00E–14		7.14E–05	
		0.3		3.00E–14		7.14E–05	
		0.3		1.50E–14		3.57E–05	
		0.3		1.50E–14		3.57E–05	
		0.3		1.50E–14		3.57E–05	
		0.3		6.00E–14		1.43E–04	
		0.3		1.50E–14		3.57E–05	
Arithmetic mean				**2.59E–14**		**6.18E–05**	
Standard Deviation				**1.71E–14**		**4.07E–05**	
Biotite gneiss, Svartboberget, Sweden	Distilled water	0.24–0.28		2.50E–14		6.00E–05	/Skagius 1986/
		0.30–0.34		3.00E–14		7.10E–05	
Biotite gneiss, Fjällveden, Sweden	Distilled water	0.27–0.28		8.10E–15	0.14	1.90E–05	
		0.26–0.28		6.00E–15	0.08	1.40E–05	
Magmatite, Studsvik, Sweden	Distilled water	0.20–0.21		9.40E–15	0.03	2.20E–05	/Skagius 1986/
Arithmetic mean				**1.57E–14**		**3.72E–05**	
Standard Deviation				**1.10E–14**		**2.63E–05**	
Granite + Fissure coatings, Finnsjön, Sweden	Distilled water	1.43–1.58		1.00E–14	0.8	2.40E–05	/Skagius 1986/
		1.03–1.19		6.30E–15	0.36	1.50E–05	
Fissure coatings, Finnsjön, Sweden	Distilled water	3.51–3.61		4.80E–13	2.83	1.14E–03	/Skagius 1986/
Arithmetic mean				**1.65E–13**		**3.94E–04**	
Standard Deviation				**2.72E–13**		**6.49E–04**	

Table A3. Literature data of porosity, bulk diffusivity, effective diffusivity, rock storage capacity and formation factor for uranin.

Rock type	Water type and ionic strength (M)	ε (%)	D_w(m^2 s^{-1})	D_e(m^2 s^{-1})	α (%)	F_f	Ref
			4.5E–10				/Skagius 1986/
Granite, Finnsjön, Sweden	Distilled water			2.20E–15	0.02	5.00E–06	/Skagius 1986/
		0.20–0.21		5.30E–15	0.02	1.20E–05	
Granite, Gideå, Sweden	Distilled water	0.15–0.17		1.30E–14	0.11	2.90E–05	
		0.18–0.19		1.30E–14	0.12	2.90E–05	
Magmatite granite, Svartboberget, Sweden	Distilled water	0.22–0.26		6.90E–14	0.3	1.53E–04	
		0.30–0.34		5.20E–14	0.94	1.16E–04	
Granite, Stripa, Sweden	Stripa groundwater, 0.006	0.3		6.00E–13		1.33E–03	/Birgersson 1988/
		0.3		1.05E–13		2.33E–04	
		0.3		1.05E–13		2.33E–04	
		0.3		1.80E–14		4.00E–05	
		0.3		1.80E–14		4.00E–05	
		0.3		1.80E–14		4.00E–05	
		0.3		1.80E–14		4.00E–05	
		0.3		1.80E–14		4.00E–05	
		0.3		1.80E–14		4.00E–05	
		0.33		6.60E–14		1.47E–04	
		0.33		2.20E–14		4.89E–05	
		0.3		9.00E–15		2.00E–05	
		0.3		1.00E–14		2.22E–05	
		0.3		2.00E–14		4.44E–05	
Granite		2.3–3.4		4.03E–13		8.96E–04	/Eriksen 1985/
				2.94E–13		6.53E–04	
				2.94E–13		6.53E–04	
Arithmetic mean				**9.52E–14**		**2.12E–04**	
Standard Deviation				**1.54E–13**		**3.43E–04**	
Gneiss, Gideå, Sweden	Distilled water	0.11–0.12		3.20E–15	0.11	7.10E–06	/Skagius 1986/
Gneiss, Svartboberget, Sweden	Distilled water	0.08–0.09		9.30E–15	0.07	2.07E–05	
		0.08–0.10		9.60E–15	0.07	2.13E–05	
Biotite gneiss, Svartboberget, Sweden	Distilled water	0.30–0.35		7.80E–14	0.52	1.73E–04	
		0.23–0.28		6.00E–14	0.62	1.33E–04	
Biotite gneiss, Fjällveden, Sweden	Distilled water	0.23–0.24		3.60E–15	0.02	8.00E–06	
		0.23–0.24		3.90E–15	0.04	9.00E–06	
Magmatite, Studsvik, Sweden	Distilled water	0.21–0.22		2.30E–14	0.22	5.10E–05	/Skagius 1986/
Arithmetic mean				**2.38E–14**		**5.29E–05**	
Standard Deviation				**2.90E–14**		**6.43E–05**	
Granite + Fissure coatings, Finnsjön, Sweden	Distilled water	3.00–3.31		1.30E–14	0.1	2.90E–05	/Skagius 1986/
		1.28–1.35		6.20E–14	3.5	1.38E–04	
		0.78–1.00		5.00E–14	2.5	1.11E–04	
				1.60E–11	24	3.56E–02	
				2.00E–11	24	4.44E–02	
		7.12–7.49		4.00E–11	8.27	8.89E–02	
		7.12–7.49		9.00E–12		2.00E–02	
		1.17–1.25		7.00E–15	0.27	1.60E–05	
		1.67–1.79		1.50E–12	2.8	3.33E–03	

Rock type	Water type and ionic strength (M)	ε (%)	D_w(m^2 s^{-1})	D_e(m^2 s^{-1})	α (%)	F_f	Ref
Fissure coatings, Finnsjön, Sweden	Distilled water			6.00E–14	1.1	1.33E–04	/Skagius 1986/
				9.00E–14		2.00E–04	
		2.67–2.79		2.20E–13	1.3	4.89E–04	
Arithmetic mean				**7.25E–12**		**1.61E–02**	
Standard Deviation				**1.25E–11**		**2.77E–02**	

Table A4. Literature data of porosity, bulk diffusivity, effective diffusivity, rock storage capacity and formation factor for iodide.

Rock type	Water type and ionic strength (M)	ε (%)	D_w(m^2 s^{-1})	D_e(m^2 s^{-1})	α (%)	F_f	Ref
			1.6E–09				/Skagius 1986/
Granite, Ossian, the UK	$NaNO_3$, 0.1 M			1.4E–12	1.2	7.00E–04	/Bradbury and Green 1985/
				1.4E–12	1.6	7.00E–04	
				1.4E–12	2.2	7.00E–04	
				1.6E–12	1.2	8.00E–04	
				9.4E–13	0.74	5.00E–04	
				7.5E–13	0.31	4.00E–04	
				6.8E–13	0.65	3.00E–04	
				6.4E–13	0.57	3.00E–04	
Granite, Scottish lowland, the UK	$NaNO_3$, 0.1 M			4.0E–13	0.22	2.00E–04	
				2.7E–13	0.27	3.00E–04	
Granite, Skene complex, the UK	$NaNO_3$, 0.1 M			2.8E–13	0.12	1.00E–04	
				7.7E–13	0.38	4.00E–04	
				5.9E–13	0.21	3.00E–04	
				7.7E–13	0.36	4.00E–04	
				5.7E–13	0.27	3.00E–04	
				6.6E–13	0.29	3.00E–04	
				7.4E–13	0.37	4.00E–04	
				6.6E–13	0.46	3.00E–04	
Granite, Cornish Carnmenellis, the UK	$NaNO_3$, 0.1 M			2.3E–14	0.06	1.20E–05	
				3.2E–14	0.088	1.60E–05	
Granite, Finnsjön, Sweden	1 M NaI and distilled water			8.40E–14	0.3	5.20E–05	/Skagius 1986/
				7.00E–14	0.2	4.40E–05	
	NaI and $NaNO_3$, 0.1 M	0.20–0.21		7.10E–14	0.27	4.40E–05	
		0.21–0.23		4.10E–14	0.18	2.60E–05	
Granite, Stripa, Sweden	1 M NaI and distilled water			1.60E–13	1.05	1.00E–04	
				1.30E–13	0.21	8.10E–05	
Granite, Gideå, Sweden	1 M NaI and distilled water, then 1M $NaNO_3$	0.16–0.17		1.00E–13	0.14	6.20E–05	
				1.30E–13	0.14	8.10E–05	
		0.15–0.16		9.00E–14	0.14	5.60E–05	
				1.10E–13	0.14	6.90E–05	
Granite. Svartboberget, Sweden	NaI and $NaNO_3$, 0.1 M	0.20–0.25		6.60E–13	1.14	4.12E–04	
		0.29–0.36		3.40E–13	0.58	2.12E–04	
		0.26–0.30		2.60E–13	0.28	1.62E–04	

Rock type	Water type and ionic strength (M)	ε (%)	$D_w(m^2\,s^{-1})$	$D_e(m^2\,s^{-1})$	α (%)	F_f	Ref
Young granite, Finnsjön, Sweden	NaI and $NaNO_3$, 0.1 M	0.27–0.34		3.90E–13	0.22	2.44E–04	/Skagius 1986/
		0.25–0.30		3.90E–13	0.09	2.44E–04	
		0.26–0.31		3.40E–13	0.2	2.12E–04	
		0.21–0.25		2.45E–13	0.15	1.53E–04	
		0.24–0.28		2.80E–13	0.11	1.75E–04	
		0.31–0.38		2.80E–13	0.17	1.75E–04	
		0.24–0.29		3.20E–13	0.1	2.00E–04	
		0.18–0.21		3.10E–13	0.24	1.94E–04	
		0.19–0.23		2.50E–13	0.13	1.56E–04	
		0.54–0.64		2.60E–13	0.37	1.62E–04	
		0.54–0.60		3.20E–13	0.32	2.00E–04	
		0.50–0.54		4.40E–13	0.26	2.75E–04	
		0.40–0.43		2.50E–13	0.38	1.56E–04	
		0.35–0.37		2.70E–13	0.34	1.69E–04	
		0.32–0.34		2.26E–13	0.34	1.41E–04	
		0.28–0.31		2.20E–13	0.36	1.38E–04	
		0.25–0.28		2.40E–13	0.28	1.50E–04	
		0.27–0.30		2.48E–13	0.41	1.55E–04	
		0.26–0.27		2.50E–13	0.37	1.56E–04	
		0.26–0.27		2.65E–13	0.47	1.66E–04	
		0.27–0.28		2.53E–13	0.41	1.58E–04	
		0.28–0.30		2.47E–13	0.37	1.54E–04	
		0.28–0.29		2.44E–13	0.32	1.52E–04	
		0.38–0.40		2.35E–13	0.34	1.47E–04	
		0.33–0.35		2.48E–13	0.37	1.55E–04	
		0.34–0.37		4.40E–13	0.65	2.75E–04	
Arithmetic mean				**4.12E–13**		**2.29E–04**	
Standard Deviation				**3.55E–13**		**1.74E–04**	
Granite, Stripa, Sweden	Stripa groundwater, 0.006	0.3		1.50E–12		9.38E–04	/Birgersson 1988/
		0.3		3.00E–13		1.88E–04	
		0.3		3.00E–13		1.88E–04	
		0.3		2.10E–13		1.31E–04	
		0.3		1.50E–12		9.38E–04	
		0.3		6.00E–14		3.75E–05	
		0.3		6.00E–14		3.75E–05	
		0.3		6.00E–14		3.75E–05	
		0.3		6.00E–14		3.75E–05	
		0.33		1.38E–13		8.63E–05	
		0.33		5.10E–13		3.19E–04	
		0.3		5.10E–13		3.19E–04	
		0.3		5.10E–14		3.19E–05	
		0.3		1.50E–13		9.38E–05	
Granite, Stripa, Sweden	Synthetic, 0.00309			3.20E–14		2.00E–05	/Ittner et al. 1998/
Granite, Studsvik, Sweden	Synthetic, 0.00744			1.50E–14		9.38E–06	/Ittner et al. 1998/
Arithmetic mean				**3.41E–13**		**2.13E–04**	
Standard Deviation				**4.79E–13**		**2.99E–04**	

Rock type	Water type and ionic strength (M)	ε (%)	D_w(m^2 s^{-1})	D_e(m^2 s^{-1})	α (%)	F_f	Ref
Gneiss, Karlshamn, Sweden	1 M NaI and distilled water			1.30E–13	0.74	8.10E–05	/Skagius 1986/
	NaI and $NaNO_3$, 0.1 M			4.80E–14	0.27	3.00E–05	
Gneiss, Gideå, Sweden	NaI and $NaNO_3$, 0.1 M	0.06		1.80E–14	0.05	1.12E–05	
Gneiss, Svartboberget, Sweden	NaI and $NaNO_3$, 0.1 M	0.07		1.90E–14	0.07	1.19E–05	
		0.07–0.09		3.50E–14	0.12	2.07E–05	
Biotite gneiss, Svartboberget, Sweden	NaI and $NaNO_3$, 0.1 M	0.29–0.34		1.50E–13	0.26	9.40E–05	
		0.28–0.32		3.60E–13	0.92	2.25E–04	
Gneiss, Fjällveden, Sweden	NaI and $NaNO_3$, 0.1 M	0.25–0.26		7.20E–14	0.36	4.50E–05	
		0.27–0.29		7.40E–14	0.15	4.60E–05	
Magmatite, Studsvik, Sweden	1 M NaI and distilled wate			1.30E–13	0.74	8.10E–05	/Skagius 1986/
	NaI and $NaNO_3$, 0.1 M			4.80E–14	0.27	3.00E–05	
Arithmetic mean				**9.85E–14**		**6.14E–05**	
Standard Deviation				**9.81E–14**		**6.14E–05**	
Granite + Fissure coatings, Finnsjön, Sweden	1 M NaI and distilled water			1.20E–13		7.50E–05	/Skagius 1986/
				6.00E–14		3.75E–05	
		2.57–3.30		6.50E–14		4.10E–05	
				9.20E–14		5.75E–05	
		1.90–2.19		2.50E–12		1.56E–03	
				1.65E–12		1.03E–03	
		1.01–1.57		1.90E–11		1.19E–02	
				1.00E–11		6.25E–03	
				6.00E–11		3.75E–02	
		7.31–7.57		2.00E–11		1.25E–02	
				2.50E–11		1.56E–02	
	NaI and $NaNO_3$, 0.1 M	0.99–1.14		3.00E–14		1.90E–05	
		1.47–1.55		1.60E–14		1.00E–05	
Fissure coatings, Finnsjön, Sweden	1 M NaI and distilled water			2.20E–15		1.40E–06	
				1.40E–14		8.80E–06	
				1.20E–13		7.50E–05	
				2.20E–13		1.38E–04	
	NaI and $NaNO_3$, 0.1 M	1.67–1.83		1.20E–12		7.50E–04	

Rock type	Water type and ionic strength (M)	ε (%)	D_w(m^2 s^{-1})	D_e(m^2 s^{-1})	α (%)	F_f	Ref
Fissure coatings, Cornish Carnmenellis, the UK	NaI and NaNO$_3$, 0.1 M	6.5 *		6.40E–12		3.20E–03	/Bradbury and Green 1985, 1986/
		0.28		4.50E–13		2.25E–04	
		0.16		1.30E–12		6.50E–04	
		1.9		1.50E–12		7.50E–04	
Fissure coating + granite, Cornish Carnmenellis, the UK	NaI and NaNO$_3$, 0.1 M	0.3		2.10E–13		1.05E–04	
		0.04		3.10E–14		1.55E–05	
		0.33		2.50E–13		1.25E–04	
		0.057		5.60E–14		2.80E–05	
		0.065		5.60E–14		2.80E–05	
		0.08		8.50E–14		4.25E–05	
		0.096		7.70E–14		3.85E–05	
		0.06		2.30E–14		1.15E–05	
		0.088		3.20E–14		1.60E–05	
Fissure coating + granite, AECL, Canada	NaI and NaNO$_3$, 0.1 M			1.32E–12		6.60E–04	
		0.58		8.30E–13		4.15E–04	
				9.90E–13		4.95E–04	
		0.88		7.50E–13		3.75E–04	
		0.29		1.40E–12		7.00E–04	
				4.70E–13		2.35E–04	
		0.93		1.60E–12		8.00E–04	
* The porosities given in /Bradbury and Green 1985/ are the values of the rock capacity α obtained through data fitting.							
Arithmetic mean				**4.16E–12**		**2.54E–03**	
Standard Deviation				**1.10E–11**		**6.89E–03**	

Table A5. Literature data of porosity, bulk diffusivity, effective diffusivity, particle size and formation factor for Cs.

Rock type	Water type and ionic strength (M)	ε (%)	D_w(m^2 s^{-1})	D_e(m^2 s^{-1})	Particle size (mm)	F_f	Ref
			2.00E–09				/Skagius 1986/
Granite, Finnsjön, Sweden	Allard, 0.0085	0.3–0.5		8.50E–13	0.100–0.120	4.25E–04	/Skagius 1986/
		0.3–0.5		2.80E–12	0.200–0.250	1.40E–03	
		0.3–0.5		3.80E–12	0.375–0.43	1.90E–03	
		0.3–0.5		3.70E–11	1.0–1.5	1.85E–02	
		0.3–0.5		1.20E–11	2.0–3.0	6.00E–03	
		0.3–0.5		1.60E–11	4.0–5.0	8.00E–03	
Granite, Stripa, Sweden	Allard, 0.0085	0.3–0.5		2.90E–13	0.100–0.120	1.45E–04	/Skagius 1986/
		0.3–0.5		9.80E–13	4.0–5.0	4.90E–04	
Granite, Finnsjön, Sweden	Allard, 0.0085	0.3–0.5		1.60E–12	Rock piece (disk of φ41X5 mm)	8.00E–04	/Skagius 1986/
		0.3–0.5		1.00E–12		5.00E–04	

Table A6. Literature data of porosity, bulk diffusivity, effective diffusivity, particle size and formation factor for Sr.

Rock type	Water type and ionic strength (M)	ε (%)	D_w(m^2 s^{-1})	D_e(m^2 s^{-1})	Crushed particle size (mm)	F_f	Ref
			1.30E–09				/Skagius 1986/
Granite, Finnsjön, Sweden	Allard, 0.0085	0.3–0.5		4.50E–13	0.100–0.120	2.25E–04	/Skagius 1986/
		0.3–0.5		1.60E–12	0.200–0.250	8.00E–04	
		0.3–0.5		3.50E–12	0.375–0.43	1.75E–03	
		0.3–0.5		4.40E–12	1.0–1.5	2.20E–03	
		0.3–0.5		1.10E–11	2.0–3.0	5.50E–03	
		0.3–0.5		2.40E–11	4.0–5.0	1.20E–02	
Granite, Stripa, Sweden	Allard, 0.0085	0.3–0.5		1.10E–14	0.100–0.120	5.50E–06	/Skagius 1986/
		0.3–0.5		1.00E–12	4.0–5.0	5.00E–04	
Granite, Finnsjön, Sweden	Allard, 0.0085	0.3–0.5		2.20E–12	Rock piece (disk of ϕ41X5 mm)	1.10E–03	/Skagius 1986/
		0.3–0.5		3.10E–12		1.55E–03	

Table B1. Literature data of porosity and formation factor for Swedish granties.

Rock type	Water type and ionic strength (M)	Tracer used	ε (%)	F_f	Ref
Granite, Finnsjön, Sweden	Allard, 0.0085	THO	0.18–0.21	7.50E–5	/Skagius 1986/
	Distilled water			5.42E–5	
	Distilled water	Cr-EDTA	0.21–0.22	1.64E–5	
				1.48E–5	
	Distilled water	Uranin		5.00E–6	
			0.20–0.21	1.20E–5	
	1 M NaI and distilled water	Iodide		5.20E–5	
				4.40E–5	
	NaI and NaNO3, 0.1 M		0.20–0.21	4.40E–5	
			0.21–0.23	2.60E–5	
Young granite, Finnsjön, Sweden	NaI and NaNO3, 0.1 M	iodide	0.27–0.34	2.44E–4	/Skagius 1986/
			0.25–0.30	2.44E–4	
			0.26–0.31	2.12E–4	
			0.21–0.25	1.53E–4	
			0.24–0.28	1.75E–4	
			0.31–0.38	1.75E–4	
			0.24–0.29	2.00E–4	
			0.18–0.21	1.94E–4	
			0.19–0.23	1.56E–4	
			0.54–0.64	1.62E–4	
			0.54–0.60	2.00E–4	
			0.50–0.54	2.75E–4	
			0.40–0.43	1.56E–4	
			0.35–0.37	1.69E–4	
			0.32–0.34	1.41E–4	
			0.28–0.31	1.38E–4	
			0.25–0.28	1.50E–4	
			0.27–0.30	1.55E–4	
			0.26–0.27	1.56E–4	
			0.26–0.27	1.66E–4	
			0.27–0.28	1.58E–4	
			0.28–0.30	1.54E–4	
			0.28–0.29	1.52E–4	
			0.38–0.40	1.47E–4	
			0.33–0.35	1.55E–4	
			0.34–0.37	2.75E–4	
Granite, Studsvik, Sweden	Synthetic, 0.00744	iodide		9.38E–6	/Ittner et al. 1998/
Granite, Gideå, Sweden	Distilled water	Cr-EDTA	0.18–0.19	3.80E–5	/Skagius 1986/
			0.17–0.18	5.50E–5	
	Distilled water	Uranin	0.15–0.17	2.90E–5	
			0.18–0.19	2.90E–5	
	1 M NaI and distilled water, then 1M NaNO3	Iodide	0.16–0.17	6.20E–5	
				8.10E–5	
			0.15–0.16	5.60E–5	
				6.90E–5	

Rock type	Water type and ionic strength (M)	Tracer used	ε (%)	F_f	Ref
Magmatite granite, Svartboberget, Sweden	Distilled water	Cr-EDTA	0.28–0.31	1.17E–4	/Skagius 1986/
			0.22–0.26	6.90E–5	
	Distilled water	Uranin	0.22–0.26	1.53E–4	
			0.30–0.34	1.16E–4	
	NaI and NaNO3, 0.1 M	Iodide	0.20–0.25	4.12E–4	
			0.29–0.36	2.12E–4	
			0.26–0.30	1.62E–4	
Granite, Stripa, Sweden	Stripa groundwater, 0.006	Cr-EDTA	0.3	3.57E–5	/Birgersson 1988/
			0.3	1.43E–4	
			0.3	7.14E–5	
			0.3	7.14E–5	
			0.3	7.14E–5	
			0.3	3.57E–5	
			0.3	3.57E–5	
			0.3	3.57E–5	
			0.3	1.43E–4	
			0.3	3.57E–5	
Granite, Stripa, Sweden	Stripa groundwater, 0.006	Uranin	0.3	1.33E–3	/Birgersson 1988/
			0.3	2.33E–4	
			0.3	2.33E–4	
			0.3	4.00E–5	
			0.3	4.00E–5	
			0.3	4.00E–5	
			0.3	4.00E–5	
			0.3	4.00E–5	
			0.3	4.00E–5	
			0.33	1.47E–4	
			0.33	4.89E–5	
			0.3	2.00E–5	
			0.3	2.22E–5	
			0.3	4.44E–5	
Granite, Stripa, Sweden	1 M NaI and distilled water	Iodide		1.00E–4	/Skagius 1986/
				8.10E–5	
	Stripa groundwater, 0.006	Iodide	0.3	9.38E–4	/Birgersson 1988/
			0.3	1.88E–4	
			0.3	1.88E–4	
			0.3	1.31E–4	
			0.3	9.38E–4	
			0.3	3.75E–5	
			0.3	3.75E–5	
			0.3	3.75E–5	
			0.3	3.75E–5	
			0.33	8.63E–5	
			0.33	3.19E–4	
			0.3	3.19E–4	
			0.3	3.19E–5	
			0.3	9.38E–5	
	Synthetic, 0.00309	Iodide		2.00E–5	/Ittner et al. 1998/

Rock type	Water type and ionic strength (M)	Tracer used	ε (%)	F_f	Ref
Granite, Sweden		Uranin	2.3–3.4	8.96E–4	/Eriksen 1985/
				6.53E–4	
				6.53E–4	
Granite, Laxemar, Sweden	NaCl, 1.0 M	Iodide		2.40E–4	/Löfgren 2004/
				1.20E–4	
				5.30E–4	

Table B2. Literature data of porosity and formation factor for other Swedish crystalline rocks.

Rock type	Water type and ionic strength (M)	Tracer used	ε (%)	F_f	Ref
Biotite gneiss, Svartboberget, Sweden	Distilled water	Cr-EDTA	0.24–0.28	6.00E–5	/Skagius 1986/
			0.30–0.34	7.10E–5	
Biotite gneiss, Fjällveden, Sweden	Distilled water		0.27–0.28	1.90E–5	
			0.26–0.28	1.40E–5	
Magmatite, Studsvik, Sweden	Distilled water		0.20–0.21	2.20E–5	/Skagius 1986/
Gneiss, Gideå, Sweden	Distilled water	Uranin	0.11–0.12	7.10E–6	/Skagius 1986/
Gneiss, Svartboberget, Sweden	Distilled water		0.08–0.09	2.07E–5	
			0.08–0.10	2.13E–5	
Biotite gneiss, Svartboberget, Sweden	Distilled water		0.30–0.35	1.73E–4	
			0.23–0.28	1.33E–4	
Biotite gneiss, Fjällveden, Sweden	Distilled water		0.23–0.24	8.00E–6	
			0.23–0.24	9.00E–6	
Magmatite, Studsvik, Sweden	Distilled water	Uranin	0.21–0.22	5.10E–5	/Skagius 1986/
Gneiss, Karlshamn, Sweden	1 M NaI and distilled water	Iodide		8.10E–5	/Skagius 1986/
	NaI and NaNO3, 0.1 M			3.00E–5	
Gneiss, Gideå, Sweden	NaI and NaNO3, 0.1 M		0.06	1.12E–5	
Gneiss, Svartboberget, Sweden	NaI and NaNO3, 0.1 M		0.07	1.19E–5	
			0.07–0.09	2.07E–5	
Biotite gneiss, Svartboberget, Sweden	NaI and NaNO3, 0.1 M		0.29–0.34	9.40E–5	
			0.28–0.32	2.25E–4	
Gneiss, Fjällveden, Sweden	NaI and NaNO3, 0.1 M		0.25–0.26	4.50E–5	
			0.27–0.29	4.60E–5	
Magmatite, Studsvik, Sweden	1 M NaI and distilled water	Iodide		8.10E–5	/Skagius 1986/
	NaI and NaNO3, 0.1 M			3.00E–5	
Diorite, Äspö, Sweden	1.00 M NaCl	Uranin		1.2E–4	/Ohlsson 2000/
				5.8E–5	
				5.1E–5	
				6.6E–5	
				2.4E–5	
				8.1E–5	
				5.5E–5	
				4.0E–5	
				4.0E–5	
				4.4E–5	

Table B3. Literature data of porosity and formation factor for international crystalline rocks.

Rock type	Water type and ionic strength (M)	Tracer used	ε (%)	F_f	Ref
Granite, Whiteshell, Canada	NaI and NaNO3, 0.1 M	Iodide	2.20E–3	1.32E–3	/Vilks et al. 2003/
			2.20E–3	1.12E–3	
Granite, Ibaraki Prefecture, Japan	0.1 M NaHCO3	UO2(CO3)3 4–	2.26E–3	2.17E–4	/Yamaguchi and Nakayama 1998/
			3.23E–3	1.99E–4	
			3.70E–1	1.78E–4	
		Pu(OH)2(CO3)2 2–	1.25E–3	2.89E–4	
			1.25E–3	2.23E–4	
			8.50E–4	1.23E–4	
Granite, Kivetty, Finland	Dilluted Allard, 4.5E–4	THO	0.05–0.16	1.63E–4	/Kaukonen et al. 1997/
				1.50E–4	
				7.08E–5	
				7.08E–5	
				9.17E–5	
				5.83E–5	
				7.08E–5	
				7.50E–5	
				1.13E–4	
				1.00E–4	
				1.00E–4	
				1.63E–4	
			0.1	2.46E–4	
				1.96E–4	
				8.75E–5	
				8.33E–5	
				1.25E–4	
				8.33E–5	
				9.58E–5	
				9.17E–5	
				1.33E–4	
				1.25E–4	
				1.46E–4	
				1.63E–4	
Granite, Ossian, the UK	NaNO3, 0.1 M	iodide	1.20 *	7.00E–4	/Bradbury and Green 1985/
			1.60	7.00E–4	
			2.20	7.00E–4	
			1.20	8.00E–4	
			0.74	5.00E–4	
			0.31	4.00E–4	
			0.65	3.00E–4	
			0.57	3.00E–4	
Granite, Scottish lowland, the UK	NaNO3, 0.1 M	iodide	0.22	2.00E–4	
			0.27	3.00E–4	

Rock type	Water type and ionic strength (M)	Tracer used	ε (%)	F_f	Ref
Granite, Skene complex, the UK	NaNO3, 0.1 M	iodide	0.12	1.00E–4	/Bradbury and Green 1985/
			0.38	4.00E–4	
			0.21	3.00E–4	
			0.36	4.00E–4	
			0.27	3.00E–4	
			0.29	3.00E–4	
			0.37	4.00E–4	
			0.46	3.00E–4	
Granite, Cornish Carnmenellis, the UK	NaNO3, 0.1 M	iodide	0.06	1.20E–5	
			0.088	1.60E–5	
Pegmatite, Olkiluoto, Finland	Olkilouto, 0.26–0.31	THO	0.13–0.39	4.53E–4	/Kaukonen et al. 1997/
				4.89E–4	
				5.79E–4	
				3.42E–4	
				5.16E–4	
				7.37E–4	
				6.84E–4	
Pegmatite, Olkiluoto, Finland	Olkilouto, 0.26–0.31	THO	0.1	4.32E–4	/Kaukonen et al. 1997/
				6.32E–4	
				8.42E–4	
				2.63E–4	
				5.79E–4	
				5.79E–4	
				6.32E–4	
Mica gneiss, Olkiluoto, Finland	Olkilouto, 0.26–0.31	THO	0.05–0.06	7.37E–5	/Kaukonen et al. 1997/
				5.26E–5	
				7.37E–5	
				4.21E–5	
				6.84E–5	
				7.89E–5	
				2.79E–4	
				5.26E–4	
			0.05	7.37E–5	
				5.26E–5	
				8.95E–5	
				5.26E–5	
				8.42E–5	
				1.00E–4	
				3.95E–4	
				6.84E–4	
Gneiss, Hästholmen. Finland	NaCl, 0.0044	THO		4.00E–5	/Lehikoinien et al. 1992/
	NaCl, 0.7			3.54E–5	
	NaCl, 1.0			3.17E–5	
Plutonic, Mäntsäälä, Finland	Allard, 0.0085	THO	0.2	4.95E–5	/Kaukonen et al. 1993/
				2.84E–4	
Plutonic, Syyry, Finland	Syyry	THO	0.26–0.43	3.05E–5	
				3.00E–5	
				4.79E–5	

Rock type	Water type and ionic strength (M)	Tracer used	ε (%)	F_f	Ref
Vulcanite	Allard, 0.0085	THO	0.27	3.34E–4	/Kaukonen et al. 1993/
				1.41E–4	
				1.93E–4	
Granodiorite, Whiteshell, Canada	NaI and NaNO3, 0.1 M	Iodide	2.60E–3	1.22E–3	/Vilks et al. 2003/
			2.80E–3	1.07E–3	
Basalt, Gunma, Japan	1.8 M NaCl	Chloride		2.05E–4	/Sato et al. 1996/
Grannodiorite, Gunma, Japan	1.8 M NaCl	Chloride		3.70E–4	/Sato et al. 1997/
Granodiorite, Grimsel, Switzerland			6.60E–3		/Möri et al. 2003/
			7.70E–3		
			6.90E–3		
			5.30E–3		
			6.80E–3		
			8.00E–3		

Table B4. Literature data of porosity and formation factor for fissure coatings.

Rock type	Water type and ionic strength (M)	Tracer used	ε (%)	F_f	Ref
Granite + Fissure coatings, Finnsjön, Sweden	Distilled water	Cr-EDTA	1.43–1.58	2.40E–5	/Skagius 1986/
			1.03–1.19	1.50E–5	
Granite + Fissure coatings, Finnsjön, Sweden	Distilled water	Uranin	3.00–3.31	2.90E–5	/Skagius 1986/
			1.28–1.35	1.38E–4	
			0.78–1.00	1.11E–4	
				3.56E–2	
				4.44E–2	
			7.12–7.49	8.89E–2	
			7.12–7.49	2.00E–2	
			1.17–1.25	1.60E–5	
			1.67–1.79	3.33E–3	
Granite + Fissure coatings, Finnsjön, Sweden	1 M NaI and distilled water	Iodide		7.50E–5	/Skagius 1986/
				3.75E–5	
			2.57–3.30	4.10E–5	
				5.75E–5	
			1.90–2.19	1.56E–3	
				1.03E–3	
			1.01–1.57	1.19E–2	
				6.25E–3	
				3.75E–2	
			7.31–7.57	1.25E–2	
				1.56E–2	
	NaI and NaNO3, 0.1 M		0.99–1.14	1.90E–5	
			1.47–1.55	1.00E–5	

Rock type	Water type and ionic strength (M)	Tracer used	ε (%)	F_f	Ref
Fissure coating + granite, Cornish Carnmenellis, the UK	NaI and NaNO3, 0.1 M	Iodide	0.3	1.05E–4	/Bradbury and Green 1985, 1986/
			0.04	1.55E–5	
			0.33	1.25E–4	
			0.057	2.80E–5	
			0.065	2.80E–5	
			0.08	4.25E–5	
			0.096	3.85E–5	
			0.06	1.15E–5	
			0.088	1.60E–5	
Fissure coating + granite, Cornish Carnmenellis, the UK	NaI and NaNO3, 0.1 M	Iodide	0.58	4.15E–4	/Bradbury and Green 1985, 1986/
				4.95E–4	
			0.88	3.75E–4	
			0.29	7.00E–4	
				2.35E–4	
			0.93	8.00E–4	
* The porosities given in /Bradbury and Green 1985/ are the values of the rock capacity a obtained through data fitting.					
Fissure coatings, Finnsjön, Sweden	Distilled water	Cr-EDTA	3.51–3.61	1.14E–3	/Skagius 1986/
Fissure coatings, Finnsjön, Sweden	Distilled water	Uranin		1.33E–4	/Skagius 1986/
				2.00E–4	
			2.67–2.79	4.89E–4	
Fissure coatings, Finnsjön, Sweden	1 M NaI and distilled water	Iodide		1.40E–6	/Skagius 1986/
				8.80E–6	
				7.50E–5	
				1.38E–4	
	NaI and NaNO3, 0.1 M		1.67–1.83	7.50E–4	
Fissure coatings, Cornish Carnmenellis, the UK	NaI and NaNO3, 0.1 M	Iodide	6.5 *	3.20E–3	/Bradbury and Green 1985, 1986/
			0.28	2.25E–4	
			0.16	6.50E–4	
			1.9	7.50E–4	

Table C. Porosities of crystalline rocks measured by different methods.

Rock type	e (%) (water-saturation)	e (%) (leaching after diffusion)	e (%) (conc. profile intercept, a)	Tracers used in the leaching method	Ref.
Granite, Finnsjön, Sweden		0.14	0.3	Iodide	/Skagius 1986/
		0.11	0.2	Iodide	
		0.06	0.02	Uranin	
	0.20–0.21	0.12	0.27	Iodide	
	0.21–0.23	0.07	0.18	Iodide	
	0.20–0.21	0.09	0.02	uranin	
	0.21–0.22	0.1	0.04	Cr-EDTA	
Granite, Stripa, Sweden		0.3	1.05	Iodide	
		0.27	0.21	Iodide	

Rock type	e (%) (water-saturation)	e (%) (leaching after diffusion)	e (%) (conc. profile intercept, a)	Tracers used in the leaching method	Ref.
Granite, Gideå, Sweden	0.16–0.17	0.12	0.14	Iodide	/Skagius 1986/
	0.15–0.16	0.11	0.14	Iodide	
	0.15–0.17	0.16	0.11	Uranin	
	0.18–0.19	0.16	0.12	Uranin	
	0.18–0.19	0.18	0.04	Cr-EDTA	
	0.17–0.18	0.19	0.08	Cr-EDTA	
Young granite, Finnsjön, Sweden	0.27–0.34	0.38	0.22	Iodide	
	0.25–0.30	0.44	0.09	Iodide	
	0.26–0.31	0.29	0.2	Iodide	
	0.21–0.25	0.35	0.15	Iodide	
	0.24–0.28	0.28	0.11	Iodide	
	0.31–0.38	0.27	0.17	Iodide	
	0.24–0.29	0.26	0.1	Iodide	
	0.18–0.21	0.23	0.24	Iodide	
	0.19–0.23	0.27	0.13	Iodide	
	0.54–0.64	0.43	0.37	Iodide	
	0.54–0.60	0.5	0.32	Iodide	
Young granite, Finnsjön, Sweden	0.50–0.54	0.39	0.26	Iodide	/Skagius 1986/
	0.40–0.43	0.43	0.38	Iodide	
	0.35–0.37	0.28	0.34	Iodide	
	0.32–0.34	0.24	0.34	Iodide	
	0.28–0.31	0.19	0.36	Iodide	
	0.25–0.28	0.3	0.28	Iodide	
	0.27–0.30	0.21	0.41	Iodide	
	0.26–0.27	0.19	0.37	Iodide	
	0.26–0.27	0.27	0.47	Iodide	
	0.27–0.28	0.26	0.41	Iodide	
	0.28–0.30	0.21	0.37	Iodide	
	0.28–0.29	0.23	0.32	Iodide	
	0.38–0.40	0.24	0.34	Iodide	
	0.33–0.35	0.25	0.37	Iodide	
	0.34–0.37	0.26	0.65	Iodide	
Magmatite granite, Svartboberget, Sweden	0.20–0.25	0.25	1.14	Iodide	
	0.22–0.26	0.33	0.3	Uranin	
	0.28–0.31	0.36	0.29	Cr-EDTA	
	0.29–0.36	0.29	0.58	Iodide	
	0.30–0.34	0.38	0.94	Uranin	
	0.22–0.26	0.34	0.13	Cr-EDTA	
	0.26–0.30	0.27	0.28	Iodide	
Gneiss, Karlshamn, Sweden		0.32	0.74	Iodide	
		0.26	0.27	Iodide	
Gneiss, Gideå, Sweden	0.06–0.08	0.06		Iodide	
	0,06	0.07	0.05	Iodide	
	0.07–0.09	0.08		Uranin	
	0.11–0.12	0.11	0.11	Uranin	
	0.07–0.08	0.05		Cr-EDTA	
Gneiss, Svartboberget, Sweden	0,07	0.07	0.07	Iodide	
	0.07–0.09	0.07	0.12	Iodide	
	0.08–0.09	0.12	0.07	Uranin	
	0.08–0.10	0.11	0.07	Uranin	
	0.06–0.07	0.11		Cr-EDTA	

Rock type	e (%) (water-saturation)	e (%) (leaching after diffusion)	e (%) (conc. profile intercept, a)	Tracers used in the leaching method	Ref.
Biotite gneiss, Svartboberget, Sweden	0.24–0.28	0.32	0.13	Cr-EDTA	/Skagius 1986/
	0.29–0.34	0.21	0.26	Iodide	
	0.30–0.35	0.38	0.52	Uranin	
	0.30–0.34	0.29	0.06	Cr-EDTA	
	0.28–0.32	0.28	0.92	Iodide	
	0.23–0.28	0.32	0.62	Uranin	
Biotite gneiss, Fjällveden, Sweden	0.25–0.26	0.1	0.36	Iodide	
	0.27–0.29	0.1	0.15	Iodide	
	0.23–0.24	0.17	0.02	Uranin	
	0.23–0.24	0.19	0.04	Uranin	
	0.27–0.28	0.33	0.14	Cr-EDTA	
	0.26–0.28	0.34	0.08	Cr-EDTA	
Magmatite, Studsvik, Sweden	0.20–0.21	0.17	0.3	Iodide	
	0.21–0.22	0.18	0.22	Uranin	
	0.20–0.21	0.16	0.03	Cr-EDTA	
Granite + Fissure coatings, Finnsjön, Sweden		0.13	1.23	Iodide	
			1.32	Iodide	
	2.57–3.30	3.62	0.5	Iodide	
	3.00–3.31	4.55	0.1	Uranin	
	1.90–2.19	1.63	27.5	Iodide	
	1.28–1.35	1.57	3.5	Uranin	
	1.10–1.57	1.11	1.05	Iodide	
	0.78–1.00	0.78	2.5	Uranin	
			24	Iodide	
			24	Uranin	
	7.31–7.57	4.2	50	Iodide	
	7.12–7.49	6.75	8.27	Uranin	
	0.99–1.14	0.89	0.4	Iodide	
	1.43–1.58	2.46	0.8	Cr-EDTA	
	1.17–1.25	0.88	0.27	Uranin	
	1.47–1.55	0.87	0.5	Iodide	
	1.67–1.79	1.55	2.8	Uranin	
	1.03–1.09	3.98	0.36	Cr-EDTA	
Fissure coatings		0.41	0.21	Iodide	/Skagius 1986/
		0.33	0.13	Iodide	
		1.46	1.43	Iodide	
		3	1.1	Uranin	
	1.67–1.83	1.52	1.55	Iodide	
	2.67–2.79	3.37	1.3	Uranin	
	3.51–3.61	4.27	2.82	Cr-EDTA	

Appendix II

Guide how to read the distribution and normal-score plots in this report

In this report there are a number of log-normal distributions, for example Figure A2-1.

Here the data points, the grey diamonds, are shown in a normal-score plot. The normal score axis is the right y-axis. The data points are ordered from lower formation factors to higher formation factors. If the data points in the normal-score plot could be fitted to a straight line, the data points are distributed according to the proposed distribution /Johnson 1994/.

From the normal score values the standard deviation could be read (see below). From the mean value and standard deviation the distribution curve is drawn (the solid black line). The scale of the normalised frequency axis (left axis) is adjusted so that the integral under the distribution curve is always 1.

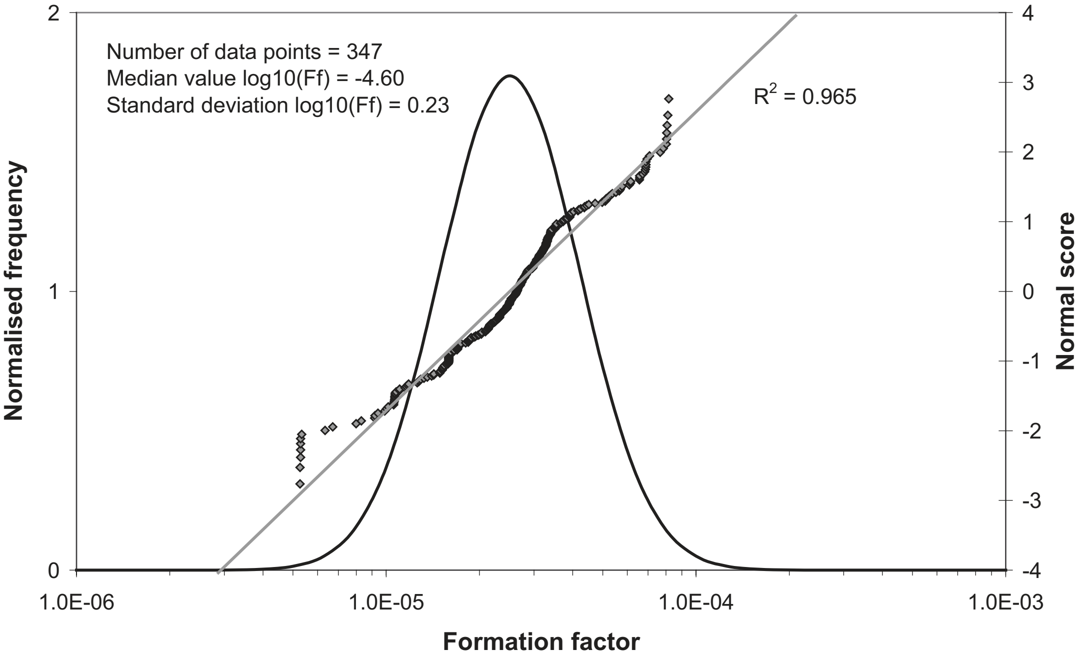
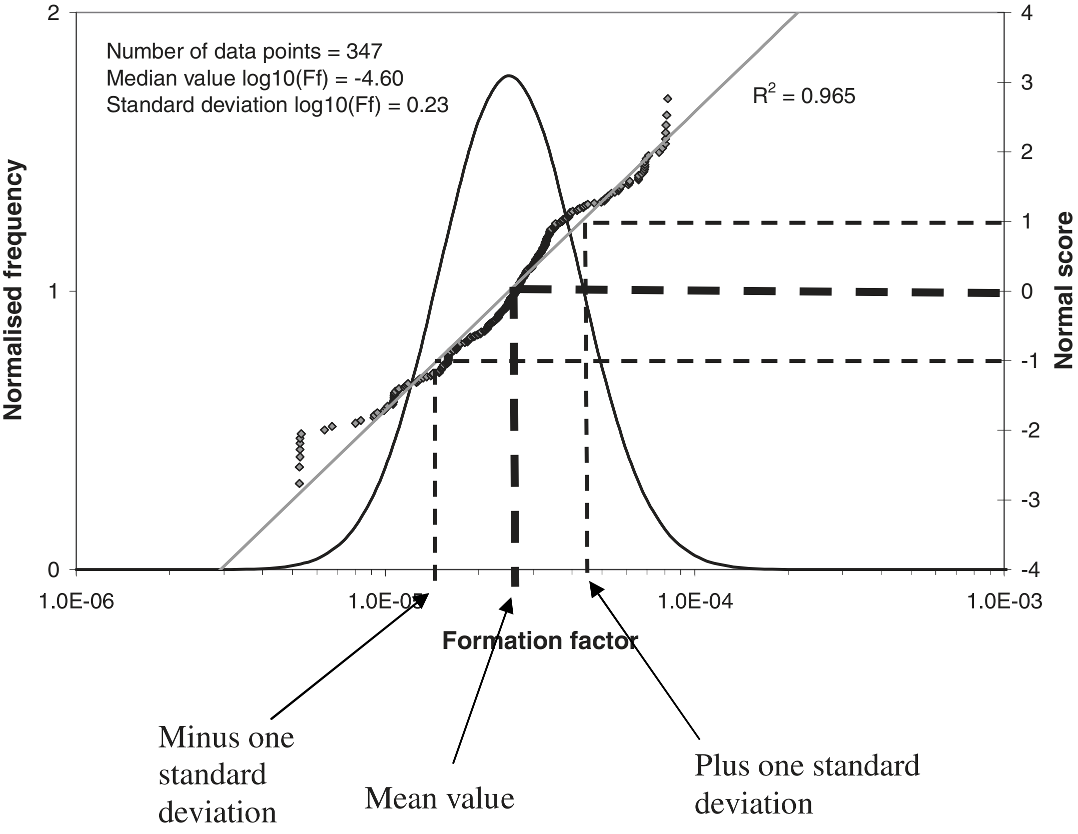


Figure A2-1. *Distribution of in-situ formation factors in Laxemar.*



Minus one standard deviation

Mean value

Plus one standard deviation