

**R-07-39**

**Tracer tests  
– possibilities and limitations  
Experience from SKB fieldwork: 1977–2007**

Martin Löfgren, James Crawford, Mark Elert  
Kemakta Konsult AB

September 2007

**Svensk Kärnbränslehantering AB**

Swedish Nuclear Fuel  
and Waste Management Co  
Box 5864  
SE-102 40 Stockholm Sweden  
Tel 08-459 84 00  
+46 8 459 84 00  
Fax 08-661 57 19  
+46 8 661 57 19



**Tracer tests  
– possibilities and limitations  
Experience from SKB fieldwork: 1977–2007**

Martin Löfgren, James Crawford, Mark Elert  
Kemakta Konsult AB

September 2007

*Keywords:* Tracer tests, In situ, Transport properties, Safety Assessment Modelling.

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

A pdf version of this document can be downloaded from [www.skb.se](http://www.skb.se).

# Abstract

Tracer tests have played, and still play, a central role in investigations relating to the understanding of radionuclide retention processes in the field. At present there is a debate within the scientific community concerning how, and to what extent, tracer tests can be used to evaluate large-scale and long-term transport and retardation of radionuclides and other solutes of interest for Safety Assessment of repositories for spent nuclear fuel.

In this report the SKB fieldwork on tracer tests performed at Swedish sites from 1977 to 2007 is described and discussed. Furthermore, the knowledge and process understanding evolved during the decades of radionuclide transport experiments and modelling within the SKB programme is summarised.

One of the main objectives of this report is to discuss what data and knowledge can be extracted from different in situ tests in a robust fashion. Given the level of complexity associated with transport processes that may occur over the timescale of a tracer test, the utility of tracer tests is considered in the context of evidence-based interpretations of data which we characterise in the form of a sequence of questions of increasing complexity. The complexity of this sequence ranges from whether connection can be confirmed between injection and withdrawal points to whether quantitative data can be extrapolated from a tracer test to be subsequently used in Safety Assessment.

The main findings of this report are that:

- Field scale tracer tests can confirm flow connectivity.
- Field scale tracer tests confirm the existence of retention.
- Field scale tracer tests alone can only broadly substantiate our process understanding. However, if performing extensive Site Characterisation and integrating the tracer test results with the full range of geoscientific information available, much support can be given to our process understanding.
- Field scale tracer tests can deliver the product of the material property group *MPG* and the F-factor, valid only for the tracer test configuration, but not separate them without introducing additional assumptions.
- Field scale tracer tests (in flowing fractures) alone cannot deliver individual transport parameters, such as effective diffusivity, flow-wetted surface, etc in the absence of additional supporting information.
- Lumped or individual transport parameters obtained from inverse modelling of tracer test data are not directly transferable to Safety Assessment flow conditions or timescales.

In addition to the above findings, another objective of this report has been to describe and highlight a number of elaborate in situ tracer tests that SKB has performed and reported. Many of these experiments have contributed immeasurably to current process understanding although may not be widely known to the general reader.

# Contents

<b>1</b>	<b>Introduction</b>	7
1.1	Background	7
1.2	Aims and scope	7
1.3	Outline of report	8
1.4	Transport and retention paradigm	9
<b>2</b>	<b>Conceptual understanding of transport and retention processes</b>	11
2.1	Introduction	11
2.2	Conceptual understanding of the bedrock	11
2.2.1	The rock mass	12
2.2.2	Deformation zones and fractures	18
2.3	Conceptual understanding of the flowing groundwater system	20
2.3.1	Hydraulic gradients	20
2.3.2	Hydraulic conductivity and fluid properties	21
2.4	Conceptual understanding of radionuclide transport	24
2.4.1	Hydrodynamic dispersion	24
2.4.2	Matrix diffusion	26
2.4.3	Partitioning	30
2.4.4	Flow-wetted surface	32
<b>3</b>	<b>SKB field tracer tests: 1977–2007</b>	35
3.1	Distinguishing characteristics of tracer tests	35
3.1.1	Environment	36
3.1.2	Flow situation	36
3.1.3	Monitoring and sampling	38
3.1.4	Tracers	38
3.2	Tracer tests at the Studsvik research centre area	38
3.2.1	Cross-hole tracer test 1 in Studsvik	38
3.2.2	Cross-hole tracer test 2 in Studsvik	40
3.3	Tracer tests at Finnsjön test area	41
3.3.1	Cross-hole tracer test in Finnsjön	41
3.3.2	The Fracture Zone Project, Phase 3	43
3.4	Tracer tests in Stripa mine	46
3.4.1	Small-scale tracer tests at the SGAB tracer test drift	46
3.4.2	Monitoring of saline tracer transport with borehole radar	47
3.4.3	In situ diffusion experiment	48
3.4.4	Migration in a single fracture, the 2D experiment	50
3.4.5	3D migration experiment	53
3.4.6	Channelling experiments	55
3.4.7	Tracer Migration Experiment in the Validation drift – TME	56
3.5	Tracer tests at Äspö Hard Rock Laboratory	58
3.5.1	Long Term Pumping and Tracer Test – LTP2	58
3.5.2	Tracer Retention Understanding Experiments – TRUE	60
3.5.3	TRUE-1	60
3.5.4	TRUE Block Scale	63
3.5.5	Äspö Task Force 6 – Understanding TRUE results	67
3.5.6	TRUE-1 Continuation	70
3.5.7	TRUE Block Scale Continuation	72
3.5.8	Long Term Sorption Diffusion Experiment – LTDE-SD	75
3.6	Tracer tests within the site investigations	77
3.6.1	Single-Well Injection-Withdrawal – SWIW	77
3.6.2	Formation factor logging in situ	78

3.6.3	Large-scale tracer test in Forsmark at drill site 1	81
3.6.4	Large-scale tracer test in Forsmark at drill site 2	82
3.6.5	Large-scale tracer test in Forsmark 3	82
3.6.6	Large-scale tracer test in Laxemar	82
<b>4</b>	<b>The constraining power of tracer tests</b>	<b>83</b>
4.1	Introduction	83
4.2	Interpretation of data at different levels of complexity	84
4.3	Confirmation of flow connectivity	85
4.4	Qualitative confirmation of retention processes	86
4.5	Confirmation of process understanding	86
4.5.1	Transport of radionuclides along discrete flowpaths	87
4.5.2	Matrix diffusion	87
4.5.3	Partitioning of solutes	88
4.6	Abstraction of lumped transport parameters	91
4.6.1	Aspects of transport processes involving matrix diffusion and sorption	91
4.7	Abstraction of individual transport parameters	95
4.8	Extrapolation of numerical data to Safety Assessment	98
4.9	Possibilities and limitations of tracer tests	101
<b>5</b>	<b>Conclusions</b>	<b>105</b>
	<b>Notation list</b>	<b>109</b>
	<b>References</b>	<b>111</b>

# 1 Introduction

## 1.1 Background

Transport and retention of solutes in geological media has been studied around the world for more than half a century. The Swedish Nuclear Fuel and Waste Management Company (SKB) has studied transport in geological media with focus upon understanding of radionuclide retention for several decades. Investigations have been carried out both in the laboratory and in situ at a number of sites in Sweden. Furthermore, SKB has performed/participated in a number of international co-operative studies where radionuclide retention has been studied in detail.

SKB is currently performing site investigations and Site Characterisations at Forsmark and Oskarshamn, resulting in much new knowledge. Much of this knowledge has been internalised in the Safety Assessment SR-Can and will be internalised in the Safety Assessment SR-Site. Recently SKB has participated in three major programmes with the focus upon understanding solute transport; the TRUE series of projects, the RETROCK project, and the Äspö Modelling Task Force – Task 6, delivering much insight into the topic of radionuclide retention. Earlier investigations included studies of field scale retention in tracer tests carried out in the Stripa mine (the OECD/NEA Stripa Project), at the Äspö Hard Rock Laboratory, and at other locations including Finnsjön and Studsvik.

Tracer tests have played, and still play, a central role in investigations relating to the understanding of retention processes in the field. At present there is a debate within the scientific community concerning how, and to what extent, tracer tests can be used to evaluate large-scale and long-term transport and retardation of radionuclides and other solutes in geological formations. Therefore, understanding of the possibilities and limitations of tracer tests is of great importance for the continued site investigation strategy of SKB.

## 1.2 Aims and scope

This report aims to discuss the possibilities and limitations of tracer tests, with focus upon in situ tracer tests performed in Sweden by SKB. The discussion is held in the context of what information of interest for Site Characterisation (SC) and Safety Assessment (SA) can be extracted from these tests. To clearly underline the scientific basis for our detailed discussions in later chapters, the conceptual understanding of transport and retention processes in crystalline rock, as we see it, is given early in this report. However, we have restrained ourselves from describing the processes in mathematical equations as is done in many other publications, among which we recommend the RETROCK project /SKB 2004/.

Much of the discussion concerning the possibilities and limitations of field experiments is centred on the re-examination of various in situ tracer tests performed by SKB from the late 1970's to the present time. In this report, the designs and outcomes of the most important field experiments are briefly described. Supporting experiments from laboratory studies and field experiments carried out in countries other than Sweden may be briefly acknowledged in various sections in the report, although it should be noted that a detailed account of these investigations and their conclusions are beyond the scope of this report and generally has not been included. For a compilation of relevant field scale tracer tests carried out in other countries than Sweden, /Andersson 1995/ is recommended.

In this present report it is shown that there are some inherent difficulties in extracting appropriate data from tracer tests for use in models applicable over the long time periods and large length scales of relevance for demonstrating repository safety. It is also shown that tracer tests do deliver some confirmatory information that is of strong qualitative significance for process

understanding and confidence building in SA models of radionuclide migration. In general our conclusions are in line with the outcome of the Äspö Modelling Task Force 6 /e.g. Hodgkinson 2007, Hodgkinson and Black 2005/ and the RETROCK project /SKB 2004/

This report, and conclusions drawn, will serve as input for the continued site investigation strategy of SKB for obtaining transport and retention parameters of fractured rock.

It has been identified during the site investigations and in the Safety Assessment SR-Can /SKB 2006d/ that the transport properties of the rock may be of high importance for repository safety not only in terms of radionuclide migration, but also for the prediction of groundwater chemistry evolution. Even so, it should be noted that this report concerns radionuclide migration only and the implications of tracer test data for the modelling of groundwater chemistry are not covered.

### **1.3 Outline of report**

In Chapter 2 of this report the conceptual understanding of transport and retention processes in fractured crystalline rock is described. The rock mass, the hydraulic system, and transport of solutes are discussed. A sound conceptual understanding is vital when extrapolating results from tracer tests carried out over short distances and experimental times to SA spatial scales and timescales.

In Chapter 3, the experimental setup and performance of in situ tracer tests carried out from the late 1970's to the present time are summarised and briefly described. This chapter does not aim to account in detail for the results and interpretations of these tests, but merely to summarise their major achievements. References to more detailed descriptions of these tracer tests are given for further reading. The short descriptions given serve as a platform for later discussions in Chapter 4 concerning what information can be extracted from tracer tests.

Concerning the TRUE series of projects, which are considered to be of extra importance for the overall SKB strategy for obtaining radionuclide retention information, a somewhat more extensive summation is given. In addition, the major achievements of the Äspö Modelling Task Force, Task 6 are summarised. It has also been considered that in this report, some extra attention should be given to the tracer tests carried out in the Stripa mine.

An important outcome of Chapter 3 is that we demonstrate that SKB has performed a number of relevant large-scale tracer tests which have been widely reported in internationally peer-reviewed publications.

In Chapter 4, the question of what data and knowledge can actually be robustly extracted from different in situ tests is discussed. Given the level of complexity associated with transport processes that may occur over the timescale of a tracer test, the utility of such tests is considered in the context of evidence-based interpretations of data which we characterise in the form of a sequence of questions of increasing complexity.

The complexity of these questions ranges from whether transport connectivity can be confirmed between injection and withdrawal points, to whether quantitative data can be extrapolated from a tracer test for use within Safety Assessment. When discussing this sequence of questions, we do so in the context of in situ tracer tests that have been performed by SKB. Given the immense body of literature that exists concerning these tests, however, it is impractical to discuss each question in the light of every single tracer test described in Chapter 3.

At the end of Chapter 4, a table is given concerning possibilities and limitations of tracer tests that may serve as a guide when planning future tracer tests.

In Chapter 5, the most important conclusions of this report are highlighted.

## **1.4 Transport and retention paradigm**

The basis for the discussions in this report is the paradigm that long-range transport occurs by advective and channelled flow in a discrete fracture network. Furthermore, the transported radionuclides are assumed to be influenced by matrix diffusion and partitioning between aqueous and solid phases, both at the fracture surface and within the rock matrix. Matrix diffusion and partitioning of solutes are thought to be the main retardation mechanisms limiting the transport of radionuclides in fractured rock. We acknowledge that our stated paradigm affects the content and scope of this report.



## **2 Conceptual understanding of transport and retention processes**

### **2.1 Introduction**

A fundamental problem when assessing radionuclide transport in geological formations from repository depth to the biosphere concerns the timescale. The residence time in the natural barrier is, for many important radionuclides, thought to be on the order of thousands or even millions of years. Long-term tracer tests, where solutes are allowed to migrate in a geological medium, are typically carried out for a few years to a decade at the most. Short-term tracer tests can span over hours to some weeks or months.

Owing to the relatively short timescales over which in situ experiments are usually carried out, in combination with the large retention capacity of the geological medium, experiments are often performed by necessity over limited length scales. If using moderately to strongly sorbing radionuclides, tracer tests are invariably carried out over short length scales if tracer recovery is expected.

As tracer tests spanning from repository depth to the biosphere are not feasible using representative sorbing radionuclides, the information gained from short-term and small-scale tracer tests needs to be extrapolated to be of use in Safety Assessments. As is always the case when performing extrapolations, unless the extrapolation is based on a sound conceptual understanding, any way of extrapolating the data outside of its range of calibration is as unsound as any other. For this reason, regardless of the quality of the data set that is obtained from the tracer test, without putting it first into the context of the (preferably evolving) conceptual understanding of the system which accounts for relevant mechanisms and processes, the data are useless for predicting radionuclide retention during repository evolution.

The conceptual understanding needed for assessing solute transport in a geological formation can be roughly divided into the following three subdivisions:

- Conceptual understanding of the bedrock, including its rock mass and fracture network.
- Conceptual understanding of the groundwater movement within the rock mass and fracture network.
- Conceptual understanding of radionuclide transport within the groundwater including chemical reactions and diffusive interaction with the rock mass.

Although tracer tests often only focus on the third subdivision, without the additional understanding of the other two subdivisions there is a substantial risk of making incorrect assumptions and extrapolations that lack physical basis. The TRUE series of projects is an excellent example of tracer tests where much weight was also given to the first two subdivisions. By performing multidisciplinary investigation in the Site Characterisation prior to the tracer tests, descriptive models on both macro and micro scales could be developed for the site. As a result of integrating tracer test results with the full range of geoscientific information available, an increased understanding of solute transport and retention was gained.

### **2.2 Conceptual understanding of the bedrock**

There are a number of general features of crystalline rock that are of interest when discussing radionuclide retention. The magnitudes and overall importance of different properties of the rock mass, such as the fracture frequency, may differ greatly from site to site. Notwithstanding this, however, the mechanisms involved in radionuclide retention are believed to be similar for

many different geological materials. The information concerning the bedrock of the Forsmark and Oskarshamn site investigation areas are summarised in a number of site descriptive model reports /SKB 2005ab, 2006ab/. Other sites investigated by SKB are described in a wealth of SKB reports (see e.g. /Milnes 2002/ for an overview). In the Forsmark and Oskarshamn site descriptions, the bedrock is divided into three main components; 1) the rock mass itself, 2) background fractures and minor deformation zones that are described stochastically, and 3) “deterministically” identified and defined deformation zones. From a viewpoint of radionuclide retention, there are reasons to divide the bedrock into the corresponding domains, as will be discussed in subsequent sections.

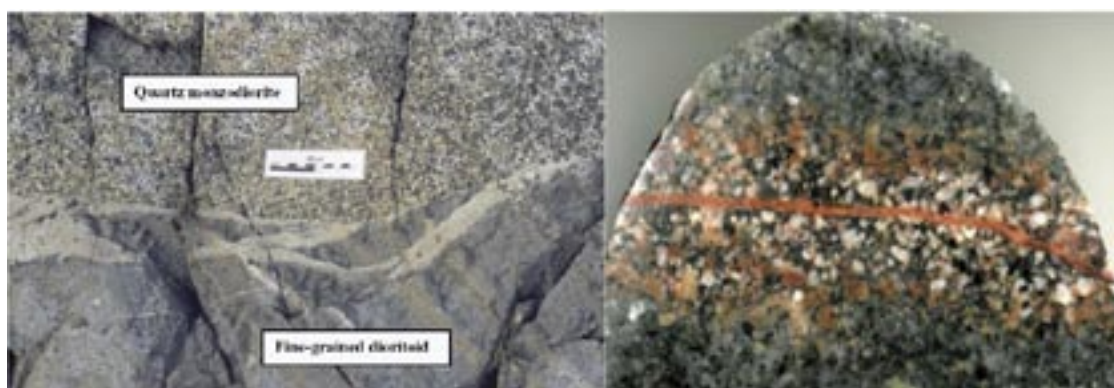
### 2.2.1 The rock mass

Along a flowpath, which is thought to be hosted in a number of connected open fractures (or at least partially open) in the rock mass, groundwater will likely contact a number of different rock types, each constituted of a number of different minerals. The rock types and mineral grains may be altered to different degrees and the degree of alteration may change along the flowpath, as well as with depth into the rock matrix from the flowpath. Depending on the setting, the alteration may be the result of metamorphic, tectonic, or hydrothermal processes or it may have occurred at low temperature under the influence of groundwater of various types interacting chemically with the rock.

To the left in Figure 2-1, an outcrop (~ 0.5×0.8 m) is shown where the rock type of the upper part is quartz monzodiorite and of the lower part is fine-grained dioritoid. Large fractures cut through the interface between the rock types and fractures along such interfaces are conceivable. Within such fractures groundwater containing solutes, such as radionuclides, may flow. Retardation of radionuclide migration is greatly dependent upon the properties of the rock matrix in contact with the flowpath, particularly for radionuclides that interact with mineral grain surfaces.

To the right in Figure 2-1, an alteration zone expanding from a now sealed fracture in a drill core sample (∅ 5 cm) is shown /Sandström and Tullborg 2005/. The width of the visibly altered rock matrix surrounding the fracture is around 2 cm. In alteration zones, host rock minerals have gradually been transformed to other minerals and in many cases, the porosity and possibly also the sorption capacity of the rock matrix may be substantially increased.

Below, the bulk rock mass will first be discussed, followed by a general discussion on the properties of altered rock adjacent to flowpaths.



**Figure 2-1.** Left: Fine-grained dioritoid and quartz monzodiorite outcrop in Laxemar. Image taken from /Wahlgren et al. 2004/. Right: Drill core from KFM05A in Forsmark, showing altered rock around a sealed fracture. Image taken from /Sandström and Tullborg 2005/.

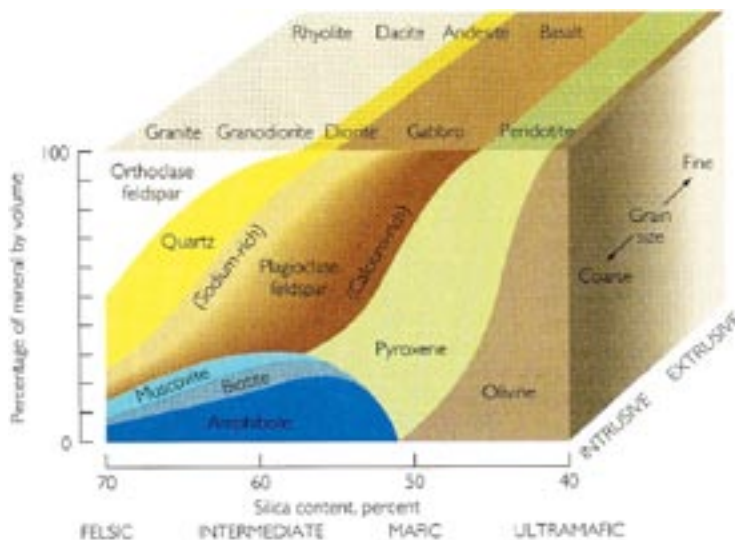
## Bulk rock mass

The bedrock in the eastern part of Sweden, including both the Forsmark and Oskarshamn site investigation areas, was formed 1,910 to 1,750 million years ago /SKB 2005b/. In the uplift that took place during the following few hundred million years, ductile deformation occurred, affecting large rock volumes. As the ductile deformation occurred, the rock was still hot ( $> 500^{\circ}\text{C}$ ) and plastic. Consequently, the rock matrix became structurally altered, although without the formation of brittle fractures. It is considered that a ductile structural alteration manifesting itself in anisotropy, for example, does not necessarily have a great effect on the retention capacity of the non-fractured rock matrix. Although the Forsmark site has been subjected to considerably more ductile deformation than the Oskarshamn site, the retention capacities of the rock matrices appear to be similar /SKB 2006c/. During the billion of years following the ductile deformation until present time, the rock matrix outside of brittle deformation zones has remained practically unchanged. As this time period vastly exceeds that of interest for repository evolution, the transport properties of the bulk rock mass can be considered to have no or little temporal variability.

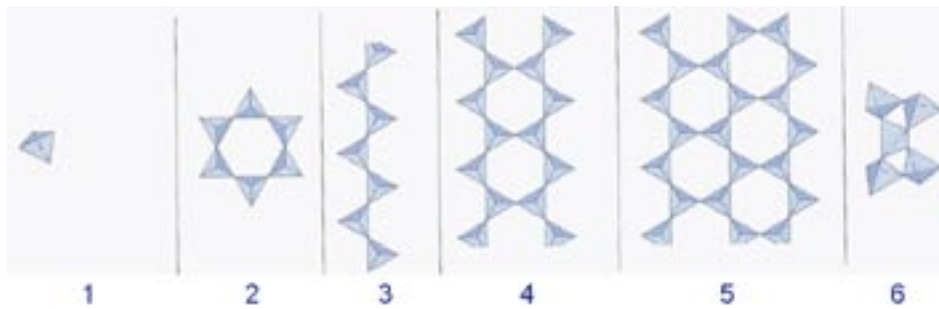
The dominating rock types at the Oskarshamn and Forsmark sites are granite, granodiorite, and diorite (more, or less metamorphosed). At the Oskarshamn site there is also a significant element of metavolcanic rock which displays an intermediate, dacitic to andesitic composition /SKB 2005b, Curtis et al. 2003/. Figure 2-2 shows the mineral composition of the most common rock types of intrusive and extrusive igneous rock.

Silicate minerals, such as those shown in Figure 2-2, are generally built by tetrahedra organised into different structures. The most common tetrahedron is  $\text{SiO}_4$ , which occurs in all abundant minerals. Another common tetrahedron is  $\text{AlO}_4$  that is abundant in, for example, feldspar. In most minerals, cations such as  $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{Mg}^{2+}$  are incorporated in the lattice between the tetrahedra, or bound at the lattice surfaces, so that electroneutrality is maintained.

Generally speaking, the lower the temperature at which the minerals crystallise, the more complex the lattice of tetrahedra becomes. Figure 2-3 shows the six major silicate structures starting with the least complex (isolated tetrahedron) and ending with the most complex structure (framework), where the corners of the tetrahedra (not necessarily all) are linked to other tetrahedra in a three-dimensional network /Press and Siever 1998/.



**Figure 2-2.** Mineralogical composition of igneous rock. Image taken from /Press and Siever 1998/.

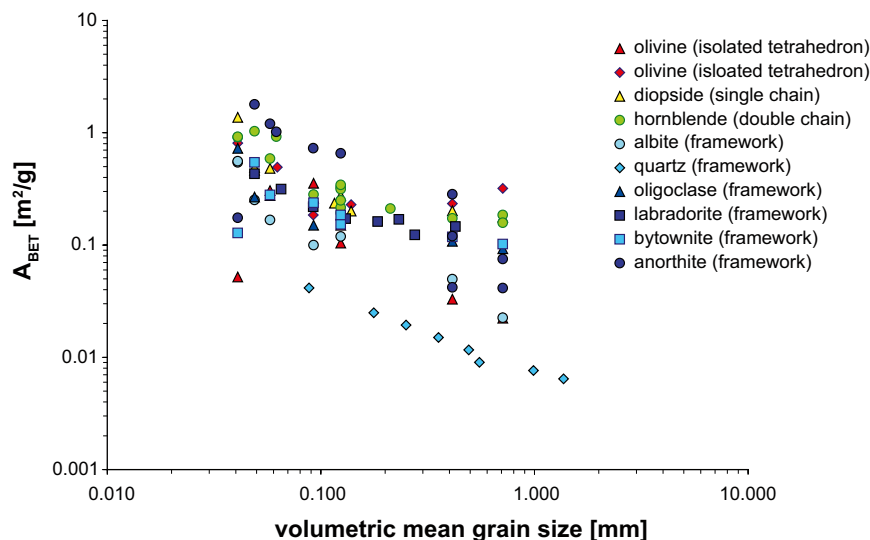


**Figure 2-3.** Major silicate structures. 1: Isolated tetrahedron, 2: Ring of tetrahedra, 3: Single chain, 4: Double chain, 5: Sheet, 6: Framework. Image taken from /Press and Siever 1998/.

Out of the minerals shown in Figure 2-2, quartz and the feldspars have a framework structure, biotite and muscovite have a sheet structure, amphibole has a double chain structure, pyroxene has a single chain structure, and olivine has an isolated tetrahedron structure. Where the lattice ends, at mineral surfaces for example, the surface may have a net negative charge owing to the unsatisfied valence charges of terminal oxygen atoms (surface oxide groups). In aqueous solutions, cations will associate with these charged sites and maintain electroneutrality. A net charge at mineral surfaces can also arise due to so-called “isomorphic substitution” of certain elements within the mineral lattice with other elements possessing a higher number of valence electrons, for example,  $\text{Al}^{3+}$  replaced by  $\text{Fe}^{2+}$  or  $\text{Mg}^{2+}$ . As a consequence, the mineral surfaces will be negatively charged and this charge is balanced by aqueous cations.

As sorption occurs by way of association of ionic solutes with charged mineral surfaces, both the surface area and surface charge of the mineral are of importance for retention. A measure of the mineral surface area is the  $\text{N}_2$ -BET surface area. Figure 2-4 shows different surface areas for minerals obtained by this method. The data sets are taken from /Brandtley and Mellot 2000/ and /Hölttä et al. 1997/.

In Figure 2-4 it can be seen that the surface area ranges over as much as two orders of magnitude for different minerals at different grain sizes. There are no data for sheet silicates in Figure 2-4 but they are reported to have significantly higher surface areas. In /Allard et al. 1983/, for example, the surface areas for biotite and muscovite are reported to be around  $7 \text{ m}^2/\text{g}$ . The different surfaces of sheet silicate minerals, however, have different charge properties owing to their lattice structure which complicates the relationship between surface area and sorptivity.



**Figure 2-4.**  $\text{N}_2$ -BET surface areas of different minerals and different grain sizes. Image adapted from /Crawford et al. 2006/.

In Figure 2-5 the surface charge for quartz ( $\text{SiO}_2$ ) and feldspar, as well as the common weathering products montmorillonite and kaolinite, are shown as a function of pH.

As can be seen by Figure 2-5 and from the surface chemistry literature, the minerals which comprise granitic rock will generally have negatively charged surfaces at all reasonable groundwater pH-values.

In addition to the electrostatic properties of mineral surfaces, the geometric structure of the microporous system of the rock matrix is of great importance for retention. The connected microporous system functions as transport conduits into the rock matrix where the solutes can be sorbed on mineral surfaces or sequestered in the water filled pores themselves.

In a study at the Grimsel Test Site in Switzerland, /Möri et al. 2003/ characterised the microporosity of undisturbed granodiorite. The most abundant pore type was found to be grain boundary pores. Other pore types found were microfractures, sheet silicate pores, and solution pores. To the left in Figure 2-6, grain boundary pores surrounding a mineral grain are shown. To the right a microfracture cutting through mineral grains is shown. The images were obtained by using Scanning Electron Microscope imaging /Möri et al. 2003/.

Microfractures are thought to originate in brittle mechanical deformation and can be found with increasing intensity around brittle deformation zones. Microfractures are also thought to form as a result of the stress arising from changes in rock conditions during the uplift.

Grain boundary pores are thought to extend in a regular and consistent fashion throughout the entire rock mass (this is the position of the authors of this report although it is noted that the issue is still contested within the scientific literature /e.g. Heath et al. 1992, Miller et al. 1994/).

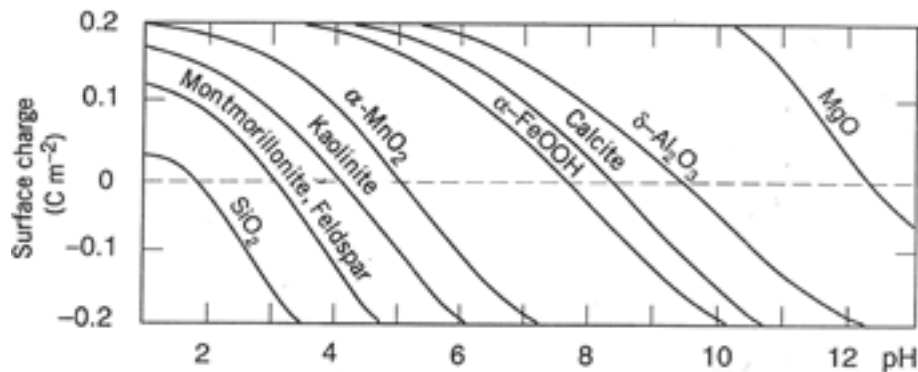


Figure 2-5. Surface charge of oxides. Image taken from /Stumm and Morgan 1996/.

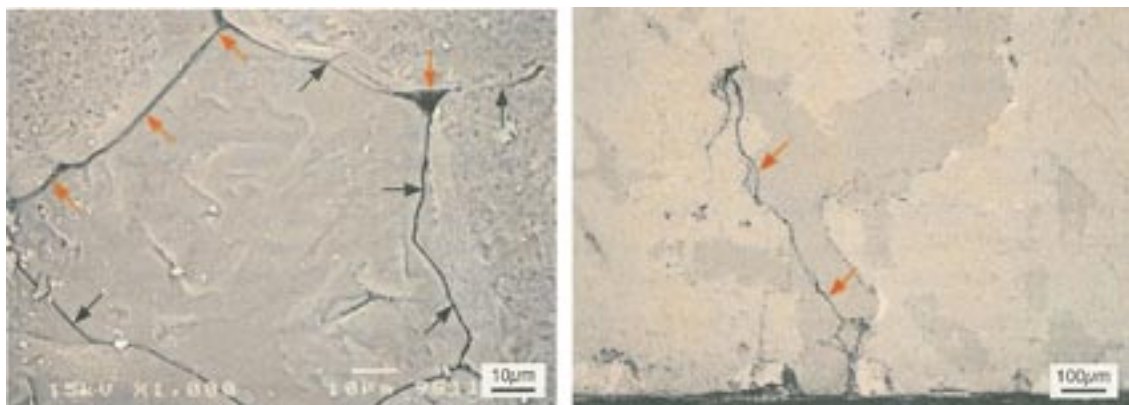


Figure 2-6. Left: Grain boundary pores. Right: Microfracture. Images taken from /Möri et al. 2003/.

A hypothesis suggested in e.g. /Löfgren 2004/ is that when the rock crystallises at great depths, the grain boundary porosity is practically non-existent. In Figure 2-6, one can see how the irregularly shaped mineral grains have crystallised so that they become as closely packed as possible. As the rock is uplifted the temperature decreases, mineral grains become increasingly rigid, and the pressure decreases. As the minerals have different thermal expansion coefficients and bulk moduli of expansion, some grains will expand while others will contract in the uplift. This uneven change in mineral grain volume will give rise to a void, i.e. the grain boundary porosity shown in Figure 2-6. Above a certain temperature, void creation is likely to be counteracted by recrystallisation and other processes associated with metamorphism. However, at relatively shallow depths in the earth's crust (although still at very large depth in comparison to repository depth), metamorphism ceases due to low temperature conditions. Uplift from these depths may create the grain boundary pores seen today at repository level. The differential expansion of mineral grains may also lead to intergranular microfracturing as an additional process also giving rise to porosity.

If this hypothesis is true, it would suggest an expansion of grain boundary pores throughout the entire rock mass. The processes involved would ensure that the grain boundary pores are connected and thus a pore connectivity extending throughout the entire rock mass can be proposed on theoretical grounds. It should be noted that Swedish crystalline rock may feature a higher content of microfractures, compared to grain boundary pores, than the rock found at the Grimsel Test Site. However, as microfractures generally cut through mineral grains, they also tend to intersect or terminate at grain boundary pores and thus become connected.

The connectivity of the matrix porosity is further discussed in subsection 2.4.2 of this report, in the context of limited and unlimited matrix diffusion. It is suggested by the authors of this report that more effort is devoted into explaining the origin and modelling the evolution of the microporous system given the controversy that still surrounds this important issue.

### ***Altered rock adjacent to flowpaths***

Owing to the exceptionally low hydraulic conductivity of crystalline igneous rock, groundwater tends to flow in discrete fractures along brittle and brittle-ductile deformation zones. Brittle and brittle-ductile deformation occurred primarily more than 1 billion years ago /SKB 2005b/. During the evolution of the bedrock, the fractures may have been sealed by precipitates and reactivated in tectonic events in a sequence of cycles. The brittle deformation occurred at rock temperatures at, or below, a few hundred degrees Celsius and the rock matrix adjacent to flowpaths may have been contacted by hot hydrothermal groundwater flowing from greater depth. Due to the elevated temperatures, the mechanical stress associated with the brittle deformation and later tectonic events, and contact with groundwater containing reactive constituents at elevated temperature (as well as in later stages), the rock adjacent to flowpaths is more or less altered.

Figure 2-7 shows a drill core sample ( $\varnothing$  5 cm) from a brittle deformation zone in Forsmark that is intersected by a number of fractures. It can be clearly seen in the figure that the fracture surfaces are rough and that the extents of fracture minerals and altered rock vary along the fracture zone and between the fractures.

In assessing the radionuclide retention properties of the rock in the immediate vicinity of a fracture, information concerning unaltered rock may not necessarily be directly transferable to the altered rock zones. Additionally, the degree and extent of alteration around one fracture may not be the same as for other fractures at a corresponding depth. This may occur, for example, where some fractures have been subject to a hydrothermal event while other fractures may have been sealed although subsequently reactivated at a later time.

This presents a major problem when assessing the rock retention capacity in many field tracer tests, as most tracers will not penetrate even the thinnest layer or alteration zone around a fracture within the experimental timeframe. It is not always clear what relevance the interaction of tracer solutes with the outermost layers of these alteration rims has for the retardation of

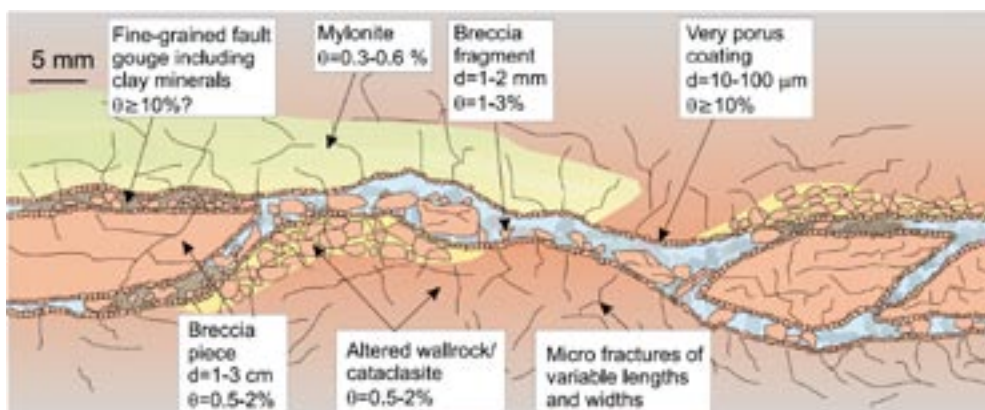


**Figure 2-7.** Fractures intersecting a drill core sample from KFM05A. Image taken from /Sandström and Tullborg 2005/.

radionuclide transport at Safety Assessment timescales where penetration depths are typically significantly greater.

In the TRUE Block Scale project /Winberg et al. 2003/, which is one of the most extensive tracer test programmes carried out in granitic rock in Sweden, the following conceptual model of a water-bearing fracture was adopted (Figure 2-8). The rock volume that is represented by Figure 2-8 is of approximately the same scale as that of the drill core sample depicted in Figure 2-7.

The numerical values shown in Figure 2-8 are representative for the specific location where the TRUE Block Scale tracer tests were carried out and only their orders of magnitude are of interest here for a general discussion. The conceptual image exhibits many of the same features discussed above. Within the fracture, small pieces of rock (or former sealing materials) called fault gauge are shown which have broken off in brittle deformation processes and other tectonic events. The rough fracture surfaces, in combination with fault gauge, create hydraulic hindrances (bottlenecks) for flowing water. At the fracture surface there may be a thin layer of fracture coatings. Precipitates and mineral alteration products constitute the coating. The extent and properties of this layer have not yet been well characterised and they potentially differ greatly from fracture to fracture. The rock surrounding the fracture may be more or less altered, both in terms of mineralogy and its porous structure. In Figure 2-8, the altered rock is represented as being mylonite and cataclasite. It is also possible for the rock matrix found on opposing fracture surfaces to have different material properties if the fracture is found at the boundary of different rock type sub-domains such as dykes, or other igneous intrusions. It may also occur in reactivated fractures if the mode of fracture reactivation leads to an asymmetric distribution of altered materials on flanking sides of the fracture.



**Figure 2-8.** Conceptual model of a water-bearing fracture. Image taken from /Winberg et al. 2003/.

Not all hydraulically conductive fractures necessarily display the same degree of complexity as the ones shown in Figure 2-7. Figure 2-9 below shows a drill core sample from Forsmark featuring two intersecting fractures, both of which are now sealed but in the past held water. It is interesting to note the different mineralogy of fracture sealing materials, as evident from the infill colours of the crossing fractures, possibly indicating that the fracture may have been open at different time periods. One can thus speculate if they have ever been part of the same flowing system. These fractures appear to have fairly smooth surfaces, compared to those in Figure 2-7, and one can suspect that the magnitude of radionuclide retention differs in the different types of fractures.

At present it is not clear to what extent hydraulically conductive features of the greatest importance for repository safety are best described by complex fractures systems, such as shown in Figure 2-7, or by single fractures, such as shown in Figure 2-9 (with the prerequisite that they are reactivated and open).

## 2.2.2 Deformation zones and fractures

In situ tracer tests are commonly carried out in “deterministically” interpreted fractures (or clusters of fractures) within brittle deformation zones. For practical reasons, the tests are generally performed in reasonably well connected and hydraulically conductive features where groundwater pumping can be performed. The rock mass surrounding the deterministically interpreted fractures is likely to be intersected by swarms or clusters of minor fractures that can only be described stochastically. Information concerning deformation zones and fractures in the rock volume of interest for a tracer test is normally obtained by drilling boreholes and examining the boreholes geologically, hydraulically, and geophysically. If drill cores are available, these are also examined.

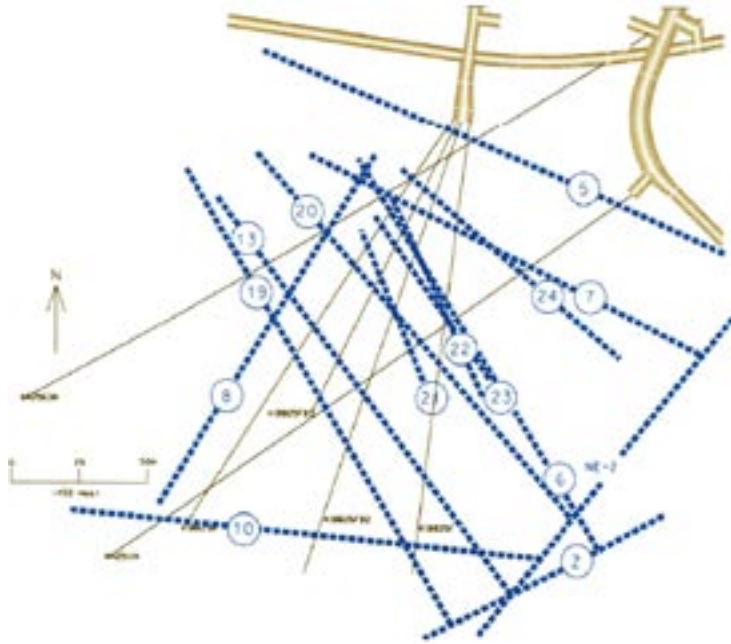
Figure 2-10 shows a model layout of some of the hydraulically conductive structures interpreted from investigations of the TRUE Block Scale rock volume (blue dashed lines). Here, the conductive features are depicted as simply-connected planar features although this does not necessarily need to be the case, as large conductive features are often comprised of clusters of smaller fractures that are interpreted en echelon as single structures. As this volume is in the vicinity of the Äspö HRL tunnel (shown in the upper right corner), it was possible to drill a number of boreholes through the rock volume (solid dark lines) to intersect the structures. This made it possible to characterise the variability/heterogeneity along a number of key structures /Andersson et al. 2002a/.

Far from all fractures observed in situ are open under present day conditions. As mentioned earlier, for example in connection with Figure 2-9, fractures may have been sealed by mineral precipitates and reactivated in tectonic events in a sequence of cycles. Adjacent fractures, especially of different fracture sets, may be open at different time periods and if so, they are not part of the same flow system.



*Figure 2-9. Drill core from KFM05A with sealed fractures. Image taken from /Sandström and Tullborg 2005/.*

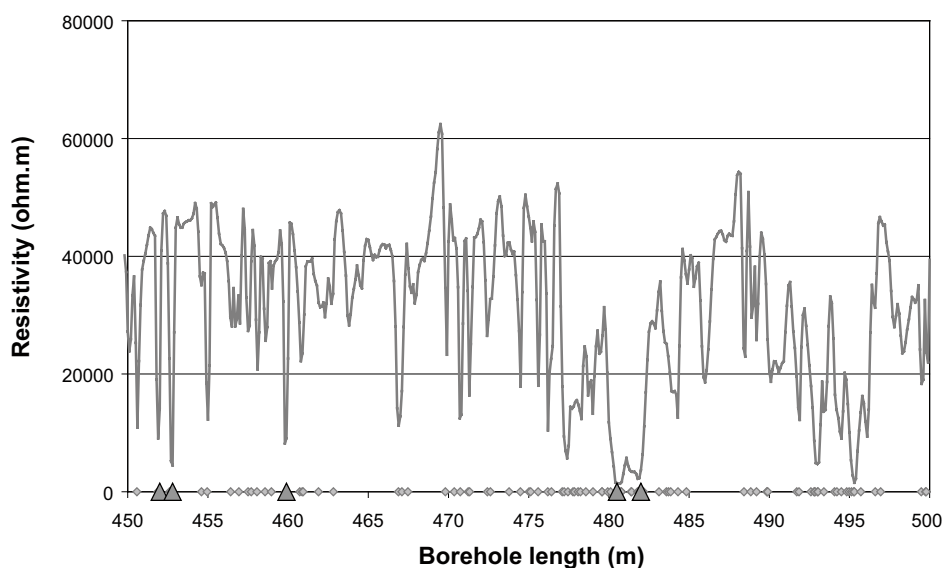




**Figure 2-10.** Hydrostructural model of the TRUE Block Scale rock volume. Image taken from /Hermanson and Doe 2000/.

The authors of this report consider that all open fractures (and all open portions of partially open fractures) are water-bearing at depth, although not all water-bearing fractures are sufficiently well connected to other water-bearing fractures in such a way that they can conduct groundwater (at measurable rates). Fractures that do conduct water are labelled as hydraulically conductive fractures in this report. We recognise that in different SKB reports, the label “water-bearing” may have different meaning.

To exemplify that fractures observed in situ may be water-bearing but not hydraulically conductive (at measurable rates), a rock resistivity log from a borehole section at roughly repository depth from the borehole KFM08A in Forsmark is shown in Figure 2-11.



**Figure 2-11.** Rock resistivity log and fractures in KFM08A. Image based on /Löfgren 2006/.

On the x-axis, which represents the borehole length measured from the surface, diamond symbols mark the position of fractures that part (i.e. physically separate) the drill core as detected in the drill core logging /Petersson et al. 2005/. The triangles mark the location of hydraulically conductive fractures detected by in situ flow logging using the Posiva difference flow meter /Sokolnicki and Rouhiainen 2005/. This tool has a lower detection limit of about 0.1 ml/min which means that there may be hydraulically conductive fractures with very small flowrates that remain undetected.

The solid grey curve in Figure 2-11 represents the rock resistivity obtained by in situ geophysical logging /Nielsen et al. 2005/. When logging the rock resistivity, an electrical current beam is sent out from a tool into the rock surrounding the borehole. A sharp anomaly (sharp dip in the curve) at a fracture suggests that the fracture is open, as the emitted current can be propagated more easily in the less resistive fracture water than in the rock matrix. What is not seen in Figure 2-11 are the locations of sealed fractures found in the core logging. In the interval shown in the figure only 40% of all fractures part the drill core, while the rest were logged as sealed /SICADA 2006/. Sealed fractures generally do not give rise to significant resistivity anomalies, although open fractures do. Amongst the fractures logged in the figure, less than 10% of the fractures parting the drill core appear to be hydraulically conductive. However, as can be seen by the many sharp resistivity anomalies, a much larger portion of the fractures seem to be open and water-bearing.

Based on the reasoning above, there appears to be a multitude of water-bearing fractures at repository depth, although the hydraulically conductive fracture network may be poorly connected. One can only speculate whether this is due to flow bottlenecks, as indicated in Figure 2-8 or due to a limited extent of, and connectivity between, the fractures. Based on the experience from investigating 20 deep core drilled boreholes within the Forsmark and Oskarshamn site investigation areas /e.g Löfgren and Neretnieks 2005, Löfgren 2006/ the authors of this report deem that the borehole section shown in Figure 2-11 is quite typical for both sites, even if the target rock volumes of interest for siting of a repository may feature less hydraulically conductive fractures.

## **2.3 Conceptual understanding of the flowing groundwater system**

Movement of groundwater through a porous system is controlled primarily by the prevailing hydraulic gradient and the hydraulic conductivity of the medium. Whether the flow occurs within the porous network of a soil, sedimentary rock or within fractures hosted in crystalline rock, in all cases the flow resistance of the medium (which is the inverse of hydraulic conductivity) arises due to viscous friction between the groundwater and the geological medium through which it flows.

### **2.3.1 Hydraulic gradients**

Hydraulic gradients generally arise from differences in altitudes and fluid densities, where differences in altitude are governed by the topography of the water table and differences in density are governed by the groundwater chemistry and temperature. The natural hydraulic gradient may also be affected to a minor extent by the air pressure, tidal effects, etc. The geological basis for a discussion of the hydraulic conductivity of the porous media, in this case a discrete fracture network, is briefly given in Section 2.2. In /Laaksoharju et al. 2004/ the following conceptual picture of the regional flow in the Oskarshamn area is given where the y-axis shows the altitude relative to sea level.

According to this model the flow is governed by the topography of the water table in inland regions. As can be seen in Figure 2-12, the contemporary regional hydraulic gradient is thought to be on the order of about 1%. During, and in the wake of a glacial period, hydraulic gradients are expected to increase considerably. At the present day in the inland regions shown in the

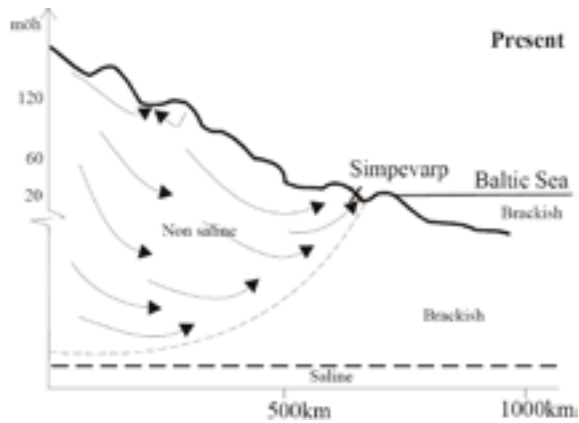


Figure 2-12. Regional groundwater flow. Image taken from /Laaksoharju et al. 2004/.

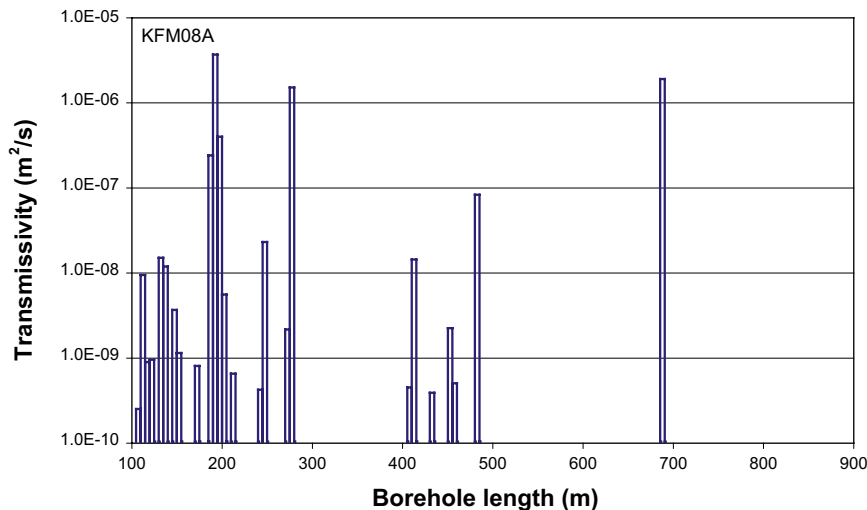
figure, the groundwater down to some depth is predominantly non-saline and originating from atmospheric precipitation (so-called “meteoric” water). At some depth, a halocline exists which restricts the vertical component of the groundwater flow, and the turnover or residence time of the saline water below the halocline is thought to be very large. At the coastline, brackish water from the Baltic Sea has infiltrated the bedrock. Brackish water is denser than non-saline water and as a consequence, the non-saline water flowing from the inland is forced towards the surface. It is thought that the coastal area of the Baltic Sea, in general, is a regional outflow area. It should also be noted that inflow and outflow areas are dynamic in nature and their location can be expected to vary during glacial cycles and the accompanying rebound of the crust.

In tracer tests, one usually performs pumping in order to achieve a reasonable tracer recovery during the timescale of the test. In most tracer tests the hydraulic gradient is increased by as much as three or more orders of magnitude relative to the naturally existing hydraulic gradient. This must be compensated for when transferring tracer test output data to Safety Assessment input data, which was done in for example the Äspö Task Force 6E /Hodgkinson 2007/. If a tracer test is performed in the vicinity of a tunnel, the presence of the tunnel is likely to induce a large hydraulic gradient towards it, which may need to be overcome by even more extensive pumping in order to achieve tracer recovery. In the TRUE series of experiments, for example, tracer tests were carried out in close proximity to the Äspö HRL tunnel. As a result, it was necessary to overcome tunnel-induced hydraulic gradients of 5–10% /Winberg et al. 2003/.

### 2.3.2 Hydraulic conductivity and fluid properties

The hydraulic conductivity of the rock/groundwater system depends on frictional forces arising between the flowing groundwater and the rock matrix. The friction depends on the geometry of the fracture network, the geometry of the fracture surfaces (including fracture coating, fault gauge etc), and the viscosity of the groundwater.

The non-fractured matrix of intrusive igneous rock can be considered, for all practical purposes, as being impermeable to flow. As a general rule of thumb, the hydraulic conductivity of this type of rock is considered to be on the order of  $10^{-10}$  m/s or lower. As an example, /Suzuki et al. 1995/ obtained a mean hydraulic conductivity of  $1.84 \cdot 10^{-11}$  m/s by performing measurements on 25 fresh biotite granite samples from Japan. As the rock matrix is largely impermeable, groundwater transport tends to occur in discrete fractures in brittle or ductile-brittle deformation zones, as discussed in Section 2.2. Depending on the degree of fracturing, the geometry of the fractures and the degree of connectivity of the fracture network, the hydraulic conductivity not only varies greatly from site to site, but also varies within a site. Figure 2-13 shows the transmissivity measured in 5 m sections by in situ flow logging in the borehole KFM08A in Forsmark /Sokolnicki and Rouhiainen 2005/. The transmissivity is the hydraulic conductivity integrated over the borehole interval measured. It is simply the proportionality linking flow and applied hydraulic gradient for a flowpath of given width.



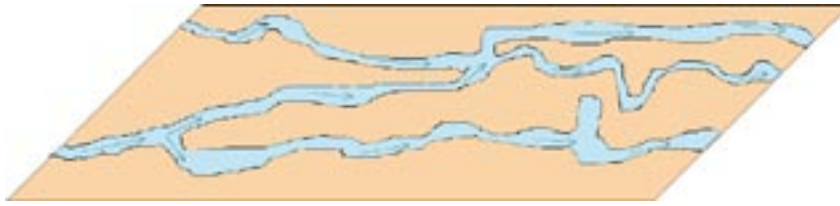
**Figure 2-13.** Transmissivity in KFM08A. Figure based on data from /Sokolnicki and Rouhiainen 2005/.

Even if the transmissivity log is very individual for different boreholes, some general features can be discerned in Figure 2-13. Firstly, the conductive fractures are very unevenly distributed in space. In some of the 5 m measurement intervals, water flows from a single fracture or many clustered fractures, while in other sections no conductive fracture can be identified. Secondly, the magnitude of the transmissivity varies greatly from section to section. The magnitude of the transmissivity is not directly correlated to the number of fractures in the section. Thirdly, shallower bedrock is more conductive than more deeply lying rock. This is thought to be at least partly due to the increasing pressure from overburden at depth, potentially giving rise to flowpaths with smaller apertures. This effect probably also has to do with processes that mechanically damage the more shallow lying rock resulting in, for example, sheet joints and a generally greater hydraulic connectivity. The fact that no transmissivity value below  $10^{-10}$  m<sup>2</sup>/s is shown in Figure 2-13 is related to the lower measurement limit of the in situ tool rather than a true physical limit.

The hydraulic conductivity is very unevenly distributed along a flowpath, which almost becomes self-evident if revisiting Figure 2-7 and Figure 2-8. If considering a fracture plane, the roughness of the fracture surfaces and its asperities (including the fracture coating and fault gouge) in combination with the hydraulic gradient and its orientation will cause the flow to become unevenly distributed over the plane. This phenomenon is often referred to as flow channelling. Figure 2-14 shows an artist's conceptualisation of channelling in a fracture plane, taken from the RETROCK project /SKB 2004/.

Channelling within fracture planes can arise due to: 1) the variable aperture of the flow porosity between adjacent fracture surfaces, 2) intersecting fractures where the fracture intersection itself acts as a flow tube or "wormhole" of enhanced transmissivity, 3) joint intersections featuring offset displacements. It is noted that both offset displacements at joint intersections and lateral displacement of mated fracture surfaces can also give rise to strongly anisotropic flow channelling effects. The presence of loose fracture filling materials and secondary mineralisations can also have a strong influence on the degree of channelling.

During the evolution of the bedrock, the hydrodynamic properties of a fracture change due to dissolution and precipitation processes within the fracture, and due to reactivation of sealed or partly sealed fractures in tectonic events. The transport of reactive constituents in strongly channelled flow can also give dissolution-precipitation feedback effects that may enhance channelling. This can have occurred previously during periods of hydrothermal alteration but may also occur at present day temperatures, if there are significant changes in the groundwater chemistry or in other conditions. In essence, water transported along fast pathways may react upon coming into contact with water or rock associated with slower flowpaths or stagnant



**Figure 2-14.** Conceptual image of channelling in a fracture plane. Image taken from /SKB 2004/.

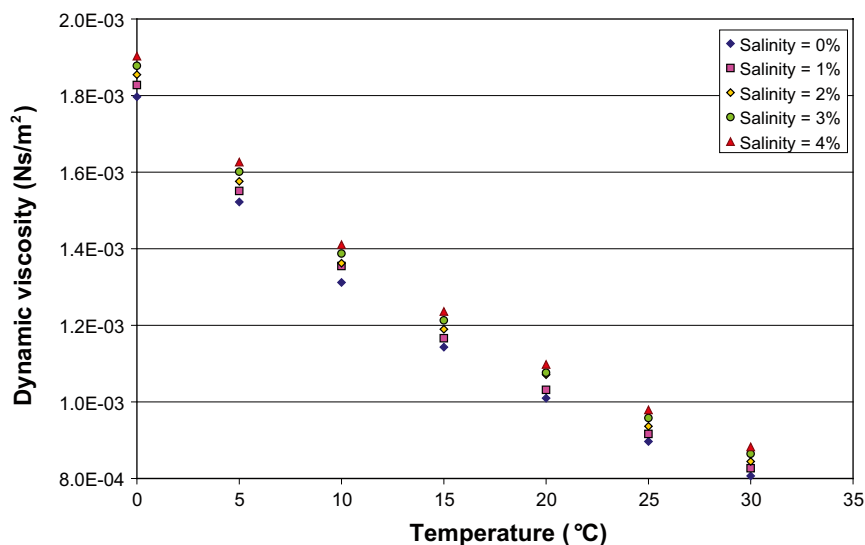
regions adjacent to the main flowpath. The reaction may be due to both differences in chemistry and in temperature. Enhancement of channelling may also occur along the fast flowpaths that may form at joint intersections, where spheroidal weathering effects can give rise to an increase in effective hydraulic aperture.

Channelling does not only affect the hydraulic properties of the fracture but also its retention capacity, especially retention by matrix diffusion, as channelling limits the contact area between flowing groundwater and the rock matrix, compared to the case where groundwater flows over the whole fracture surface /SKB 2004/. This is further discussed in Section 2.4.

When considering a three dimensional fracture network, the geometry of the individual fractures in combination with the degree of connectivity greatly affects the hydraulic conductivity of the rock volume as a whole. As a consequence, when scaling up from a tracer test, the flow situation becomes increasingly complex over greater length scales. In reality, the flowpath geometry is far more complex than the hydraulically conductive structures shown, for example, in Figure 2-10.

The flowrate in a conductive fracture is not only dependent on the hydraulic gradient and the fracture transmissivity. In addition, the viscosity of the groundwater is an important factor. Figure 2-15 shows how the viscosity of water of different salinities varies with temperature. The blue line corresponds to meteoric water, the green line to typical saline water, and the purple line to brine.

The flow situation does not only affect the groundwater chemistry as the groundwater chemistry can also influence the flow situation owing to, for example, buoyancy effects. In a long term perspective, the groundwater chemistry can also influence flowpaths as solutes precipitate and dissolve in different locations and times.



**Figure 2-15.** Viscosity of water, based on data from /Arthur D Little Inc 1962/.

In a tracer test, the groundwater situation becomes greatly disturbed due to excavations and pumping. Many of the precipitates on fracture surfaces are sensitive to changes in pH, redox potential, and solute concentrations. Therefore, the flowpaths may be altered during a campaign. It should be considered possible that the fracture system chosen for a tracer test may have become poorly representative for the rock volume at undisturbed conditions even before the tracer test begins.

## 2.4 Conceptual understanding of radionuclide transport

In granitic rock, all long-range radionuclide transport occurs by advective flow of groundwater in discrete fractures. As the pore water in the rock matrix is considered to be immobile, long-range transport through the solid rock mass can be neglected. Radionuclides can either be transported in the groundwater as free solutes (including aqueous complexes) or bound in some manner to colloids carried by the groundwater flow.

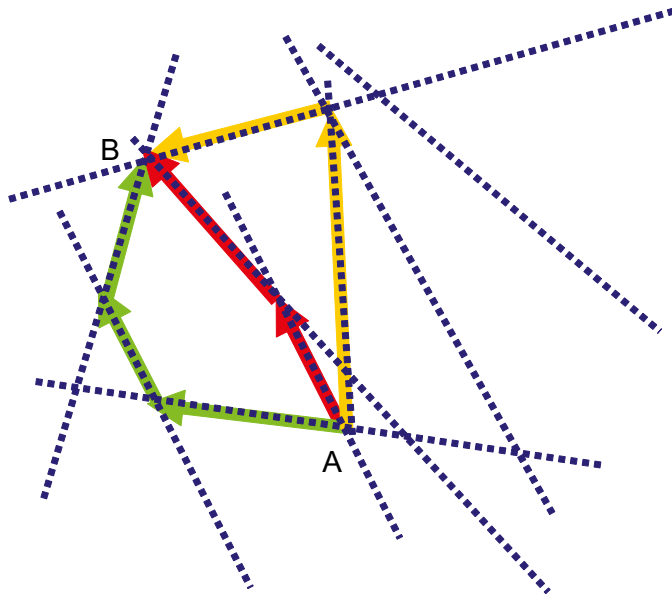
There are a number of processes that retard the transport of radionuclides in such a way that they are transported at a slower rate than the groundwater they are carried with. The most important process leading to retardation is matrix diffusion, accompanied by partitioning between aqueous and solid phases for many radionuclides. As discussed in previous sections, the rock/groundwater system is very complex and this gives rise to much spatial variability in parameters essential for radionuclide transport. An attempt to quantify the spatial variability is made in the Safety Assessment SR-Can /SKB 2006c/, to which the reader is referred. The complexity of the system and spatial variability of transport parameters should always be kept in mind when dealing with tracer tests. For an overview of solute transport mechanisms in fractured rock, the RETROCK Project /SKB 2004/ and /Neretnieks 1993/ are recommended.

### 2.4.1 Hydrodynamic dispersion

The description of solute dispersion in fractured rock flow systems is extremely complex and encompasses a range of different phenomena that give rise to a general spreading of the solute residence time distribution in the system. There is also an enormous amount of literature and research that has been carried out in the past two decades dealing with these phenomena (see e.g. /Berkowitz 2002/ for a broad overview). For the purposes of this report, however, we make a clear distinction between the dispersive effects of matrix interaction effects (here taken to include matrix diffusion and partitioning) and “true” hydrodynamic dispersion. In spite of this, we note that the effects of matrix interaction and hydrodynamic dispersion are frequently combined in formalised mathematical descriptions of anomalous dispersion /e.g. Margolin and Berkowitz 2000, Benson et al. 2000, Berkowitz et al. 2001/.

Put simply, hydrodynamic dispersion, other than Taylor dispersion, arises when different flowpaths conduct groundwater at different rates over a given geometric distance. In a fracture network the actual lengths of flowpaths in different fractures, or fracture clusters, from point A to B may differ. This is illustrated in Figure 2-16 where three flowpaths in hydraulically conductive features are highlighted in red, green, and orange. Figure 2-16 can be compared to Figure 2-10, but the rock volume should be considered generic.

The flowpath from point A to B, depicted by red arrows, is clearly shorter than those depicted by green and orange arrows. This does not necessarily mean that the red flowpath conducts water at a higher rate than the others, as each flowpath may have a different overall hydraulic transmissivity that may range over several orders of magnitude. In addition, other additional channelling effects (discussed below) may make the effective flowpath along the red path line longer than those along the green and orange ones.



**Figure 2-16.** Different flowpaths in structures in a generic rock volume close to an underground laboratory.

Hydrodynamic dispersion is not only a result of transport along flowpaths of different length in separate deformation zones. As can be expected from Figure 2-7, different subordinate flowpaths within a deformation zone may conduct water more or less effectively. As discussed previously, deformation zones are typically made up of clusters of partially oriented fractures and fracture splays that are interpreted as single “en echelon” features in hydrostructural models. As different members of these fracture clusters have different transmissivities and connectivities, it is not unexpected that they contribute substantially to the observed dispersion in a field scale experiment by way of fracture-to-fracture flow channelling effects. The interconnectivity and flow propagation properties of such fracture clusters (and fracture networks in general) is the subject of much discussion in the scientific literature and although there is broad agreement on a number of overarching issues, many of the fine details that are of importance for understanding solute transport are poorly understood or the subject of controversy /e.g. Berkowitz 2002/.

Within a single fracture, different parts of the fracture plane may also conduct water at different rates. This in-plane, flow channelling is illustrated in Figure 2-14 and discussed in Section 2.3. If the groundwater transported along different flowpaths is repeatedly and fully mixed along flowpath elements with spatially non-correlated and otherwise isotropic transmissive properties, the dispersion will be asymptotically Gaussian in the absence of matrix diffusion. In such a case, the hydrodynamic dispersion would only give rise to a symmetrical spreading of a tracer pulse with a constant apparent dispersivity as a function of transport distance. The tracer pulse would thus have the same mean residence time as the average volumetric residence time of the groundwater in the sampled flowpath. However, in tracer tests the flowpaths do not necessarily meet repeatedly, channel lengths may be correlated with transmissivity, and mixing may be poor. All of these features may result in non-Gaussian dispersion. For the transport of non-reactive or linearly sorbing solutes, this non-Gaussian dispersion typically manifests itself in the form of fast initial arrival times, multi-modal breakthroughs, and long tailing effects due to advection of solutes along poorly transmissive network paths.

Consequently, the apparent dispersivity observed in tracer tests may increase with geometric distance between injection and withdrawal points. Indeed, this scale-dependent dispersion is what is commonly observed in tracer tests (see the reviews by e.g. /Gelhar et al. 1992, Neretnieks 1993/). In the review by /Berkowitz 2002/ it is even suggested that Gaussian dispersion is exceptional and that non-Gaussian behaviour is the norm for most natural systems.

## 2.4.2 Matrix diffusion

Matrix diffusion is considered to be a major retention process that occurs within the stagnant pore water of the rock matrix /Neretnieks 1980/. According to classical Fickian diffusion theory, as long as a concentration gradient exists in a solution, solutes will diffuse down this gradient as a result of random molecular movement. This theory was originally developed for gases and later transferred to liquid binary mixtures. Stringently speaking, the theory is not applicable for multicomponent mixtures such as groundwater unless in very dilute solutions where the errors become insignificant. In addition it should be noted that Fickian diffusion theory does not take electrostatic forces into account and may therefore be unsuitable for the rock/groundwater system, where practically all solutes and mineral surfaces are charged.

It may seem peculiar that the use of Fickian diffusion theory is so widespread within the field of contaminant hydrology. However, as for the  $K_d$  concept for partitioning, decades of practical experience have shown that the approach is workable and gives reasonable results. If the radionuclide transport occurs in a rock/groundwater system that is reasonably close to diffusive equilibrium (i.e. with respect to the major solution components) and if the tracer concentration is small, multicomponent mass transfer effects become small. This is often the case when performing diffusion studies in the laboratory, where small tracer concentrations are used and where the rock sample in general is equilibrated with the background solution prior to the study. In many cases, small deviations from Fickian diffusion are easily masked by the general complexity of the system.

If the rock/groundwater system is far from equilibrium, for example in the wake of a glacial period, multicomponent mass transfer effects may become considerable. In most cases this only affects the magnitude of the diffusive flux, but in extreme cases species may even diffuse up their concentration gradients. It should be noted that in situ tracer tests are often performed in rock/groundwater systems that are greatly disturbed and may be far from equilibrium. Whether multicomponent mass transfer can significantly affect radionuclide retention in a tracer test or during repository evolution is at present not well understood.

A diffusion theory that may be more rigorous for the rock/groundwater system is the Maxwell-Stefan diffusion theory /Krishna and Wesselingh 1997/ where species diffuse down their so-called "potential" gradient. The potential gradient here is the sum of different potentials such as chemical potential, electrostatic potential, etc. In the Maxwell-Stefan diffusion theory, the diffusivity for a solute is determined not only by interactions with the solution but also with other solutes as well as with solids. The major drawback of the theory is that it requires a number of input parameters for each interaction that at present are not numerically well constrained by measurement data.

For the purposes of SKB, the authors of this report do not generally recommend the use of Maxwell-Stefan diffusion, or any other diffusion theory, over Fickian diffusion. However, one should always be aware of the fact that in such complex systems as the rock/groundwater system, models can only mirror reality to a limited degree. This should always be kept in mind when drawing conclusions from modelling interpretation of experimental results.

Dispersion of a transported solute arising from matrix diffusion can be considered mechanistically distinct from hydrodynamic dispersion although we note that some researchers consider matrix diffusion as part of a continuum of random-walk processes that give rise to non-Gaussian solute dispersion (e.g. /Margolin and Berkowitz 2000, Berkowitz et al. 2001, Cortis et al. 2004, Dentz et al. 2004/).

Matrix diffusion is customarily considered to comprise diffusion in the stagnant pore water that saturates the microporous system of the rock matrix. However, diffusion from flowpaths to stagnant water volumes can also occur in fault gouge, fracture coatings and in stagnant water within the fracture plane. Furthermore, it has been suggested that diffusion in water-bearing, but hydraulically non-conductive, fractures may be significant for retention of non-sorbing species /Löfgren and Neretnieks 2005/. Diffusion within stagnant water-filled fractures may



have important consequences for solute transport when consideration is also given to the additional matrix surface area potentially available for solute interaction within these fractures /Neretnieks 2006/. The fact that species can diffuse in multiple immobile zones possessing different diffusivities is well known. In the TRUE series of projects, much attention was given to developing a microstructural model of the pore space and the surrounding matrix at fracture surfaces. For further reading on how stagnant water adjacent to flowpaths affects transport, we direct the reader to e.g. /Poteri et al. 2002/. Phenomenologically, these processes are similar although characterised by different time constants. While diffusion in fracture infilling material is of great importance in a tracer test, it may be of much less importance during the evolution of the repository. In the latter case, transport over much larger timescales will allow radionuclides to penetrate into the rock matrix beyond the thin layers of fracture coatings.

The rate of diffusive transport in the rock matrix is (in addition to the concentration gradient) determined by:

- The diffusivity of the species in the pore water.
- Partitioning reactions.
- The geometry of the porous system (the formation factor).
- Non-partitioning interactions between solutes and mineral surfaces.

Often the diffusivity in the pore water is approximated by the diffusivity of the species at infinite dilution. However, as the diffusivity is proportional to the pore water temperature and inversely proportional to the pore water viscosity (which itself is also a function of temperature), corrections should be made. The intrinsic temperature correction, from for example room temperature to in situ temperature, only affects the diffusivity by a few percent. The temperature correction for viscosity is of greater importance, as the viscosity is significantly affected by the pore water temperature, in addition to the salinity (see Figure 2-15).

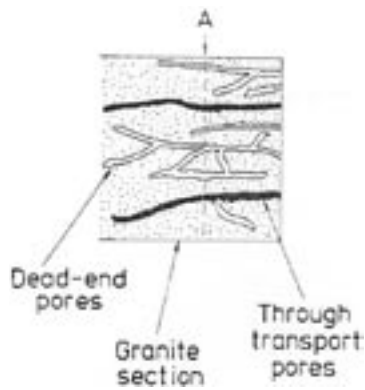
Partitioning is discussed in the following subsection although it can be mentioned already here that the immobilisation of species at mineral surfaces decreases their apparent diffusivity in the rock matrix. On the other hand, it also enables greater amounts of solutes to be transported into the rock matrix, as concentrations in the pore water are kept low for longer periods of time.

There are four main entities associated with the geometry of the porous system that affect the diffusive transport. These are; the porosity, the constrictivity, the tortuosity, and the pore connectivity. The combined effect on the effective diffusivity from the porosity, tortuosity, and constrictivity is internalised in the formation factor. As transport can only occur in the pore water and generally not through mineral lattices themselves, the volume available for transport is restricted. The connected porosity can be divided into transport porosity and dead-end porosity /e.g. Bradbury and Green 1986/. Non-connected porosity, such as fluid inclusions, is believed to be of no importance for matrix diffusion.

In the literature, transport porosity is often thought to be comprised of pores where both ends are connected to the micropore network. Dead-end porosity is often thought to be comprised of pores where only one end of the pore is connected to the rest of the micropore network. Solutes can thus be transported in or out of the pore, but not through the pore /e.g. Ohlsson 2000/. This way of conceptualising the porous system is well illustrated in, for example, /Bradbury and Green 1986/ (see Figure 2-17).

Dead-end porosity is also referred to as storage porosity /e.g. Skagius 1986/, although by using the latter nomenclature there is a risk of confusion with the so-called storage capacity of the porous system which is the sum of transport and dead-end porosity.

Picturing pores in granitic rock to generally have “ends” is in the view of the authors of this report somewhat misleading, as it suggests that most pores are wormhole like structures through the solid rock matrix. It is not inconceivable that such wormhole structures exist, originating in ruptured fluid inclusions, for example. However, grain boundary pores and



*Figure 2-17. Illustration of the dead-end pore concept. Image taken from /Bradbury and Green 1986/.*

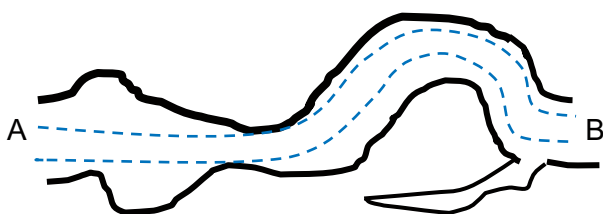
microfractures (see Figure 2-6), which have been identified as very important for matrix diffusion /Möri et al. 2003/, are better described as voids surrounding or intersecting mineral grains in a three-dimensional network, very much deviating from the two-dimensional pores in Figure 2-17). When considering grain boundary pores one may wonder how it is conceived that this three-dimensionally connected void should have “ends” as such.

Figure 2-18 shows a sketch of a tortuous and constrictive pore. The sketch could be viewed as, for example, a cross section of a plane between two mineral grains.

The dashed blue line shows a hypothetical volume where all transport could occur without constrictivity effects. In addition to constrictivity effects, the diffusive flux is also influenced by the tortuosity of the micropore network, as radionuclides need to be transported a longer distance from point A to B (in Figure 2-18) than that described by the nominal straight line distance.

The question of how well the microporous system is connected is, together with the contact time, of great importance for how deeply solutes can penetrate into the rock matrix, and thus how much of the retention capacity of the rock matrix one can utilise in Safety Assessment. It is stated in RETROCK Project /SKB 2004/ that “the value assigned to the penetration depth has a profound effect on the release rates to the biosphere” but still a consensus on the topic could not be reached and the issue was categorised as an unresolved problem. In subsection 2.2.1 of this present report we argue for the concept of unlimited (or at least long-range) matrix diffusion.

In recent years the notion of anomalous or non-Fickian diffusion processes has gained considerable attention in the scientific literature (see e.g. /Sahimi 1995, Haggerty and Gorelick 1995, Haggerty 1999, Saadatfar and Sahimi 2002/). The concept of dead-end porosity is central to the phenomenological description of anomalous diffusion. In /Haggerty 1999/ (and citations within) the porous system is described as fractal, where fractures and pores branch off in ever smaller fractures or pore spaces. Such a fractal system inevitably gives pores with such small apertures that diffusion of species becomes sterically restricted and, at some level, the pores will function as dead-end pores. Diffusion in such a system will according to /e.g. Haggerty 1999/ be non-Fickian and is called anomalous. The degree to which diffusion is actually anomalous, however, is largely dependent upon the length scale under consideration as it is intrinsically



*Figure 2-18. Sketch of a constricted and tortuous pore.*

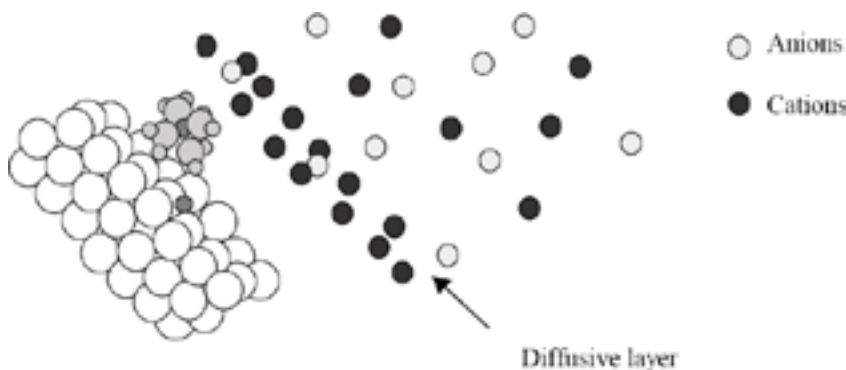
linked to the mathematical description of the immobile zone as a continuum. This means that on the level of individual grain boundary pores and microfractures, diffusion is almost always amenable to a Fickian or Maxwell-Stefan treatment (where there are significant multicomponent or electrostatic effects). This is largely true even when steric effects, anion exclusion, and surface diffusion in the electrical double layer are considered.

From a conceptual point of view, the paragraphs above highlight that there are different views upon the geometry of the porous system. In subsection 2.2.1 of this report, the porous system is conceptualised in a way which implicitly assumes the porous system to be connected throughout the rock mass. In /Haggerty 1999/, a conclusion is that the assumed fractal porous system becomes poorly connected over longer distances, giving rise to “anomalous” diffusion and diffusivities that approach zero over increasing length scales. The porous system has also been suggested to be close to the percolation threshold /e.g. Haggerty 2002/. This may occur in both fractal and non-fractal systems if the pore connectivity is sufficiently low. A porous system close to the percolation threshold would indeed manifest itself in the form of a decreasing effective diffusivity with increasing length-scale of the diffusion distance.

To solve the question of which conceptualisation is most appropriate, information from traditional field-scale tracer tests is of little use as the penetration depth is too small and the general uncertainties associated with tracer tests would mask effects from the geometry of the porous system. Instead, information obtained from the laboratory and also from geophysics, geology, and rock mechanics should be used. In /Löfgren and Neretnieks 2006/ it is proposed that the effective diffusivity of undisturbed intrusive igneous rock does not generally decrease with the length-scale of diffusive pathway. The porous system would thus not be close to the percolation threshold. This is based on interpretation of results from tracer tests performed in the laboratory on small-scale (cm) and medium-scale (dm) samples, and on geophysical measurements performed in situ on larger scales (m). In the small- and medium-scale investigations, tracers actually penetrated the samples in the way that would be expected if the porosity was well-connected over longer distances. The large-scale investigation was performed by a method described in subsection 3.6.2 of this report and the results support the same conclusion.

As solutes in general are in the form of ionic charged species, they will interact with mineral surfaces that predominantly are negatively charged. Such interactions are well described in the sorption literature (see e.g. /Stumm and Morgan 1996/). In Figure 2-19 the negatively charged mineral lattice is shown to the left and ionic solutes are shown to the right.

At the mineral surface, non-hydrated cations form inner-sphere complexes and are thought to be covalently bound to functional groups of the lattice (shown by a single grey sphere at the surface in Figure 2-19). These cations are considered to be so closely bound to specific sites that they are immobile. Inner-sphere complexes do not generally balance the entire negative charge of mineral surfaces in granitic rock. As a result, hydrated cations are bound by electrostatic forces to the surface as outer-sphere complexes (shown by the grey sphere surrounded by water



**Figure 2-19.** Solute at a mineral surface. Image taken from /Löfgren and Neretnieks 2002/ and based on /Stumm and Morgan 1996/.

molecules in the figure). It is conceptualised that as the outer-sphere complexes are so close to the surface, they are more or less bound to specific functional groups. Therefore, their mobility is considered to be significantly reduced, if they are considered to be mobile at all.

In many cases, even the inner- and outer-sphere complexes together cannot balance the total negative charge of mineral surface. Therefore more hydrated cations are attracted towards the surface. At some distance, the more closely bound cations shield the functional groups of the lattice from less closely bound cations to such an extent that only the potential gradient normal to the surface affects the movement of the less closely bound cations. For these cations, transport parallel to the surface is relatively unhindered while movement normal to the surface is restricted by electrostatic forces. The volume these cations occupy is often referred to as the diffuse double layer or the electrical double layer (EDL) /e.g. Stumm and Morgan 1996/. At what distance from the surface the outer-sphere complexes become cations in the electrical double layer is not well-defined.

As an accumulated concentration of dissolved cations can occur in the electrical double layer, this may give rise to a large concentration gradient laterally along the mineral surface. If, at the same time cationic movement parallel to the mineral surface is unrestricted, it is hypothesised that this may give rise to an increased diffusive flux /e.g. Ohlsson and Neretnieks 1998/. In a number of publications, such enhanced diffusion has been called “surface diffusion” /e.g. Axe et al. 2002/, while in other publications this is called “diffusion in the electrical double layer” or “surface related diffusion” /e.g. Marimon 2002/.

While cations are attracted to the negatively charged mineral surfaces, anions are repelled. This limits the cross sectional area of the pore space available for anion diffusion and storage and the phenomenon is thereby called anion exclusion. Another exclusion phenomenon is size exclusion, where species are excluded from some pores as they are too large. At present, exclusion effects in crystalline rock have been investigated at atmospheric pressure in the laboratory. How the increased stress at depth affects anion and size exclusion, however, is not well established.

### **2.4.3 Partitioning**

Partitioning of radionuclides between the aqueous and solid phases is of utmost importance for the safety of a repository, in case the integrity of the engineered barriers becomes compromised. Here we consider partitioning of solutes to include radionuclides being precipitated and co-precipitated, but also sorbed in some manner to solid surfaces in the manner described in previous discussions. In the case where the solids are immobile, such as minerals in the rock matrix or larger particles of fault gouge etc, partitioning will retard the radionuclide transport. In the case where the solid particles are so small that they are transported with the groundwater in the form of colloids, partitioning may increase the radionuclide transport rate compared to the transport rate of freely dissolved solutes. Such small particles may be organic or inorganic in origin. In a similar fashion, complexing ligands of various kinds in the groundwater may also reduce the sorptivity of transported solutes, thus reducing retention. Even though partitioning may both increase and decrease radionuclide retention, it is thought that the overall effect of partitioning on repository safety is substantially beneficial in that it reduces radionuclide transport.

At present there is much discussion within the scientific community as to what extent different partitioning processes affect radionuclide retention. On the one hand, there are arguments that precipitation and co-precipitation are of crucial importance for understanding retention /e.g. Miller et al. 2000/. This may be of importance when the groundwater chemistry and other conditions are changing, for example in the wake of a glacial period or during a tracer test. On the other hand, there are also counter-arguments that it is sufficient, when demonstrating the safety of the repository, to only account for partitioning due to linear sorption of radionuclides to mineral surfaces /e.g. Westall 1995/.

### ***Precipitation and co-precipitation***

Precipitation and co-precipitation processes are to a large extent determined by the hydrogeochemical conditions of the site. Cations and anions will precipitate in the form of secondary minerals when the groundwater becomes oversaturated. This may occur if the groundwater composition or temperature are subject to change. In a similar fashion, precipitates may dissolve if the composition or temperature conditions change in such a way that this is thermodynamically favourable. As the groundwater/rock system is a multicomponent system and there are a number of competitive precipitation/dissolution reactions that may occur, fully understanding and modelling such systems have proved challenging. When modelling the precipitation/dissolution reactions, consideration of chemical kinetics may be appropriate in addition to thermodynamic process descriptions, especially in the case of short-term tracer tests.

The situation is additionally complicated by the fact that co-precipitates can occur at concentrations well below what would be predicted from pure phase solubilities. In such cases, the solute may be incorporated in secondary minerals formed by the precipitation of other groundwater constituents (e.g. calcite, barite, etc). This may be of some importance as radionuclides are seldom expected to be at high enough concentrations to precipitate as pure mineral phases. During dissolution of a co-precipitate, the solubility of the minor constituent may differ significantly from that of its pure precipitate /Stumm and Morgan 1996/.

Owing to the presence of the EDL, the chemical conditions at a mineral surface may differ from those in the bulk solution. In many cases the electrostatic potential originates from functional groups at the surface, which may also function as counter ions in a precipitated solute. Due to the altered conditions, surface precipitates may therefore form at concentrations lower than that required for precipitation in the bulk solution. At very high sorbate/sorbent ratios, surface precipitation may become the dominant apparent sorption mechanism /Stumm and Morgan 1996/.

### ***Linear sorption – the $K_d$ concept***

In safety cases where radionuclides escape from the engineered barriers in a repository, it is generally argued that concentrations in the groundwater will be sufficiently low that partitioning can be approximately described as a linear sorption process. The linear sorption concept assumes that the sorption of a radionuclide is not affected by surface loading or multicomponent competitive effects, or at least that this is internalised in the partitioning ratio determined in laboratory experiments.

For a radionuclide to sorb linearly there are a few prerequisites. Firstly the sorption site should be unoccupied by an equal or a competing radionuclide, which is likely if the number of sorption sites vastly exceeds the number of radionuclides in the dilute solution. Secondly, the radionuclides sorbed to adjacent or nearby sites should not affect the sorptivity of dissolved radionuclides to otherwise unoccupied sites. Again, as the concentrations are small, it is thought that the radionuclides on average sorb to sites with sufficient distance between them so that they do not affect each other to a large extent. It is also implicitly dependent on the groundwater composition being sufficiently constant that the changing competitive effects of naturally occurring solutes do not alter the sorptivity of the radionuclide in an unpredictable fashion. A third prerequisite is that the solute binding affinity of each site, with respect to a specific radionuclide, is of equal strength. As discussed in connection to Figure 2-19, sorption at different sites may occur by different mechanisms and different sites featuring similar binding mechanisms may also sorb with different affinities for specific solutes.

With this pre-requisite in mind it becomes apparent that the  $K_d$  concept is not mechanistic, as one simply disregards the effect of different sorption mechanisms. Instead it is assumed that solutes “on average” sorb with a certain affinity to a surface. The reasoning above is equally valid for de-sorption processes.

What does affect the partitioning in this concept is the equilibrium between sorbed species and solutes. With the prerequisites discussed above, linear equilibrium is assumed with the constant partitioning coefficient or  $K_d$  ( $\text{m}^3/\text{kg}$ ). Even though the equilibrium is assumed to be unaffected by the sorption of other radionuclides, it is influenced by the concentrations of major groundwater species and by other factors. Some of these factors are well defined, or at least reasonably easy to identify in a Safety Assessments as important variables influencing sorption variability. Principal variables of this kind might include rock type, mineral surface area, temperature, ionic strength, pH, redox state, and the presence or absence of complexing ligands.

As the  $K_d$  concept has non-mechanistic basis, it has a limited ability to extrapolate data obtained under specific physicochemical conditions to other conditions. As stated in Crawford et al. 2006/ a  $K_d$  value “is an empirical snap-shot of a system that is only strictly applicable under those exact conditions under which it is measured and is not explicitly dependent upon any specific sorption mechanism or speciation considerations”. It should be mentioned that there have been attempts to approach partitioning from a mechanistic standpoint although much work remains before such an approach is practical to use in an engineering safety sense for a real rock/groundwater system.

### ***On neglecting partitioning mechanisms***

As mentioned previously, there is much discussion within the scientific community as to what extent sorption or alternatively precipitation reaction mechanisms should be relied upon as a description of retardation in a Safety Assessment. It should be clarified that basing a Safety Assessment on sorption and not precipitation, or vice versa, does not mean that one believes that the excluded partitioning mechanism is of no importance in nature. It only means that one is confident that sufficient repository safety can be demonstrated also when conservatively excluding one partitioning mechanism. It must be cautioned however that what is conservative in one situation, for example in a base case, may be non-conservative in other situations.

Even if one can argue that sufficient retention can be demonstrated in a Safety Assessment where one or more partitioning mechanisms are excluded, such arguments are not directly transferable to evaluation of tracer tests. In the analysis of tracer tests one often tries to model breakthrough curves so that they resemble those obtained in the test. Input parameters in such a modelling exercise, such as the porosity, matrix diffusivity, sorption capacity etc are often later transferred to Safety Assessments. Unless all significant partitioning mechanisms contributing to the retention manifested in the measured breakthrough curve are reasonably well accounted for, there is a risk that the modelling becomes merely a curve fitting exercise.

Using data extrapolated from such curve fitting exercises in a Safety Assessment without further consideration may be unwise. For example, the retention obtained in a tracer test may be significantly affected by precipitation processes. If this is not accounted for in the data evaluation, rationalised by the fact that precipitation is excluded in the subsequent Safety Assessment, the  $K_d$  value may be substantially overestimated and may not be physically meaningful.

#### **2.4.4 Flow-wetted surface**

If revisiting Figure 2-16, showing different flowpaths within a generic rock volume, one can see that the red flowpath is shorter than the green and orange ones. Thus, if the fractures were planar and fracture surfaces smooth, one could expect the contact area between the flowpath and the surrounding rock to be smaller for the red flowpath than for the others. However, as can be seen in Figure 2-7 and Figure 2-8, fractures are generally not planar and fracture surfaces are rough. Therefore, any of the flowpaths may have the larger contact area towards the surrounding rock.

This contact area is often referred to as the flow-wetted surface (*FWS*) and is of great importance for solute retention, as a larger contact area will allow for more solutes to diffuse into and interact with the rock matrix. It should be noted that the flow-wetted surface traditionally only

includes the contact area between flowing water and rock (including rock matrix, fault gouge, etc) and that the contact area between stagnant fracture water and rock, as well as between flowing and stagnant fracture water, is excluded.

The concept of flow-wetted surface is not clear-cut from a radionuclide retention perspective. One fundamental issue concerns how one defines the contact area of a rough surface. This is illustrated in Figure 2-20 by the analogy of a curved line, where the same line is shown but with different resolutions in the y-axis. It appears that the first line (the red) could be well approximated by a straight line. However, if increasing the resolution of the y-axis, it becomes apparent that the straight line assumption is incorrect. The same applies for the contact areas in crystalline fractured rock. The larger resolution, the larger the contact area becomes. This is a particular conceptual problem if we consider fracture surfaces to be fractal, where the surface area is dependent upon the length scale of measurement.

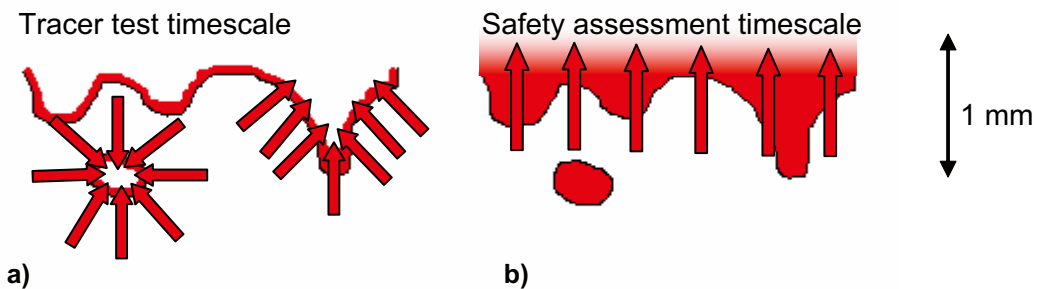
One way of dealing with surface roughness, often used in solute transport modelling, is to assume that the mineral surface facing the flowing fracture water becomes instantaneously equilibrated. In fact the concept can also be applied to a thin layer at and beyond the surface, including the zone of small-scale surface roughness. The actual thickness of this zone is determined by what equilibration time can be approximated as instantaneous, comparing to the timescale of the tracer test or transport modelling. As a result of such a simplification, small-scale roughness can be smoothed and does not need to be considered when assessing the flow-wetted surface. However, surface roughness on a larger scale will affect the flow-wetted surface through which a diffusive flux into the immobile zone can occur.

This is illustrated in Figure 2-21, showing a rock matrix surface with the maximum roughness on the millimetre scale and a small particle of fault gouge. In Figure 2-21a, the thin layer where instantaneous equilibration can be assumed on a tracer test timescale is marked in red. The red arrows symbolise the diffusive flux that can occur over the flow-wetted surface after instantaneous equilibration during the relatively short termed tracer test.

On a Safety Assessment timescale, however, even for sorbing species penetration may occur over much larger distances and the small-scale roughness and fracture fillings will soon become fully saturated by the tracer. When this has happened, the small-scale curvatures of surface imperfections are of little importance, and the diffusive flux can effectively only occur normal to the fracture plane, which is shown in Figure 2-21b. Thus, when assessing the flow-wetted surface for Safety Assessment it is more conservative to assume smooth fracture surfaces and no fracture fillings.



**Figure 2-20.** The effect of surface roughness on contact area.



**Figure 2-21.** The effect of surface roughness on retention at different timescales.

In this subsection it is argued that the apparent numerical value of the flow-wetted surface changes with the timescale of the experiment, giving the flow-wetted surface a transient characteristic in data evaluation. Due to this behavior, extrapolation of the flow-wetted surface estimated from tracer tests to Safety Assessment spatial and timescales may be problematic.

It should be noted that transient nature of the apparent flow-wetted surface described above is not consistent with how the entity is normally described by hydrogeologists. Unless the flow-field changes, the flow-wetted surface is considered constant in a hydrogeological perspective, where focus is generally upon the transport of groundwater itself rather than the solutes the groundwater contains. When discussing the flow-wetted surface, it is wise to recognise the different conceptualisations of the entity by the fields of hydrogeology and solute transport.

From an SKB perspective, the flow-wetted surface presently delivered from hydrogeological modelling, as an input to radionuclide transport modelling in Site Descriptive Modelling and Safety Assessment, is based on the assumption of planar and smooth surfaces, as defined in the Hydro-DFN (Hydrogeological Discrete Fracture Network) modelling. To compensate for channelling effects, in the Safety Assessment SR-Can the F-factor (that directly relates to the flow-wetted surface) was reduced by a factor of 10. Although thought to be a conservative approach in SR-Can, this premise must be properly tested as the existence of channelling can also impact the sparsity of flowing features modelled in the Hydro-DFN, given that it is constructed by conditioning to borehole hydraulic data where channels of very limited extent are not easily identified.



### 3 SKB field tracer tests: 1977–2007

In this chapter we aim to summarise and briefly describe the in situ tracer tests performed by SKB in Sweden from the late 1970's to the present time. Furthermore, references to more detailed descriptions of these tracer tests are given for further reading. The short descriptions serve as a platform for later discussions in Chapter 4 concerning what information can be extracted from tracer tests for use in Site Characterisation (SC) and Safety Assessment (SA). An important outcome of Chapter 3 is that we demonstrate that SKB has indeed performed a number of relevant large-scale tracer tests. It should be kept in mind that many early campaigns had equipment and methodology development as a primary aim and are perhaps of limited relevance today. Descriptions of tracer tests performed in the laboratory or in situ in other countries are omitted as this was outside the scope of this investigation.

Before presenting the SKB tracer tests, it is necessary to define what we mean by a tracer test, as our definition may be broader than that customarily assumed. Our definition of a tracer test is:

*“An investigation where tracer migration within a geological medium is monitored”*

As tracer tests can be categorised in many different ways, we simply divide them based on where they were performed and describe them in chronological order. The tracer tests discussed in this report have been performed at:

- Studsvik research centre area.
- Finnsjön test site.
- The Stripa mine.
- The Äspö Hard Rock Laboratory (Äspö HRL).
- The Forsmark or Oskarshamn sites within the site investigation programme.

The geology, geophysics, hydrology, groundwater chemistry, etc of these sites are well described in numerous of other SKB reports and site descriptions are omitted from this chapter (see e.g. /SKB 2005b, SKB 2006a/, or /Milnes 2002/ for a broad overview of relevant SKB reports).

#### 3.1 Distinguishing characteristics of tracer tests

Before describing the individual tracer tests, a few characteristics that distinguish them are discussed below. The distinguishing characteristics given in this report are environment, flow situation, monitoring and sampling, and tracer. In the list below, environment generally refers to whether the investigation is carried out in situ or in the laboratory. In the case of in situ investigations, they can be made from boreholes drilled from ground surface or from tunnels. Flow situation refers to whether the investigation is carried out in stagnant (immobile) water only or also in flowing (mobile) water. Monitoring and sampling refers to whether the monitoring and/or sampling are carried out within the mobile or immobile zone. Tracer refers to what kind of tracer that is used.

**Table 3-1. Distinguishing features of tracer tests.**

<b>Environment</b>	<b>Monitoring and sampling</b>
<ul style="list-style-type: none"><li>• In situ – Holes from ground surface</li><li>• In situ – Holes from tunnels</li><li>• Laboratory</li></ul>	<ul style="list-style-type: none"><li>• Mobile</li><li>• Immobile</li></ul>
<b>Flow situation</b>	<b>Tracers</b>
<ul style="list-style-type: none"><li>• Mobile/immobile</li><li>• Immobile</li></ul>	<ul style="list-style-type: none"><li>• Intentionally introduced</li><li>• Unintentionally introduced</li><li>• Natural</li><li>• Physical analogues</li></ul>

### 3.1.1 Environment

Tracer tests performed by SKB are either conducted in situ or in the laboratory. In situ tracer tests are generally conducted from boreholes drilled from ground surface or from tunnels. In the site investigations, all boreholes are drilled from ground surface and tracer tests performed in the vicinity of tunnels are not applicable. In the construction phase of the repository, however, tracer tests performed in the vicinity of tunnels will become possible. In supporting investigations, tracer tests have been performed utilising boreholes drilled from tunnels, such as at the Äspö HRL and the Stripa mine. Laboratory tracer tests are generally conducted on rock samples that are predominantly taken from drill cores.

Two different categories of tracer test can be performed using boreholes. The investigations could either be performed around a borehole section without hydraulically conductive fractures, or around a section featuring one or more conductive fractures. In the former case, matrix diffusion and interactions between tracers and rock matrix are studied. In the latter case, tracer migration in a flowing system is studied.

Tracer tests that are performed in the laboratory are of subordinate importance for this report. Information obtained from such investigations, however, can be very useful when interpreting data obtained from in situ tracer tests. Investigations carried out in the laboratory are often performed on intact drill core samples or on crushed rock. Examples of important complementary investigations, which are not included in our definition of tracer tests, are laboratory porosity measurements and batch sorption measurements. Information on laboratory measurements of retention parameters can be found elsewhere /e.g. Widestrand et al. 2003/.

### 3.1.2 Flow situation

In field tracer tests, the tracers could either be injected into flowing (mobile) or stagnant (immobile) water. The former refers to tracer tests carried out in hydraulically conductive fractures. In these tests, long-range transport occurs in the mobile zone while the short-range transport associated with retention occurs in the immobile zone. Here, we consider the immobile zone to be the stagnant pore water in the rock matrix as well as the mineral surfaces where sorption occurs. In a broader sense it is also taken to include stagnant zones within fractures where advective flow does not take place. The aim of such tracer tests is to investigate hydraulic and transport properties of the fracture or fracture system of interest with special focus upon fracture connectivity, water residence time, and tracer retention. Before a tracer test is initiated, investigations are often made to characterise the feature or features where the test is intended to be performed. This may be done, for example, by using pressure interference tests, flow logging, tracer dilution tests, and tracer tests of a preliminary design character. The characterisation data is used for selecting the geometry of the actual tracer test and also to give supporting information for data evaluation.

In inter-well tracer tests, also called cross-hole tracer tests, investigations are made using two or more boreholes that intersect the same conductive fracture or fracture system. Injection and withdrawal of tracers is performed in different boreholes. If the hydraulic gradient and transmissivity of the structures connecting the injection and pumping locations is too low, only a limited breakthrough may be achieved within a reasonable time period. This may occur if the boreholes are far apart, if the fracture connectivity is poor, if flowpaths are extremely tortuous, or if the fracture apertures are small. Even though experiments with limited breakthrough cannot be used to assess retention properties of the flowpath, the connection between injection and withdrawal points can be confirmed.

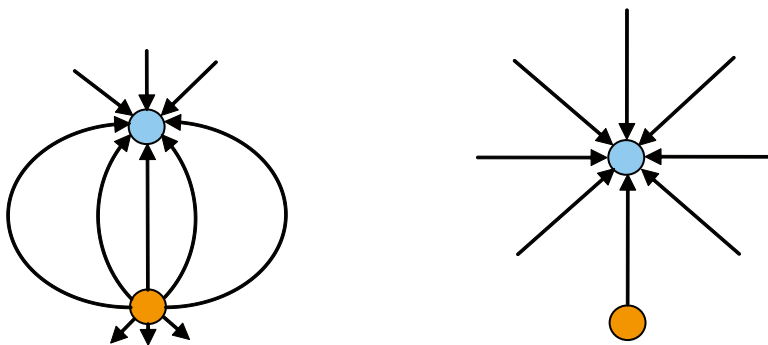
Inter-well tests can either be dipolar (potentially multipolar in case of more than two boreholes) or radially converging (see Figure 3-1). In a dipolar experiment, groundwater is actively pumped into the fracture system together with the tracer in the injection borehole. At the same time groundwater is withdrawn by pumping at another borehole. In this way a flow-field emitted from one borehole and withdrawn in another is created. This is shown in Figure 3-1 where the orange “borehole” is the injection hole and the blue the withdrawal hole.

In an equal dipolar tracer test, the pumping rates match reasonably well. In an unequal dipolar tracer test the pumping rates do not match and most often the injection rate is much lower than the withdrawal rate. If the injection rate is very much lower than the withdrawal rate, the configuration more resembles radially converging than unequal dipole and it is not clear-cut where to draw the line between the configurations, which is noted from differences in terminology used in different reports.

In a radially converging test, the tracer solution is generally added in a hydraulically conductive structure isolated by packers. In this case, no active injection of the tracer into the structure is performed. By pumping in the withdrawal borehole only, a radially converging flow field is created around the borehole. The tracer is carried with groundwater flowing towards the withdrawal borehole.

Tracer tests in hydraulically conductive fractures can also be performed from a single borehole. This has advantages when boreholes are too far apart to feasibly perform inter-well tracer tests, or if only one borehole is drilled. In such tracer tests, a borehole section is packed-off around a fracture or fracture zone and tracer-spiked groundwater is injected. After some time the hydraulic gradient is reversed and the tracer-spiked groundwater is withdrawn and analysed. This particular kind of tracer test is referred to within SKB as a Single Well Injection Withdrawal (SWIW) test /e.g. Nordqvist and Gustafsson 2002/, although it is also sometimes referred to in the literature as a push-pull test /e.g. Snodgrass and Kitandis 1998/.

By injecting tracers in stagnant water in contact with a rock surface, tracer migration into the rock matrix can be studied. In such investigations, typically a borehole section containing no hydraulically conductive fracture is packed-off. A tracer solution is introduced into the packed-off section and the tracer is allowed to diffuse into the rock matrix over a long period of time, spanning from months to years.



*Figure 3-1. Radially converging and dipole flow-fields.*

### 3.1.3 Monitoring and sampling

Monitoring and sampling may be performed either in the mobile or immobile zone. Monitoring and sampling in the mobile zone is generally accomplished by collecting withdrawn groundwater and analysing its tracer concentration. From this information, breakthrough (i.e. concentration vs. time) curves can be constructed. By comparing these with the corresponding tracer injection curves, various processes influencing the tracer transport in the mobile zone can be studied.

Monitoring and sampling in the immobile zone customarily includes overcoring and analyses of the overcored rock matrix. This may be achieved, for example, by leaching and analysis of pore fluid, extraction of sorbing tracers, or by direct measurement of tracer concentration on the solid phase. By thin-sectioning samples from the overcored rock or stepwise ablation of the sample surface, depth profiles of the tracer concentration can be obtained. If a tracer test is performed in a conductive fracture in the vicinity of the ground surface or a tunnel, monitoring and sampling in both the mobile and immobile zone may be possible and the combined information from such efforts can prove to be very useful.

### 3.1.4 Tracers

There are a number of tracers that can be used in field investigations. In most tracer tests the tracers are intentionally introduced by way of injection in a controlled fashion at the initiation of a tracer test. Within the SKB programme, non-sorbing (e.g. I<sup>-</sup>, Uranine, HTO), weakly sorbing (e.g. Sr<sup>2+</sup>) and moderately sorbing (e.g. Cs<sup>+</sup>, Co<sup>2+</sup>) species are commonly used in field experiments if breakthrough is expected. When using species more strongly sorbing than Cs<sup>+</sup>, a breakthrough is generally not expected. For radiological protection reasons, many of the radionuclide tracers used are relatively short-lived, which limits the possibility to perform long term tests. Within the SKB programme, tracers with a wide range of properties have been used, such as the dissolved gas <sup>3</sup>He, with high diffusivity, and the possibly sterically hindered macromolecule Dextran, with a low diffusivity.

Unintentionally introduced tracers have been released into the environment as a result of human activity over the last few decades and have penetrated into the bedrock. Examples of these are CFC's, HTO, and <sup>85</sup>Kr. Natural tracers are substances that exist naturally in the geochemical system and not necessarily at tracer concentrations. Examples are Cl<sup>-</sup> (also the isotope <sup>36</sup>Cl), oxygen isotopes (<sup>16</sup>O/<sup>17</sup>O), <sup>4</sup>He, radon and isotopes of the uranium disequilibrium series. Neither unintentionally introduced nor natural tracers are a focus of attention in this report, although we note that they also play an important role in site understanding and are used within the site investigations. When discussing physical analogues to tracers, the processes involved in the investigations may, or may not, fall within our definition of a tracer test. A physical analogue used within the SKB site investigation programme is the analogy between diffusivity and ionic mobility, which is used when obtaining the diffusive properties of the rock matrix by electrical methods.

## 3.2 Tracer tests at the Studsvik research centre area

### 3.2.1 Cross-hole tracer test 1 in Studsvik

The cross-hole tracer tests /Landström et al. 1978/ at the Studsvik research centre area, outside Nyköping, Sweden, were the first tracer tests carried out in a natural non-disturbed fracture system of crystalline rock in Sweden, at least within the research field of spent nuclear fuel management. The aim was both to study retention processes in the natural system and to develop suitable methodologies and instrumental technology for performing tracer tests. Eight boreholes were drilled from ground surface at the site and the boreholes were investigated by a number of geophysical and hydrological methods, in order to characterise the site and find

suitable flowpaths for the subsequent tracer tests. Two suitable flowpaths were found, where the main flowpath was between boreholes B8 and B2 and the secondary flowpath between boreholes B8 and B7 (see Figure 3-2).

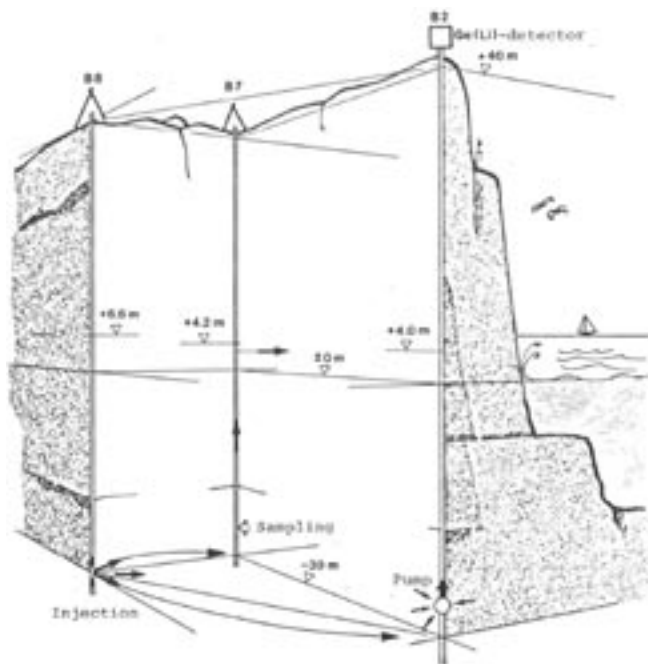
In the selected flowpaths, tracer tests with both non-sorbing and sorbing tracers were performed in different runs. Forced tracer injection was performed from a packed-off section in Borehole B8 and monitoring and sampling was made on groundwater withdrawn from the surrounding boreholes B7 and B2. The experiment was carried out in fairly shallow rock where injection was made at about 72 m depth below ground surface and 30 m depth below sea level. In preliminary tests, performed as a part of identifying the flowpaths, radioactive  $^{82}\text{Br}^-$  was used as the tracer. The two conductive paths had the lengths 22 m (B8–B7 injection-recovery well distance) and 51 m (B8–B2) with advective travel times on the order of some tens of hours for the applied hydraulic gradient.

These preliminary tests were followed up by a campaign of three separate tracer tests using instantaneous pulse injections of different sorbing and non-sorbing tracer cocktails including  $^{24}\text{Na}^+$ ,  $^{75}\text{SeO}_3^{2-}$ ,  $^{82}\text{Br}^-$ ,  $^{85}\text{Sr}^{2+}$ ,  $^{99\text{m}}\text{TcO}_4^-$ ,  $^{113}\text{Sn}^{4+}$ ,  $^{131}\text{Cs}^+$ ,  $^{131}\text{I}^-$ , and  $^{147}\text{Nd}^{3+}$ . Monitoring and sampling was carried out for a few hundred hours.

In addition to the tracer tests mentioned above, an attempt was made to inject bentonite into the fracture system around the injection hole and thereafter perform a cross-hole tracer test between boreholes B8 and B2. The result was that the injected bentonite was very effective in reducing the migration of  $^{82}\text{Br}^-$  and  $^{85}\text{Sr}^{2+}$  in the fracture system, as no tracer could be detected in the monitoring boreholes.

### **Major achievements**

As this tracer test campaign was the first performed in situ in Sweden within the research field, a major achievement was the development of methods for conducting the experiment, even if today one can consider details in the techniques used as somewhat crude. Much progress was also achieved in the development of Site Characterisation (SC) techniques which were adapted and refined during the course of this campaign.



**Figure 3-2.** Sketch of the Studsvik cross-hole tracer test 1. Image taken from /Landström et al. 1978/.

Concerning breakthroughs in the three main tracer tests, the breakthrough curves of different species were compared to the breakthrough curve of  $\text{Br}^-$ , which was assumed to be non-retarded (this was before retardation due to matrix diffusion was suggested). From the comparisons clear retention could be observed and the following conclusions were drawn /Landström et al. 1978/:

- $^{99\text{m}}\text{TcO}_4^-$  was assumed to be non-retarded.
- $^{131}\text{I}^-$  and  $^{24}\text{Na}^+$  had been slightly retarded.
- $^{85}\text{Sr}^{2+}$  had a retardation factor of about 6.
- $^{75}\text{SeO}_3^{2-}$  and  $^{113}\text{Sn}^{4+}$  were to different degrees affected by precipitation. However, it appears that fractions of the tracers were soluble and that tracers migrated at different rates in different chemical forms.
- $^{147}\text{Nd}^{3+}$  and  $^{131}\text{Cs}^+$  were thought to have high retardation factors, as they could not be detected during the 60 days of monitoring period.

### 3.2.2 Cross-hole tracer test 2 in Studsvik

A second cross-hole tracer test campaign /Landström et al. 1983/ was carried out at Studsvik, although at a different location to the first campaign. Eight percussion-drilled boreholes (B1N–B8N) and one core-drilled borehole (K1N) were drilled from ground surface at the site and the boreholes were investigated geophysically, hydrogeochemically and hydrologically. In addition, the drill-core of borehole KN1 was subjected to geological logging, where fissure minerals of water-bearing fractures were examined. Based on the Site Characterisation, borehole B1N was chosen for tracer withdrawal and the three surrounding boreholes, B5N, B6N, and B8N were chosen for tracer injection (see Figure 3-3).

Approximate water residence times of the chosen flowpaths were obtained by preliminary tracer tests using  $^{131}\text{I}^-$  as a non-sorbing tracer (assumed to be non-retarded). Based on the information obtained in these scoping tests, the flowpath between boreholes B8N–B6N was rejected for subsequent tracer tests on the basis of poor hydraulic connection.

In the main campaign the tracers  $^{85}\text{Sr}^{2+}$  and  $^{134}\text{Cs}^+$ , as well as the non-sorbing reference tracers HTO and  $^{131}\text{I}^-$ , were used in two separate dipolar tracer tests. The injection occurred from packed-off sections between the borehole length 91–92 m in B1N and 78.8–80.1 m in B5N. The distances between injection and withdrawal locations were estimated to be 11.8 m for boreholes B1N–B6N and 14.6 m for boreholes B5N–B6N.

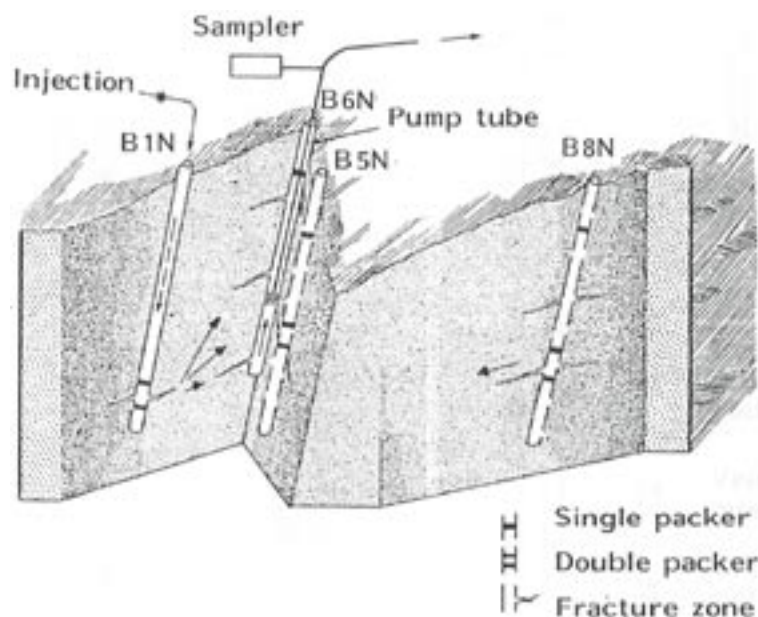


Figure 3-3. Sketch of the Studsvik cross-hole tracer test 2. Image taken from /Landström et al. 1983/.

Before and after injection of the tracer cocktail pulse, non-traced groundwater was injected continually in the flowpath to obtain a steady state flow-field. In the first tracer test, between boreholes B5N–B6N, all tracers were used but in the subsequent tracer test, between boreholes B1N–B6N,  $^{134}\text{Cs}^+$  was excluded. By pumping in borehole B6N, monitoring and sampling could be carried out on withdrawn groundwater for almost 7,000 hours.

### **Major achievements**

As in the first Studsvik tracer test campaign, much new knowledge was acquired concerning methodological and technological issues during both the Site Characterisation phase and the tracer tests. As one borehole was core-drilled and the drill core could be examined, an insight into the mineralogy of fracture infillings was gained. For the investigated fracture surfaces chlorite, clay minerals, and calcite dominated. Generally the mineral layers were less than 1 mm thick.

Concerning the tracer test results, clear indications of retention could be inferred in the test results by comparing the breakthrough curves of the non-sorbing and sorbing tracers. The following conclusions were drawn in /Landström et al. 1983/:

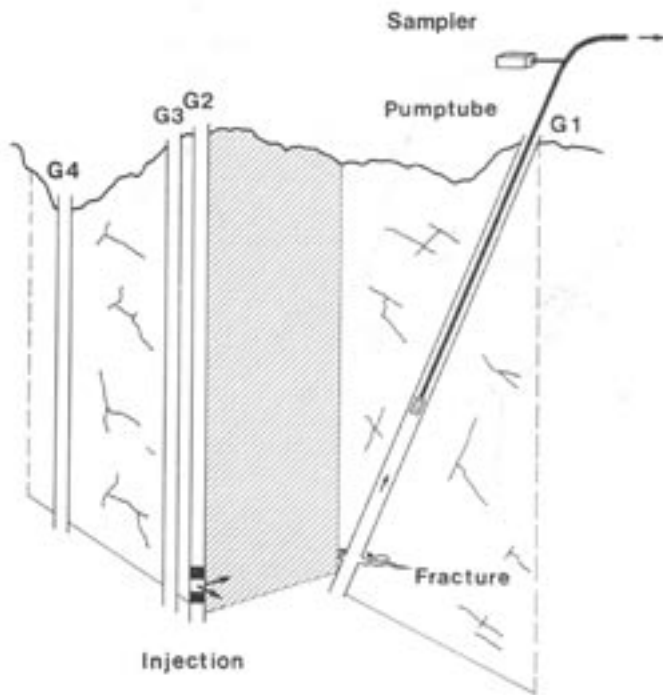
- In both tracer tests, the HTO breakthrough curve was almost identical to that of  $^{131}\text{I}^-$ .
- The majority of the  $^{85}\text{Sr}^{2+}$  in the flowpath B1N–B6N had a retardation factor of 17, compared to the non-sorbing tracers.
- The majority of the  $^{85}\text{Sr}^{2+}$  in the flowpath B5N–B6N had a retardation factor of 30, compared to the non-sorbing tracers.
- In both tracer tests a small fraction of  $^{85}\text{Sr}^{2+}$  remained practically non-retarded, compared to the non-sorbing tracers. An explanation offered was that some of the tracer was bound to mobile particles and organic complexes, e.g. Fe(III) precipitation in colloidal form and humic-/fulvic-acid complexes, carried by the flowing groundwater.
- After 5,000 hours of monitoring, no  $^{134}\text{Cs}^+$  had been detected in borehole BN6, which suggested a retardation factor of over 1,000. This was in line with results delivered from the parallel laboratory programme, investigating the retention properties of the tracers.
- No fast arrival of  $^{134}\text{Cs}^+$  was obtained, which suggested that  $\text{Cs}^+$  did not interact (to the same extent) with particles and organic complexes as  $\text{Sr}^{2+}$  was inferred to have done.

## **3.3 Tracer tests at Finnsjön test area**

### **3.3.1 Cross-hole tracer test in Finnsjön**

At the Finnsjön site, in northern Uppland County, Sweden, a series of cross-hole tracer tests were made in a minor fracture zone /Gustafsson and Klockars 1981, 1984/. At the site, ten approximately 100 m long percussion-drilled boreholes and two 460 and 730 m long core-drilled boreholes were drilled. Prior to the tracer test campaign the site had been previously characterised /e.g. Almén et al. 1978/. Therefore, the campaign only comprised two phases in the field; hydraulic testing and tracer tests.

In the hydraulic testing phase the boreholes were logged by water injection tests. Also pressure interference testing was performed by pumping in borehole G1 (see Figure 3-4) and recording the water pressure changes in the other boreholes. Based on the hydraulic testing a hydraulically conductive structure was chosen for the subsequent tracer tests. For tracer injection, the section 91–93 m in borehole G2 was chosen while for tracer withdrawal, the fracture zone at 100–102 m in borehole G1 was found to be appropriate. The geometric distance between injection and withdrawal points was around 30 m.



**Figure 3-4.** Sketch of the Finnsjön cross-hole tracer test. Image taken from /Gustafsson and Klockars 1981/.

The tracer tests were carried out and reported in two different parts of the campaign. In the first part the non-sorbing or very weakly sorbing tracers Rhodamine WT,  $\text{NO}_3^-$ ,  $\text{I}^-$ ,  $\text{Br}^-$ , and  $\text{Cr-EDTA}^-$  were used in four radially converging tracer tests /Gustafsson and Klockars 1981/. In the second part the sorbing tracers  $\text{Sr}^{2+}$  and  $\text{Cs}^+$  were used together with  $\text{I}^-$  in two radially converging tracer tests /Gustafsson and Klockars 1984/. All tracer tests were carried out in the same flowpath.

In the first three tests, an instantaneous tracer injection was performed. In the tests the tracer pairs Rhodamine WT/ $\text{NO}_3^-$ , Rhodamine WT/ $\text{I}^-$ , and  $\text{Br}^-/\text{NO}_3^-$  were used. Immediately prior to injection, groundwater was pumped from the packed-off section in borehole G2 to ground surface. At ground surface, the tracers were injected in a fraction of this groundwater and pumped back down the borehole. Thereafter, the rest of the groundwater brought to the surface was reinjected in the borehole.

In the fourth tracer test, the tracer pair  $\text{Cr-EDTA}/\text{I}^-$  was used and continuous injection was performed. In this injection, groundwater was injected into the packed-off section for a few days prior to tracer injection to achieve a steady state flow situation. Thereafter the tracers were injected for a long period of time. Continuous injection was also used for the second part of the campaign where the tracer pairs  $\text{Sr}^{2+}/\text{I}^-$  and  $\text{Cs}^+/\text{I}^-$  were used in two different runs.

Monitoring and sampling was carried out for 100–200 hours in the first part and for about 1,000 hours in the second part of the campaign.

### **Major achievements**

In the first part of the campaign, it was found that  $\text{NO}_3^-$ ,  $\text{I}^-$ , and  $\text{Cr-EDTA}$  had very similar transport properties as  $\text{Br}^-$ , which at the time was assumed to be non-retarded. Rhodamine WT had very similar peak arrival time and shape of the breakthrough curve as  $\text{NO}_3^-$  and  $\text{I}^-$ , but the recovery was somewhat less. It was interpreted as Rhodamine WT exhibits weak retardation and sorption /Gustafsson and Klockars 1981/.



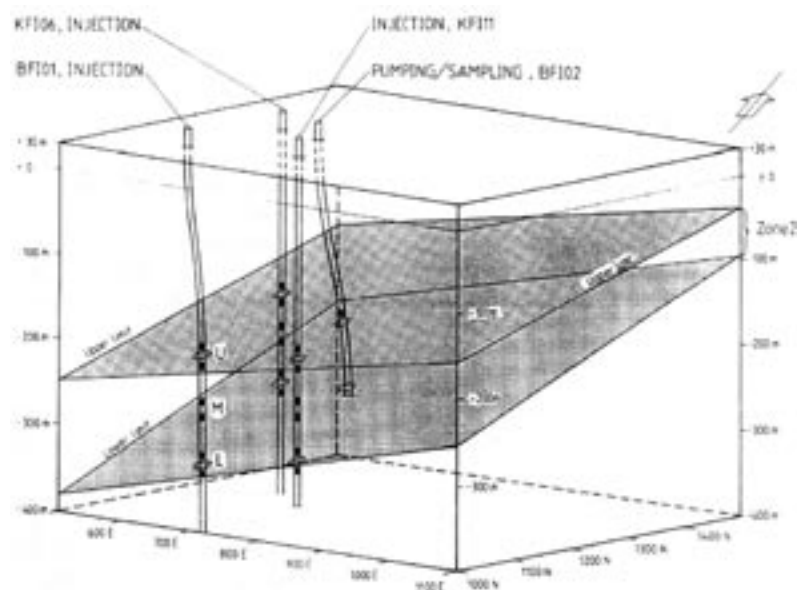
In the second part of the campaign some retardation of  $\text{Sr}^{2+}$  and  $\text{Cs}^+$ , as compared to  $\text{I}^-$ , was seen. However, the retardation factors were much smaller than in the Studsvik tracer tests described in Section 3.2. For  $\text{Sr}^{2+}$  and  $\text{Cs}^+$  the retardation factors 1.15 and 2.50 were obtained in Finnsjön, respectively. This should be compared with the Studsvik retardation factors between 6 and 30 for  $\text{Sr}^{2+}$  and over 1,000 for  $\text{Cs}^+$ . A multimodal breakthrough peak for both  $\text{I}^-$  and  $\text{Sr}^{2+}$  suggests at least two independent transport pathways, although the non-ideal tracer injection made interpretation of breakthrough curves difficult /Moreno et al. 1983/. Owing to the high concentration of tracer used in the experiment, the sorption of  $\text{Sr}^{2+}$  was non-linear thereby further complicating the data interpretation. One explanation of the observed weak retardation offered in /Gustafsson and Klockars 1984/ is that the most prominent fracture mineral in the area was calcite, a mineral on which the investigated species sorb poorly.

### 3.3.2 The Fracture Zone Project, Phase 3

At a different location of the Finnsjön test area, tracer tests were performed in the gently dipping fracture zone, designated as Zone 2. At the site all boreholes were drilled from ground surface. The campaign was divided into three phases, where hydraulic testing and tracer tests were performed in Phase 3 /Andersson et al. 1989a, 1993, Gustafsson and Nordqvist 1993/. In addition, a new borehole (BFI02) used as pumping borehole in subsequent tracer tests was drilled as a part of Phase 3. In the preceding phases, Site Characterisation was performed (references within /Andersson et al. 1989a/).

Prior to the tracer tests of Phase 3, a long-range qualitative cross-hole tracer test was performed during the drilling of borehole KFI11 /Gustafsson and Andersson 1991/. In this test the flushing fluid, used to cool the drill bit and remove drill cuttings, was spiked with Uranine. By pumping in the borehole HFI01, at a distance of 440 m away from KFI11, Uranine could be recovered and thereby the existence of a hydraulic connection could be established.

In the initial part of Phase 3, hydraulic interference tests were made in boreholes intersecting Zone 2. Pumping was performed in borehole BFI02 in different packed-off sections and the hydraulic responses in a large number of packed-off sections in surrounding observation boreholes, many of them hundreds of meters away, were recorded /Andersson et al. 1989a/. Based on the available information, boreholes and sections were chosen for a preliminary tracer test of radially converging configuration. The borehole chosen for pumping was BFI02 and those chosen for injection were BFI01, KFI06, and KFI11 /Andersson et al. 1989a/. Figure 3-5 shows Zone 2 and the four boreholes intersecting the zone.



**Figure 3-5.** Sketch of the Phase 3 tracer test set-up in Finnsjön. Image taken from /Gustafsson and Nordqvist 1993/.

In each injection borehole, a section of some tens of meters in length was packed-off and from each section a separate tracer was released in a short pulse. The non-sorbing tracers Uranine, iodide, and Amino-G acid were used in the injections. The intended flowpaths corresponded to the upper and highly conductive part of Zone 2 at about 200 m depth.

From the preliminary testing, water residence times from around 10 hours to 100 hours were obtained and from the shape of the breakthrough curves it was suggested that transport in the upper part of Zone 2 occurred in a number of fractures. Monitoring of the water withdrawn in the pumping borehole was carried on for roughly 400 hours and sufficient recovery was obtained to proceed with the planned, subsequent tracer tests.

In a second radially converging tracer test /Gustafsson and Nordqvist 1993/, the same injection/withdrawal borehole configuration as shown in Figure 3-5 was used. The tracer test was carried out at a depth of about 200 to 350 m. In this campaign, three short sections in the upper, middle, and lower part of Zone 2 were packed-off in each injection borehole. These sections were chosen on the basis of tracer dilution tests performed prior to the cross-hole test and the length of the packed-off sections varied between 3 and 19 m.

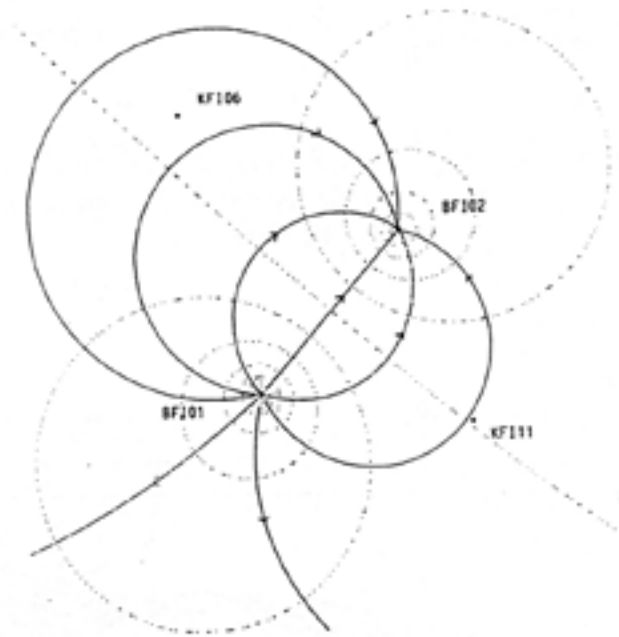
In total, 11 different non-sorbing tracers were injected from the nine packed-off injection sections. In each section, one unique tracer was injected apart from one section where three tracers were injected. The non-sorbing or very weakly sorbing tracers used were: In, Dy, Ho, Er, Tm, and Yb as M-EDTA<sup>-</sup> complexes, Gd as an M-DTPA<sup>-</sup> complex, I<sup>-</sup>, ReO<sub>4</sub><sup>-</sup>, Uranine, Amino-G acid, and Rhodamine WT. The tracers were injected continually in most cases although two pulse injections were also made. Pumping in borehole BFI02 was performed over the whole thickness of Zone 2 and monitoring and sampling was carried on for 4,510 hours. Distances between injection and withdrawal ranged from 155–200 m. First tracer arrival ranged between 24 and 3,200 hours where the most rapid first tracer arrivals were found in the upper and most conductive part of Zone 2. Recoveries varied from less than a percent to almost complete recovery.

The dipole experiment /Andersson et al. 1993/ was carried out in the same boreholes as the radially converging tracer tests. Groundwater was circulated between boreholes BFI02 and BFI01 by withdrawing water from BFI02 and re-injecting the withdrawn water into BFI01. Only the upper and most conductive part of Zone 2 was utilised. Thus, only one packed-off section was used in each borehole. The peripheral boreholes KFI06 and KFI11 were utilised for flow field measurements and KFI11 was also used for some of the tracer injections. An idealised layout showing the flow field of the experiment is shown in Figure 3-6.

A total of 15 non-sorbing or weakly sorbing tracers (short-lived when radioactive) were injected in the borehole BFI01, although not all simultaneously. Instead, a sequence of 14 injections was performed. In each injection a single tracer or a cocktail of tracers were injected for up to about 200 hours. In addition, one short injection was performed in the observation borehole KFI11.

The tracers used were: <sup>82</sup>Br<sup>-</sup>, <sup>131</sup>I<sup>-</sup>, <sup>58</sup>Co<sup>2+</sup>, <sup>24</sup>Na<sup>+</sup>, <sup>86</sup>Rb<sup>+</sup>, and <sup>201</sup>Tl<sup>+</sup> as freely solved species, <sup>51</sup>Cr, <sup>111</sup>In, In, <sup>160</sup>Tb, <sup>169</sup>Yb, <sup>58</sup>Co, and Tm(III) as M-EDTA<sup>-</sup> complexes, <sup>140</sup>La, <sup>177</sup>Lu, and Gd(III) as M-DTPA<sup>-</sup> complexes, <sup>99m</sup>TcO<sub>4</sub><sup>-</sup>, <sup>186</sup>ReO<sub>4</sub><sup>-</sup>, the organic dye Rhodamine WT, and the macro molecule Blue Dextran 2000. The distance between BFI01 and BFI02 was 168 m. The time of pumping was 1,486 hours but as the water was circulated and injections were performed in a sequence, the effective time of the tracer tests was less or even considerably less in case of quick tracer arrival.

Tracer recovery was obtained for the non-sorbing or weakly sorbing solutes but not for the more sorbing solutes, <sup>58</sup>Co<sup>2+</sup>, <sup>86</sup>Rb<sup>+</sup>, and <sup>201</sup>Tl<sup>+</sup>. In addition, <sup>99m</sup>TcO<sub>4</sub><sup>-</sup> was not recovered (a possible mechanism for this null result was suggested that involved the reduction and immobilisation of pertechnate).



**Figure 3-6.** Idealised flow-field in dipole tracer test. Image taken from /Andersson et al. 1993/.

### **Major achievements**

A major achievement of the Fracture Zone project was that a major fracture zone and its surrounding rock mass was characterised. Based on the Site Characterisation both a fracture zone model and a rock domain model of the surrounding rock were produced. Furthermore, the geological information obtained together with the information from the hydrological programme increased the understanding of the complexity of a major fracture zone. Together with the tracer tests, process understanding of flow in fractured rock was improved both on the smaller and larger scale.

As for the previously performed tracer tests, the Phase 3 tracer tests in Finnsjön also had as an objective to develop techniques and methods. This objective was met as the tracer tests in general were performed successfully.

In the second radially converging tracer test /Gustafsson and Nordqvist 1993/, specific flow wetted surfaces ranging from 1–92 m<sup>2</sup>/m<sup>3</sup> (based on the rock volume) were suggested for explaining the breakthrough curves. It is not within the scope of this present report to review these figures but what should be noted is the great range in flow-wetted surface suggested for different flowpaths within the same fracture zone. Furthermore, in both radially converging tracer test campaigns /Andersson et al. 1989a, Gustafsson and Nordqvist 1993/, clear evidence of anisotropic flow conditions were seen.

In the dipole tracer tests, the breakthroughs in the pumping hole were consistent with predictions. However, breakthroughs in the peripheral boreholes deviated significantly from those expected, indicating that the idealised flow-field shown in Figure 3-6 did not accurately describe the true flow-field. From the dipole tracer test /Andersson et al. 1993/ an anisotropy factor of 8 was suggested.

It was also concluded from the dipole tracer test that pertechnetate (TcO<sub>4</sub><sup>-</sup>) was reduced in situ to immobile Tc(IV) /Andersson et al. 1993/.

### 3.4 Tracer tests in Stripa mine

The Stripa project was an international co-operation between a number of organisations under the auspices of the OECD/NEA, initiated in 1980 and finishing in 1992. The investigations were carried out in the Stripa mine tunnel, in Västmanland County, Sweden (see Figure 3-7).

Amongst the many objectives of this project were the development of instruments and procedures to characterise candidate repository sites, and to increase the understanding and modelling of groundwater flow and solute transport in crystalline rock. Although described briefly in the following sections, for a summary of the entire project in the natural barrier, the OECD/NEA overview is recommended /Fairhurst et al. 1993, Gnirk 1993/.

As the tracer tests carried out at the Stripa mine were very early in the timeline of tracer test development, there were certainly a great number of achievements concerning methodological and technological issues that are not accounted for in detail in this present report.

#### 3.4.1 Small-scale tracer tests at the SGAB tracer test drift

A small-scale tracer test was performed at the at 360 m level at the SGAB tracer test drift /Andersson and Klockars 1985/. The objective was to obtain information on flow field heterogeneity and anisotropy in a well-defined rock mass of low hydraulic conductivity.

The tracer test was carried out from an injection hole that was surrounded by eight detection holes in a cylindrical configuration at a distance of 1.5 m from the central hole. All boreholes were parallel, about 20 m deep, and drilled into low conductive rock (see Figure 3-8).

The tracer test was preceded by hydraulic characterisation of the boreholes by using both single-hole and cross-hole tests. The lower 9 m of the injection hole was packed-off and used for continuous injection of Uranine. Tracer collection was performed in the lower 9 m of the detection holes, which were packed-off into two equally sized sections. In the injection, a substantial excess pressure was used and therefore, prior to the injection a steady state flow field had been established by injecting groundwater containing no tracer. Monitoring and sampling of the Uranine concentration in the 16 sections of the detection holes was carried on for up to 1,700 hours.

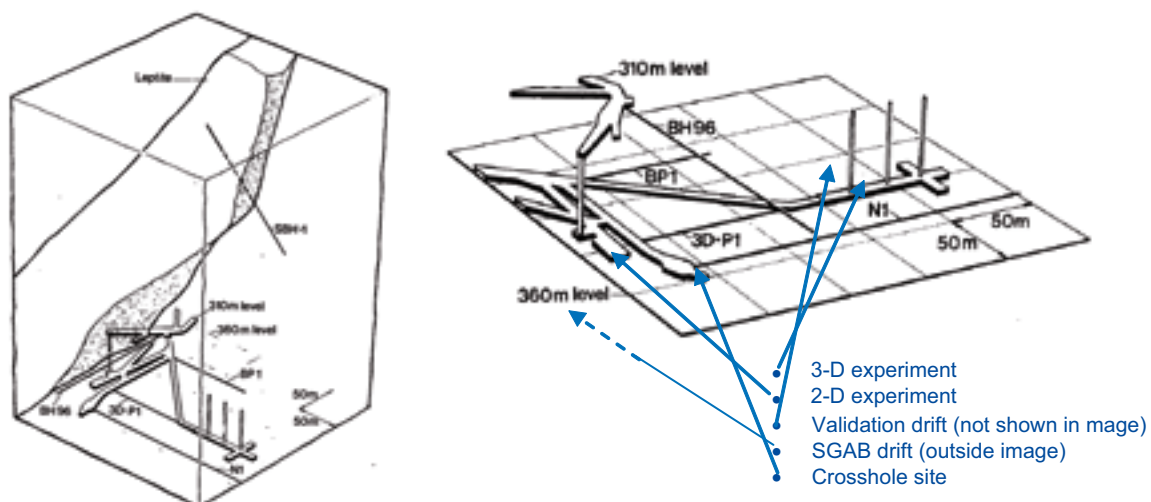
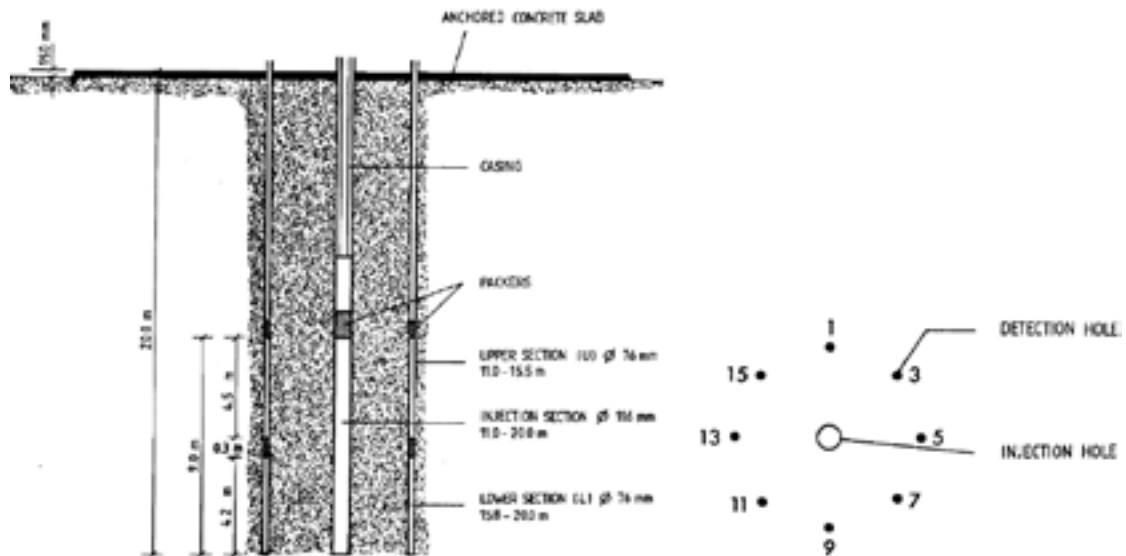


Figure 3-7. Part of the Stripa mine. Images taken and modified from /Abelin and Birgersson 1987/.



**Figure 3-8.** Configuration of the boreholes in the tracer test at the SGAB drift. Images taken from /Andersson and Klockars 1985/.

### Major achievements

In this tracer test the heterogeneity and anisotropy of the flow field and transport properties of a rock block were investigated. It was shown that the flow field in the well-defined and low conductive rock volume was very heterogeneous. Only 3% of the fractures intersecting the volume were found to be hydraulically conductive and the study suggested both channelling and anisotropy effects. It was concluded from this investigation that the rock mass could not be modelled as a homogenous porous medium.

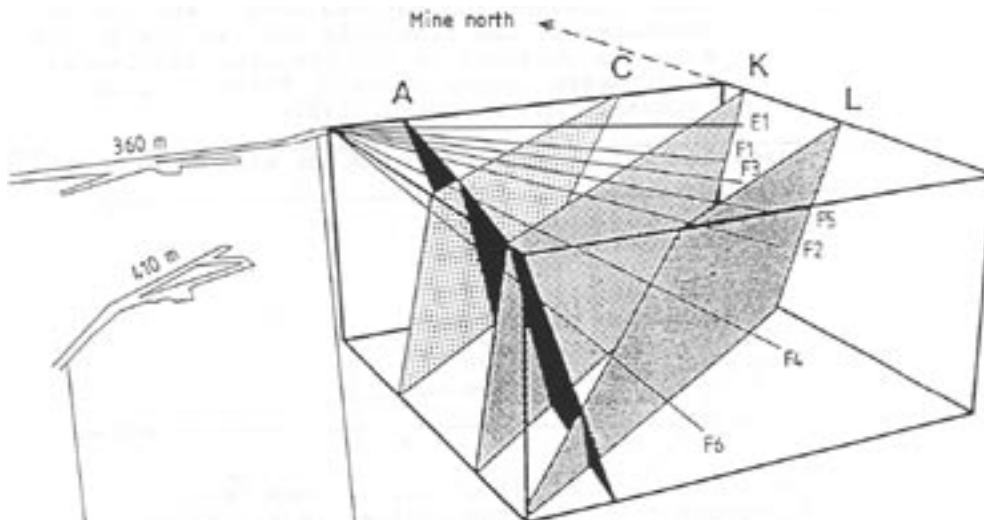
Hydraulic single-hole tests in 1 m long sections indicated hydraulic conductivities of the rock mass in the range of  $1 \cdot 10^{-9}$ – $7 \cdot 10^{-12}$  m/s. Flow porosities were estimated to be between  $6.7 \cdot 10^{-5}$  to  $3.8 \cdot 10^{-4}$ .

### 3.4.2 Monitoring of saline tracer transport with borehole radar

Two interesting “tracer tests” were performed at the 360 m level in the Stripa mine at the Crosshole site /Andersson et al. 1989b/ and at the Validation drift /Olsson et al. 1991/. The Crosshole site is shown in Figure 3-9 and Validation drift in Figure 3-18. These tests were similar to each other and the one performed at the Crosshole site is briefly described below.

At the Crosshole site, 7 boreholes between 200 m and 300 m long fan out and intersect a number of fracture zones labelled A, C, K, and L in Figure 3-9. The test was focused on zone C and the general objectives were to characterise the zone by borehole radar measurements, then inject a saline solution into the zone from borehole F3 and thereafter to re-measure with borehole radar. Radar characterisations were made both by cross-hole difference tomography and single-hole reflection. By interpreting the radar measurements one can, in theory, detect the distribution of saline and non-saline groundwater in the fracture system. The prerequisite is that the contrast between the responses from the different waters is high enough that it can be measured over the background noise.

The tracer solution of 0.5% KBr was injected in large quantities for 905 hours with an injection flow rate of 800 ml/min. To ensure that the second round of radar measurements was performed at steady state, the breakthrough of the saline front was measured in all other boreholes by means of electrical conductivity measurements. This information was also used to calibrate the radar measurements.



*Figure 3-9. The Crosshole site in the Stripa mine. Image taken from /Andersson et al. 1989b/.*

The investigation at the Validation drift was similar to that at the Crosshole site although at the end of the saline injection, a small amount of Amino-G acid was injected as a pulse to increase the resolution of the late arrival breakthrough curves.

### **Major achievements**

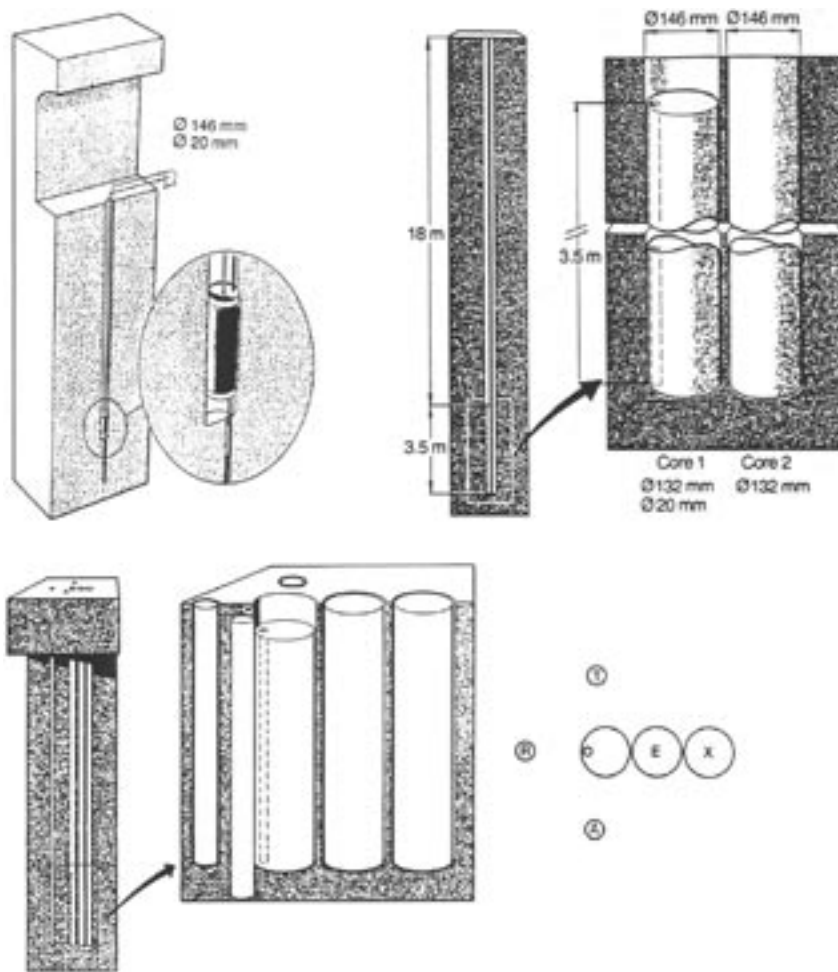
These tests can be seen as an innovative combination of a traditional tracer test and a geophysical analogue, where a saline tracer was injected in a flowing field and its spreading was monitored by geophysical tools. The resolution of the measurements was good enough to at least get the broader picture of the flow geometry in the investigated rock volume.

### **3.4.3 In situ diffusion experiment**

The in situ diffusion experiment was performed in the undisturbed rock matrix away from hydraulically conductive fractures. The aim of the study was to address the question of long-range matrix diffusion in the undisturbed rock matrix under native stress conditions /Birgersson and Neretnieks 1990, Birgersson et al. 1992a/.

The campaign comprised three similar tracer tests, referred to as Part I, II, and III, that were mainly differentiated by the experimental time frame. All tests were performed at different, but closely situated, locations at the 360 m level of the Stripa mine. In each tracer test a large diameter borehole ( $\varnothing$  146 mm) was drilled from the Stripa mine tunnel to a distance where the stress situation in the rock was expected to be not influenced by the presence of the tunnel. From the end of the large diameter borehole, a small diameter borehole ( $\varnothing$  20 mm) a few metres in length was drilled (see Figure 3-10, upper left). The small diameter borehole was packed-off and used as a tracer reservoir. In the reservoir, a slight overpressure was applied (0.5–0.9 MPa). The non-sorbing tracers used in all three parts were I<sup>-</sup>, Uranine, and Cr-EDTA.

In Part I, the tracers were allowed to diffuse into the rock matrix for three months. The small borehole was then overcored by extending the large diameter borehole. The overcored rock material was cut and sectioned into a large number of small samples (5 cm in length,  $\varnothing$  1 cm) that were leached in the laboratory. When evaluating the pore water tracer concentrations, a constant porosity was assumed. In this way a local concentration profile of the tracers could be obtained in the overcored rock matrix.



**Figure 3-10.** Sketches of the in situ diffusion experiment. Images taken from /Birgersson et al. 1992a/.

In Part II, the tracers were allowed to diffuse into the rock matrix for six months. In this case, in addition to overcoring by extending the large diameter borehole, another adjacent large diameter borehole was drilled (see Figure 3-10, upper right). Rock samples were prepared and leached as in Part I, with the exception that the porosity of each sample was measured. In this way, tracer concentration profiles extending 20–30 cm from the small diameter borehole could be obtained.

In Part III, the tracers were allowed to diffuse into the rock matrix for 3.5 years. After termination of the experiment, the rock of interest was overcored by a number of boreholes (see Figure 3-10 lower). Rock samples were prepared and leached as in Part II. In this way concentration profiles extending up to 40 cm from the small diameter borehole could be obtained. It was assumed that the stress situation of the rock matrix at a distance of 40 cm from the small diameter borehole was unaffected by the presence of the borehole /Birgersson and Neretnieks 1990/.

### **Major achievements**

In this campaign, diffusion in the microporous system of the undisturbed rock matrix was studied. The rock volume investigated held no hydraulically conductive fractures and was located far enough away from the nearest tunnel so that it was not affected by stress release. Furthermore the borehole from where the tracers were released had a small diameter, to reduce the surrounding zone of stress released rock.

It was shown that the microporous system was well connected over the investigated rock volume. This was a very important result as, at the time, it was debated whether or not tracers could penetrate more than a few centimetres into the undisturbed rock matrix. However, at the same time the transport rate through the microporous system appeared to be very heterogenous. At some locations, tracers could be found at high concentrations at the outer part of the overcored rock volume, indicating penetration well beyond the distance of 40 cm from tracer injection. At other locations, the same tracers had only penetrated a couple of decimetres into the rock matrix. This is shown in Figure 3-11 where tracer concentration profiles of Part III, at the same borehole depth but different directions, are shown.

It should be noted that when fitting the breakthrough curves, the matrix diffusivities used corresponded well to those obtained in laboratory measurements.

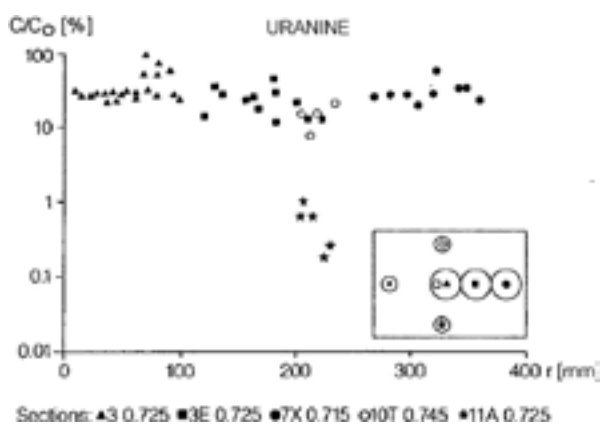
### 3.4.4 Migration in a single fracture, the 2D experiment

In the Migration in a single fracture experiment /Abelin et al. 1985, Birgersson et al. 1992a/, tracer migration in two interpreted single fractures was studied. The experiment was performed at the 360 m level of the Stripa mine where two fractures intersect the tunnel (see Figure 3-12 below).

The aim of the study was to investigate to what extent laboratory results could be applied to a real environment with migration distances up to 10 m. Furthermore, an attempt was also made to determine the extent of channelling within fractures.

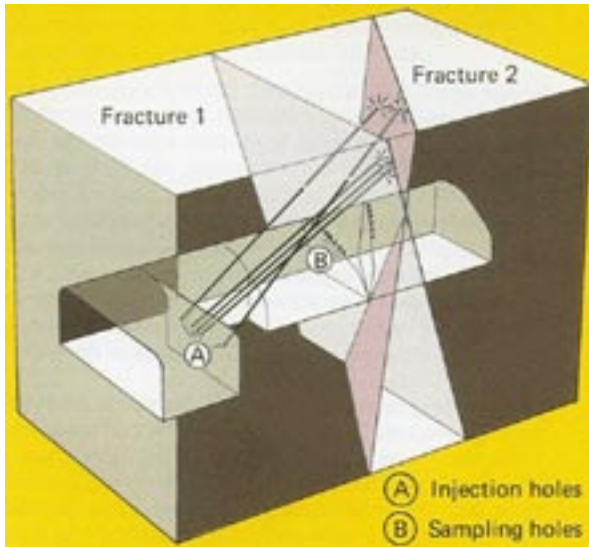
Five injection boreholes were drilled from the tunnel to intersect both fracture planes, wherein tracers were intended to migrate. As a result of the presence of the tunnel, the water flow in the fractures was directed towards the tunnel. In order to collect inflowing water in the tunnel, that should carry dissolved tracers, short sampling boreholes were drilled from the roof of the tunnel where it was intersected by the fractures. In total 13 short boreholes were drilled into Fracture 1 and 15 into Fracture 2. In addition, 3 sampling holes were drilled into the rock away from the fractures. The multitude of sampling boreholes was intended to allow the recovery of tracer to be spatially resolved.

From packed-off sections in four of the injection boreholes, tracers were injected into Fracture A. This was done using a very slight overpressure of 10% above the natural pressure at the injection point. The non-radioactive sorbing species  $Cs^+$ ,  $Sr^{2+}$ ,  $Eu(III)$ ,  $Nd(III)$ ,  $Th(IV)$ , and  $U(IV/VI)$  were continuously injected from borehole H2 at a constant concentration for more than half a year. In addition the non-sorbing tracers Cr-EDTA, iodide, Bromothymol blue, Elbenyl, Eosin Y, and Uranine were injected in different boreholes at different times in shorter tracer runs /Abelin and Gidlund 1985/. Finally coloured resin was injected from borehole H2 to, upon excavation, facilitate the identification of exactly which fractures had been active in the



**Figure 3-11.** Tracer concentrations in samples taken from overcored rock. Images taken from /Birgersson et al. 1992a/.



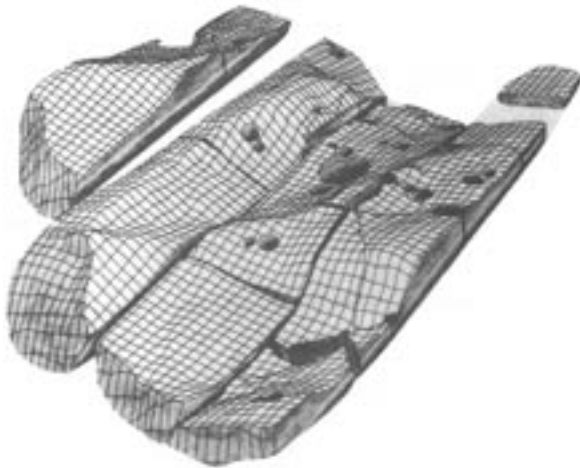


**Figure 3-12.** Sketch of the two fractures intersecting the tunnel, the injection and sampling holes. Images taken from /SKB 1989/.

injection. Both the water and tracer flows were monitored in the sampling holes and the tracer was recovered for up to 3,000 hours after injection.

The sorbing species were not expected to reach the sampling holes at the tunnel within the experimental timeframe, with the possible exception of  $\text{Sr}^{2+}$ . Therefore, after termination of the monitoring and sampling in the sampling boreholes, parts of the fracture surfaces close to the injection hole H2 were excavated by drilling parallel to Fracture 2. Figure 3-13 shows a representation of the fracture surface of Fracture 2 excavated from around the injection hole H2.

As can be seen, four adjacent excavation drill cores ( $\varnothing$  200 mm) were used to obtain the about 0.5 m<sup>2</sup> large fracture surface shown in Figure 3-13. In the figure, the injection hole H2 can be seen as the larger one in the centre surrounded by smaller holes, where rock samples were taken for analysis in the laboratory. In total 16 excavation boreholes were drilled with the objective of including Fracture A, with the average length of 5 m, and a larger surface area than shown in Figure 3-13 could be characterised. The fracture surfaces were characterised by neutron activation analysis and where samples had been taken, surface and thin-section depth profiling of solute concentration for Cs, Eu, Nd, Th, and U were performed. Atomic absorption analysis was also used to obtain depth and surface profiles of Sr and Cs concentrations.



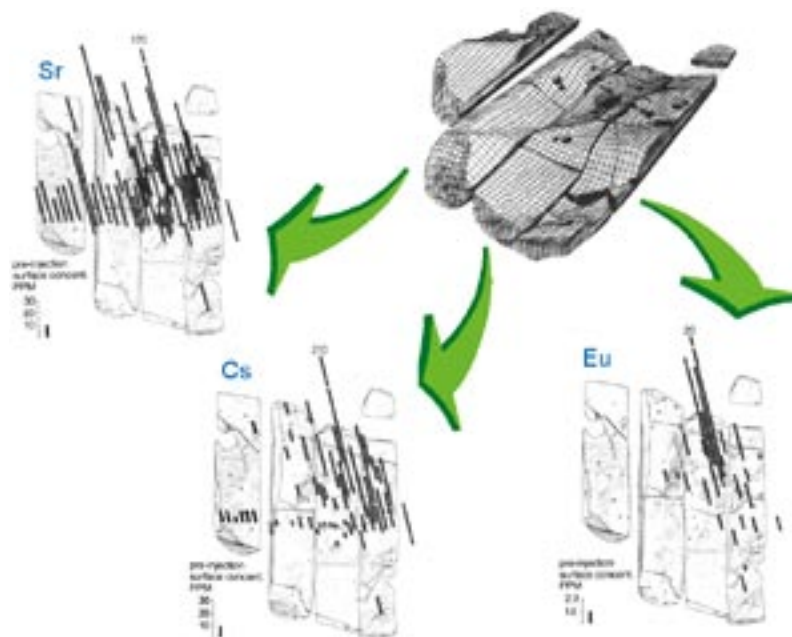
**Figure 3-13.** Representation of an excavated fracture surface close to an injection hole. Images taken from /Abelin et al. 1985/.

## Major achievements

This campaign had as a prime objective to characterise retention and channelling by means of obtaining sorption patterns at the fracture surfaces of Fracture A. Figure 3-14 shows an example of such sorption patterns immediately around the H2 injection hole.

By characterising the fracture surface up to a distance of 5 m from the injection hole (though in general with a lower resolution than that shown in Figure 3-14) surface concentration profiles could be obtained. Also a few samples at distance from the injection hole H2 were taken for depth profiling. Due to 1) the varying sampling density over the fracture plane more distant to injection hole H2 and 2) the fact that at some locations the drill cores did not include the flowpath, no complete characterisation of the flowpath's sorption pattern could be obtained. However what could be seen was that /Abelin et al. 1985/:

- For the species originally thought to be sorbing, sorption (or at least partitioning) did occur at the fracture surfaces.
- The results of the depth profiling analysis gave strong indications of both matrix diffusion and partitioning of solute within the rock matrix.
- The surface concentrations of the sorbed species generally declined with distance from injection hole H2.
- The sorption pattern indicated heterogeneous sorption on the fracture surfaces and/or heterogeneous flow (channelling).
- The breakthrough curves of the non-sorbing species in the short sampling holes, taken together with groundwater flow rates and other results from hydraulic testing indicated channelling in the fracture planes.
- $\text{Sr}^{2+}$  was found to be more strongly retarded than originally expected, possibly due to clay infillings in the fractures.
- The spatial distribution of the tracers indicated that the solutes were transported all the way to the edge of the excavated sampled part of Fracture A, indicating a noteworthy spread of the injected tracer.



**Figure 3-14.** Sorption patterns for Sr, Cs and Eu around the injection borehole H2 in the 2D migration experiment. Images taken and modified from /Abelin et al. 1985/.

### 3.4.5 3D migration experiment

The aim of the 3D migration experiment /Abelin and Birgersson 1987, Abelin et al. 1987ab, Birgersson et al. 1992a/ was to obtain information on channelling and preferential flowpaths and to determine flow porosities and longitudinal and transverse dispersion.

#### **The site and site characterisation**

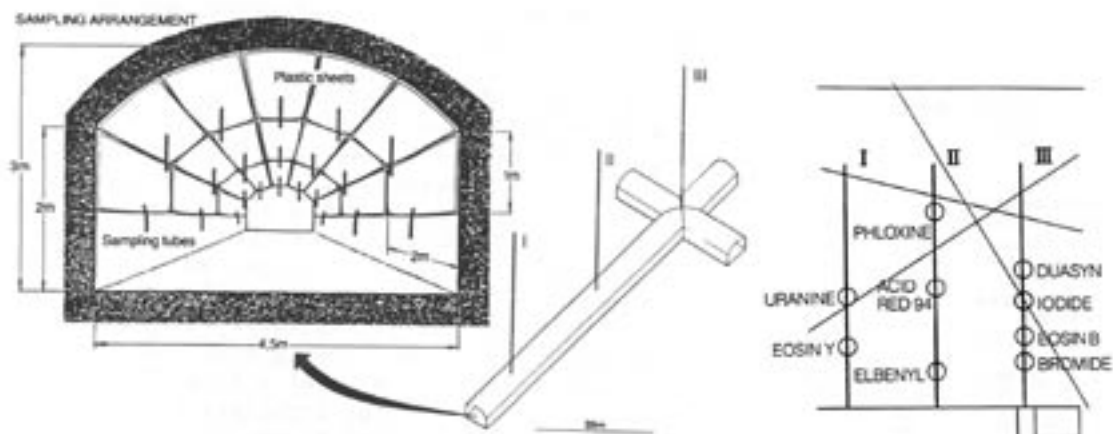
Prior to the 3D experiment, a new drift was excavated at the 360 m level, about 100 m from the old Stripa mine tunnels to reduce the risk of disturbances. The experimental site consisted of a 75 m long main drift with a crossing arm 25 m in length, as seen in Figure 3-15 below.

From the roof of the main drift, three 70 m long boreholes were drilled and investigated by means of core logging, borehole camera inspection, water inflow monitoring, pressure monitoring, and radar measurements. Not only the boreholes, but the entire site including the drifts, access tunnels, etc was subjected to a detailed site characterisation. This included mapping of all fractures intersecting the drifts and the characterisation of the groundwater chemistry of the site.

Based on these investigations, nine sections for the subsequent tracer injections were selected. The linear distances between injection points and the main drift ranged from 10–55 m.

The roof and walls of the drifts were lined with 375 plastic sheets, each about 2 m<sup>2</sup> in area. Each of these sheets collected groundwater dripping from fractures intersecting the drifts with the aim of both characterising inflow rates to the drift as well as to serve as tracer recovery points. The inflow into the tunnel was generally low with a total inflow of approximately 700 ml/h over the entire covered area. During the flow characterisation phase, it was found that 50% of the total inflow was restricted to about 3% of the sampled area. Furthermore, it was found that one of the sheets was responsible for about 10% of the total water inflow.

The tracer tests were performed using non-radioactive and non-sorbing tracers that were actively injected at low flow rate into hydraulically conductive fractures in the nine packed-off sections previously selected. The tracers used were Uranine, Eosin Y, Phloxine, Acid Red 94, Elbenyl, Duasyn, Eosin B, I<sup>-</sup>, F<sup>-</sup>, STR-7, and Br<sup>-</sup>. Figure 3-15 (far right) shows the injection locations for the different tracers and also the interpreted major fracture zones. F<sup>-</sup> and STR-7 were injected over a period of time simultaneously with, and from the same section as, Uranine. Injection times spanned between 7,000–16,000 hours.



**Figure 3-15.** Left: Layout of drifts. Right: Tracer injection points, Images taken from /Birgersson et al. 1992a/.

Groundwater entering the drifts was collected during the injection period and for an additional 6 months after the cessation of injection. The tracer recovery data gave a wealth of information about fracture connectivity and advective travel times. Major tracer recovery locations and water inflow rates are shown in Figure 3-16 below.

An interesting observation in the tracer experiment was that the tracers appeared exclusively in the central part of the main drift and there was essentially no tracer recovery via the main flow locations in the right hand end of the cross arm in spite of the fact that these were closer in proximity to the injection locations in borehole III. Some of the injected tracers (Phloxine, Acid Red 94, STR-7, and F<sup>-</sup>) were never recovered during the course of the experiment, and one tracer Duasyn was only recovered in small amounts (< 0.002% recovery) after many months of sampling. The other tracers had recoveries between 2.8–65% and average residence times varied between 2,000–7,000 hours. However, for individual sheets the residence times could be considerably shorter as well as longer.

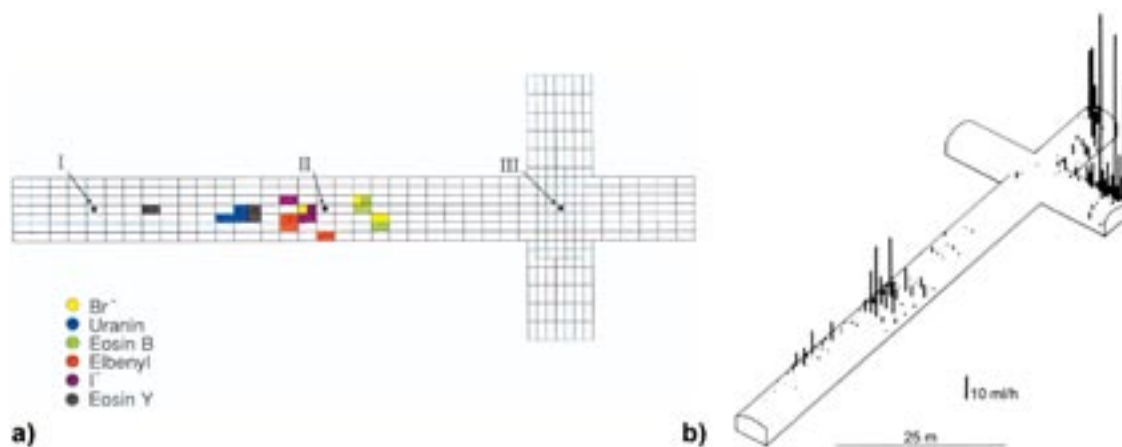
### Major achievements

This very elaborate campaign gave clear indication of the heterogeneity of the large-scale flow field at a site. As can be seen in Figure 3-16, tracers preferentially travelled through the rock mass in a few fractures intersecting the middle of the main drift. Also the groundwater flow was focused to a few locations within the drifts. It should be noted that from the fracture mapping of the drift walls, the density of major structures appeared to be fairly evenly distributed in all parts of the drifts.

The tests also demonstrated the difficulty in predicting flowpaths carrying tracers. One might have expected that the tracers injected in borehole III would preferentially end up in the closely located right hand section of the crossing arm, which is the area with the highest groundwater inflows, or the outer end of the main drift where large inflows were detected. However, the results showed that this was not the case.

It is also interesting to note that the Duasyn tracer was first detected in a location at the far end of the drift and was not found in combination with any of the other injected tracers. This indicates that the Duasyn travelled along flowpaths that were separate to those traversed by the other injected tracers.

During the course of the experiment, none of the tracers were found in any other sampling locations within the mine site outside the 3D drift. After the conclusion of the 3D experiment and during excavation operations for the next phase of the Stripa programme, however, a significant



**Figure 3-16.** The 3D migration experiment at Stripa showing (a) locations in drift where the highest tracer concentrations were found, and (b) water inflow rates to drift prior to drilling of injection holes. Images taken from /Birgersson et al. 1992a/.

fraction of the injected Eosin Y (> 10%) was found in a newly excavated gallery some 150 m distant from the injection location. This, although an unintended result, was indicative of extensive connectivity of the fracture system over a substantial distance through the rock.

One of the aims of the experiment was to obtain flow porosities for the different flowpaths. From the tests, flow porosities ranging from  $2 \cdot 10^{-5}$  to  $1.6 \cdot 10^{-4}$  were estimated for different pathways.

The 3D migration experiment is, together with the TME experiment addressed in subsection 3.4.7, a unique test and considerable advances were made in methodological and technological developments.

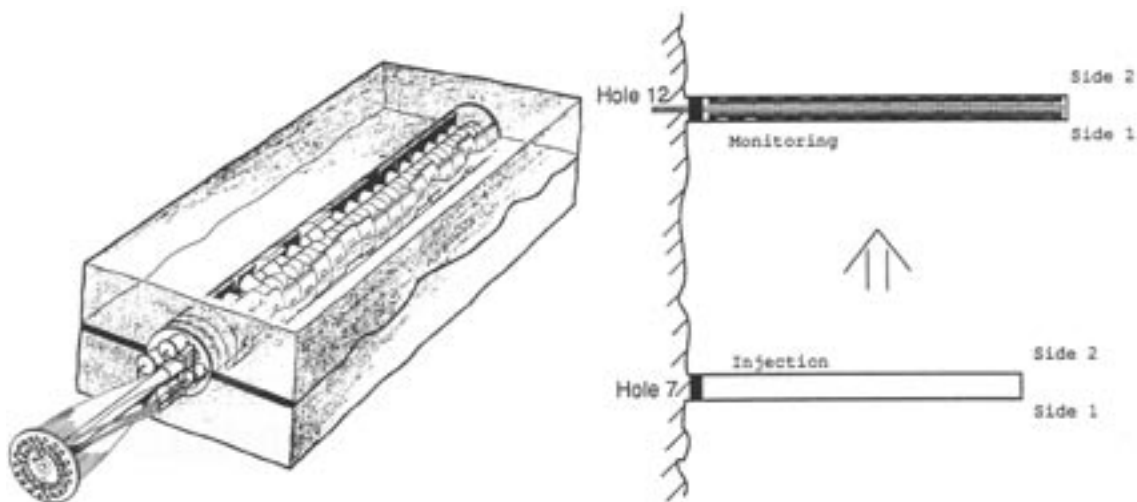
### 3.4.6 Channelling experiments

The Channelling experiment was performed at the 360 m level in the Stripa mine /Birgersson et al. 1992a, Abelin et al. 1990, 1994/. The aim was to study local channelling effects within a fracture plane. This was done both by single-hole hydraulic injection tests and by cross-hole tracer tests. In the single-hole tests, a borehole ( $\varnothing$  200 mm) was drilled from the tunnel along the plane of a hydraulically conductive fracture to a depth of at least 2 m. A “multipede” packer system, featuring 20 sections each 50 mm wide, as shown in Figure 3-17 (left), was installed.

Hydraulic injection tests could be performed with a resolution of 50 mm. By taking photographs of the fracture plane prior to the installation of the multipede, the flow rates and visible apertures could be compared.

In the cross-hole tracer test, a second borehole was drilled at a distance of about 2 m from an existing borehole, which had been found to be suitable on the basis of previous single-hole injection tests. The second borehole was drilled along the same fracture plane as the first and as a preparation for the tracer test, the fracture plane between the two boreholes as seen from the drift was sealed off with an epoxy glue. Prior to the tracer test the second borehole was subjected to single-hole injection tests and a series of cross-hole pressure interference tests were also performed.

The entire length of the second borehole was used for tracer injection. However, the borehole was packed-off into five sections, each functioning as a reservoir for the five non-sorbing dye tracers Uranine, Eosin Y, Elbenyl, Duasyn, and Phloxine. By using the multipede packer system for monitoring and sampling, the five different tracers could be withdrawn over the 2 m long multipede with a resolution of 50 mm (as seen in Figure 3-17) and channelling effects could thereby be studied.



**Figure 3-17.** Left: Multipede packer system used in the Channelling experiment. Right: Boreholes for injection and monitoring. Images taken from /Birgersson et al. 1992a/.

## Major achievements

In this tracer test, small-scale channelling could be studied. The results indicated the presence of dead end channels and also intersecting fractures that diverted the flow from the main fracture. The channel widths were estimated to be 10 cm or less and the flow apertures were estimated to be on the order of several hundred micrometers.

From examining the photographs of the fracture intersecting the borehole it was concluded that the description of a fracture as made up of two parallel plates is not very good. The visible apertures were better described by an array of small holes in an otherwise nearly closed fracture. By comparing the visible apertures with the injection flow rates it was also concluded that there was no correlation between the entities.

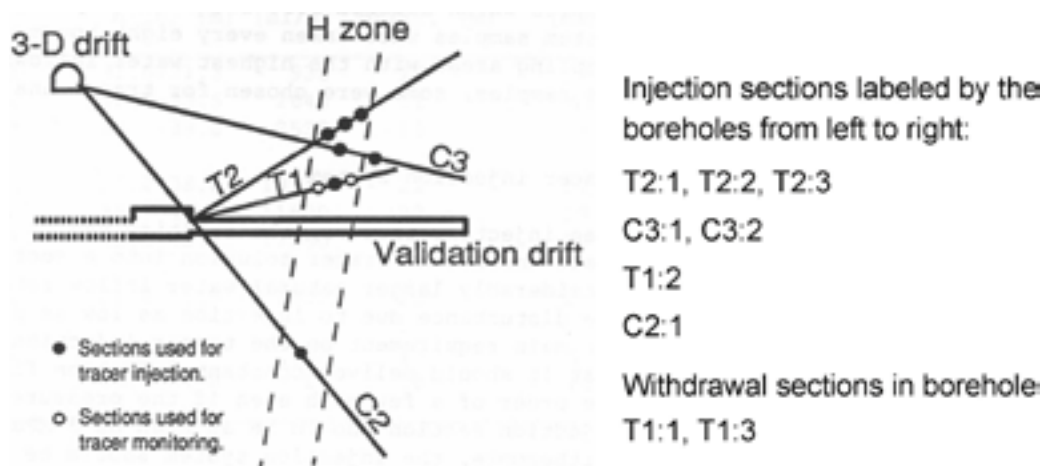
By analysing the flow results from the injection tests, variograms could be made and the correlation lengths for flow rate spatial variability were estimated to be somewhere between 5 cm (i.e. the lowest achievable resolution) and 50 cm.

### 3.4.7 Tracer Migration Experiment in the Validation drift – TME

The Tracer Migration Experiment (TME) /Birgersson et al. 1992ab, Birgersson and Ågren 1992/ resembles the 3D migration experiment with the exception that the Validation drift is intersected by a large fracture zone. The aim was to study tracer transport and retention in a fracture zone where the inflow into the drift was roughly one order of magnitude larger than that observed in the 3D drift.

The Validation drift was excavated at a depth of 385 m below ground level in the Stripa mine to transverse a large fracture zone. The TME was a part of the Site Characterisation and Validation (SCV) project that aimed at validating all techniques used in Site Characterisation. Therefore the site was subjected to a broad range of geoscientific investigations, including geological, geophysical, hydrogeological, and hydrogeochemical characterisations.

As in the 3D migration experiment, the roof and walls of the Validation drift were lined with plastic sheets for collection of groundwater flowing into the drift. In the TME about 150 such sheets were used. To increase the accuracy of water inflow measurements in the Validation drift, measurements of water content in ventilation air and water inflow to sump holes drilled in the tunnel floor were used. The total groundwater inflow in the entire Validation drift was about 6,000 ml/h and about 99% of that flowed from a few individual fractures in the approximately 6 m wide fracture zone H. Figure 3-18 shows the location of the 50 m long validation drift, the 3D drift, the major fracture zone H, and also four boreholes drilled into the SCV rock volume that were used for tracer injection.

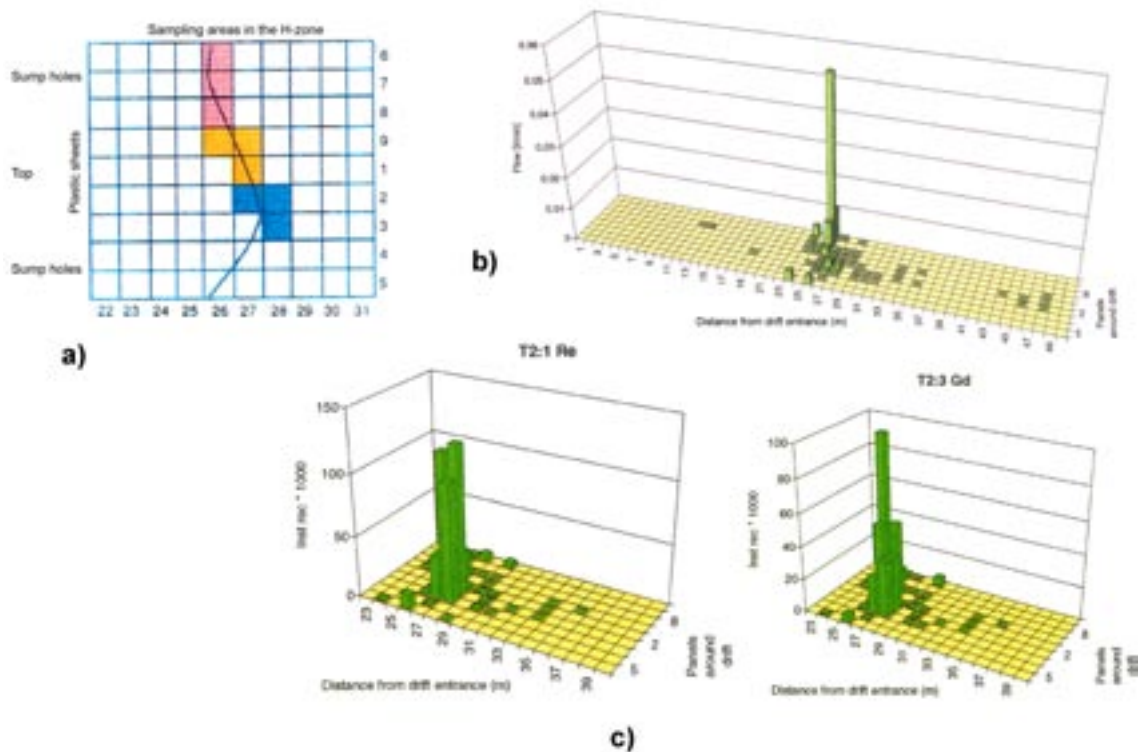


**Figure 3-18.** Location of Validation and 3D drifts in the Stripa mine. Image taken from /Birgersson et al. 1992a/.

The campaign was comprised of two different runs of tracer tests. In the first run, tracers were injected from the boreholes and collected in the drift. In the second run, tracers were injected in borehole T2 and withdrawn in borehole T1. Prior to the tracer tests, all boreholes were sealed off using bentonite with the exception of nine short sections. Seven of these sections were used for tracer injection where one, T2:2, was only used in the second tracer test run. The two withdrawal sections in borehole T1 were only used in the second run, in which their pressure was maintained at the same level as in the drift. All sections were located in the H zone except for one injection section, C3:2, located in low permeability rock adjacent to the zone (see Figure 3-18). Injection was performed actively for between two and seven months in the first tracer test run but only for a week in the second run. The injection flow was kept low with the intention of not excessively disturbing the natural flow field.

As dye tracers had previously been used in the 3D migration experiment and there was a risk of tracer cross-contamination between the tests, six trivalent metal tracers were used in combination with six dyes. Five of the metals used were the lanthanides Eu, Gd, Tb, Dy, and Ho, bound to EDTA or DTPA to render them non-sorbing and the sixth metal was  $\text{ReO}_4^-$ . The dyes used were Ebnyl, Rose B, Eosine Y, Phloxine B, Uranine, and Duasyn. One dye and one metal tracer were used in each injection point. In addition, an injection with  $^{86}\text{Sr}^{2+}/^{88}\text{Sr}^{2+}$  was made. For the exact injection sequence we refer to /Birgersson et al. 1992b/. The linear distances from the injection points to the drift ranged from 10–25 m. Monitoring and sampling was carried on for about ten months in the drift in the first tracer test run. For the second tracer test run monitoring and sampling in borehole T1 was only carried on for about a month. During this time, monitoring and sampling was also continued in the drift.

The six tracer injections in the first run were associated with different breakthrough patterns in the drift, although the tracer recovery was largely isolated to a couple of sections with high flow. Figure 3-19 shows a selection of the obtained results.



**Figure 3-19.** a) Major tracer recovery locations for different injections. b) Map of water inflow rates to drift; c) Typical maps showing recovery locations of  $\text{ReO}_4^-$  and Gd-EDTA. Images taken from /Birgersson et al. 1992a/.

In Figure 3-19b the groundwater flow rate into the drift is shown. As can be seen the flow is highly concentrated to the middle of the drift where it is intersected by the H zone. Figure 3-19c shows the tracer inflow rates for  $\text{ReO}_4^-$  and Gd-EDTA, also corresponding to the location of the H zone. In the first tracer test run, the main flowpaths between the different injection sections and the drift appeared to be located in the same fracture in the H zone but not in the same parts of the fracture plane. Figure 3-19a indicates the most hydraulically conductive fractures in the zone intersecting the drift by the curved black line. Tracers from injection section C2:1 (under the drift) preferentially ended up in the sampling areas marked by pink in the figure. Tracers from injection sections T1:2, T2:3, C3:1, and C3:2 preferentially ended up in the sampling areas marked by orange and tracers from T2:1 ended up in the sampling areas marked by blue. Almost 100% of the recovered tracers ended up in the 10 sampling areas coloured in Figure 3-19a. For the metal tracers, with the exception of  $\text{Sr}^{2+}$ , the recovery varied from 10 to 65%. For the dyes, the recovery varied between only a few percent to  $\sim 40\%$ , although tracers were still emerging when the experiment was discontinued. No  $\text{Sr}^{2+}$  detected in the drift could be derived from the injection. The mean residence time for the tracers varied between 1,500 and 4,000 hours. As in the 3D migration experiment, for individual sampling areas the residence times could be considerably shorter as well as longer than the mean.

Concerning the second tracer test run, from boreholes T2 to T1, it was suspected, based on hydraulic testing before and after the first run, that bentonite had entered the fracture system. Also tracers from the first run remained in the system in the second run, rendering the obtained breakthrough curves difficult to interpret.

### **Major achievements**

From a methodological point of view, the Tracer Migration Experiment in the Validation drift in many ways resembles the 3D migration experiment, with the exception of a few improvements made to better measure the total groundwater inflow into the drift. Non-sorbing metal complexes were also used in pair combinations with dye tracers unlike the 3D migration experiment.

A major result was that even in the most hydraulically conductive part of a fracture zone, in this case interpreted as an individual fracture, tracers did not spread over the entire fracture plane. Even though a large majority of all tracers exited from the same fracture, tracers from different injection points travelled along spatially separated flowpaths within the fracture plane, as seen in Figure 3-19a.

Another interesting observation was that 99% of the groundwater flowing into the drift emerged from the fracture zone H and 50% of this flow was collected in a single  $1 \text{ m}^2$  large sampling sheet. This shows that the flow within a hydraulically conductive zone may be very heterogeneous. A conclusion that could be drawn from this is that when assigning a representative flow-wetted surface for a zone, this should be based on hydraulic information and not geological fracture mapping. In the Validation drift, 50% of the inflowing water potentially encountered only a relatively small flow-wetted surface.

From modelling, flow porosities between  $3.5 \cdot 10^{-3}$  and  $1.6 \cdot 10^{-3}$  were obtained, where the estimated flow porosity decreased slightly with distance from the drift.

## **3.5 Tracer tests at Äspö Hard Rock Laboratory**

### **3.5.1 Long Term Pumping and Tracer Test – LTP2**

Prior to the construction of the Äspö HRL, SKB performed a series of cross-hole tracer tests in boreholes drilled from the ground surface. This was done as a part of the Long Term Pumping and Tracer Test campaign – LTP2 /Rhen et al. 1992/. One open borehole, KAS06, was used for tracer withdrawal and six packed-off sections in five surrounding boreholes were used for tracer injection. The test geometry was essentially a radially converging flow system. Non-sorbing



tracers were injected in hydraulically conductive fractures and monitoring and sampling was performed on withdrawn groundwater.

The main objectives of the LTP2 were primarily to determine how major fracture zones at the site were interconnected and secondarily to determine transport parameters such as residence time, dispersivity, flow porosity, and hydraulic conductivity. A sketch of the experiment is shown in Figure 3-20, where packed-off borehole sections at identified hydraulically conductive fracture zones were used for tracer injection. Pumping was performed in the entire central borehole but by using a multisampler, groundwater could be withdrawn from different parts of the borehole.

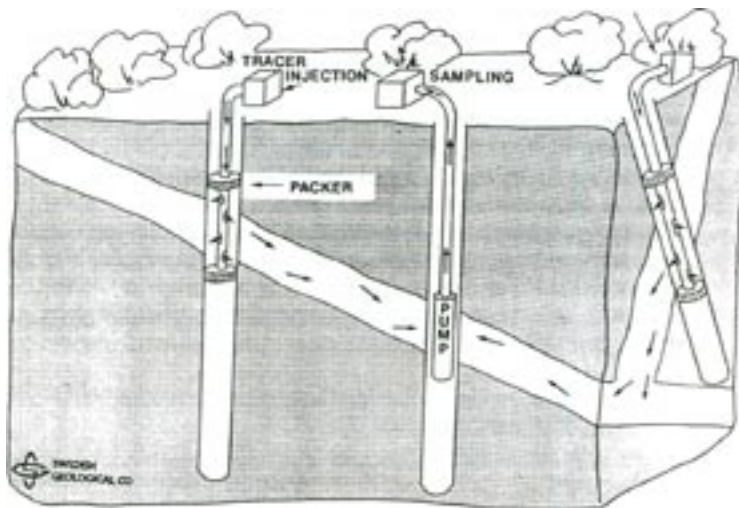
Before performing the cross-hole tracer tests, a number of dilution tests were performed in 30–100 m long packed-off sections of the injection boreholes. This was done to identify appropriate fracture zones that could be utilised for the cross-hole tracer tests.

In the cross-hole tracer tests, the non-sorbing radioactive tracers  $^{114}\text{In}$ ,  $^{131}\text{I}$ , and  $^{186}\text{Re}$  were used together with Uranine. Prior to the tracer tests, pumping was performed for two weeks in order to achieve a steady state flow situation. The tracer injection took the form of an intermittent decaying pulse injection (i.e. successive pulses of tracer with intervening lag time). The tracer tests were performed for 76 days at a depth ranging from 140–600 m below sea level. The geometric distance from points of injection to point of withdrawal ranged from 100 to 300 m. The flowpaths were estimated to be somewhat longer.

### **Major achievements**

The major achievement of the campaign was that the hydrostructural model for the Äspö site could be further strengthened. Concerning the tracer tests, during the 76 days the test was performed, clear recovery from three out of the six injection points was achieved and from one section it was uncertain whether recovery was achieved or not. Using a multisampler in an open borehole made interpretation of tracer breakthrough complicated, as the sampling locations in the borehole were not hydraulically isolated. The tracer tests confirmed connection between three (possibly four) injection points and the pumping borehole KAS06, but did not disqualify connection between KAS06 and the other injection points.

Prior to the test the tracer residence times were predicted and when comparing to the test result, the advective travel time predictions were found to be somewhat overestimated. The offered explanation was that an overly large flow porosity was used in the prediction. Based on the tracer test results, flow porosities between  $2.0 \cdot 10^{-4}$  and  $5.0 \cdot 10^{-2}$  were assessed. Furthermore, from the results it was judged that macrodispersion was an important process during the



**Figure 3-20.** Sketch of LTP2, showing injection and withdrawal boreholes. Image taken from /Rhén et al. 1992/.

prevailing pump conditions. The experimental design and performance was also judged to be very successful.

### 3.5.2 Tracer Retention Understanding Experiments – TRUE

The TRUE (Tracer Retention Understanding Experiments) series of projects constitutes TRUE-1, TRUE Block Scale, TRUE-1 Continuation, and TRUE Block Scale Continuation. Results from TRUE-1 and TRUE Block Scale, and their implications for SC and SA modelling have been examined and discussed by an international collaborative group, the Äspö Task Force, and Task 6 is shortly summarised in this report. The TRUE series of projects include extensive investigations both in the field and in the laboratory. However, the laboratory programme is not discussed in this report.

The location for the TRUE series of projects is the Äspö HRL at a depth of about 400–500 m below sea level. Äspö HRL has undergone a major Site Characterisation using a broad range of geoscientific investigation techniques, both prior to and during the TRUE series of projects.

The overall objectives of the TRUE series of projects are, according to /Winberg et al. 2000/:

- To develop the understanding of radionuclide migration and retention in fractured rock.
- To evaluate to what extent concepts used in models are based on realistic descriptions of fractured rock and if adequate data can be collected in Site Characterisation.
- To evaluate the usefulness and feasibility of different approaches to model radionuclide migration and retention.
- To provide in situ data on radionuclide migration and retention.

Presently, TRUE-1 and TRUE Block Scale are completed while continuation programmes are underway. Figure 3-21 shows the layout of Äspö HRL. TRUE-1 Continuation is mainly carried out at the TRUE-1 site while TRUE Block Scale Continuation has its focus on the True Block Scale site /SKB 2005c/.

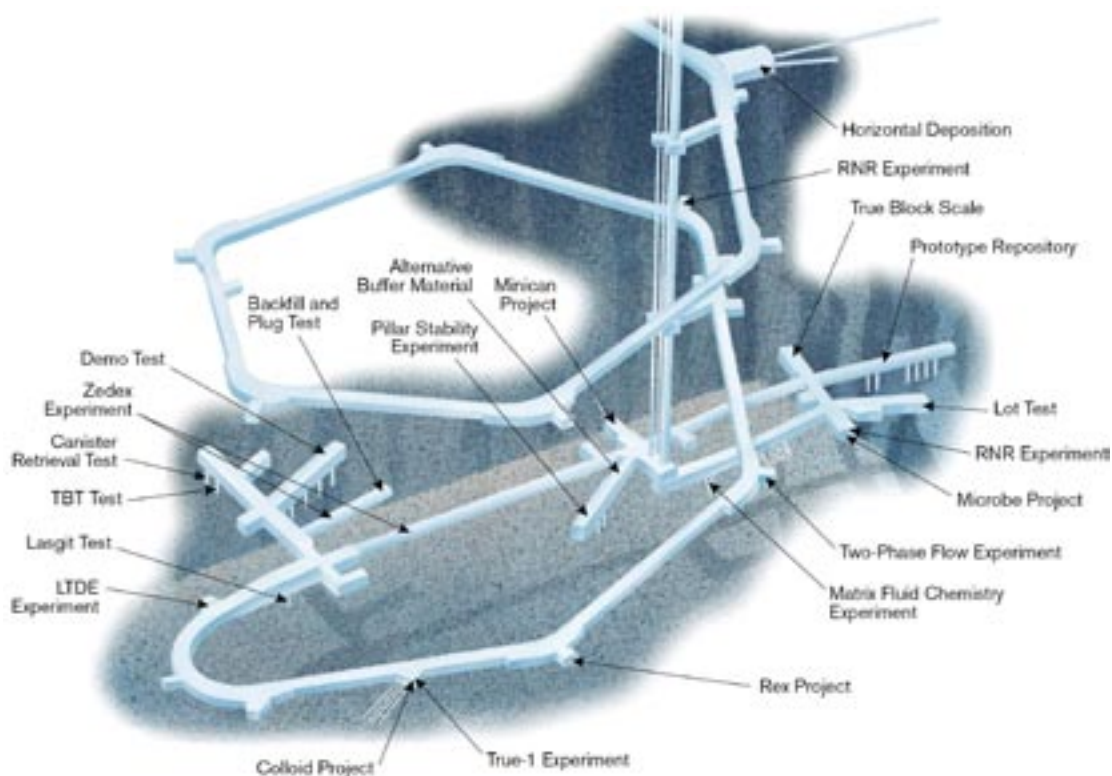


Figure 3-21. Layout of Äspö HRL. Image taken from /SKB 2006g/.

### 3.5.3 TRUE-1

The basic idea behind the TRUE series of projects was to perform a series of tracer tests with successively increasing complexity. Each campaign consisted of a cycle of activities, beginning with geological characterisation and design modelling followed by hydraulic testing and tracer tests.

The first test cycle was TRUE-1, reported in /Winberg et al. 2000/, which aimed at understanding radionuclide transport and retention in a single fracture. In TRUE-1 the tracer tests were conducted over a limited scale (less than 10 m) and had a limited duration. The primary aim of TRUE-1 was methodology and technology development and initially only non-sorbing tracers were intended to be used. However, as the project progressed the objectives were changed to include the use of sorbing tracers and to determine retention parameters.

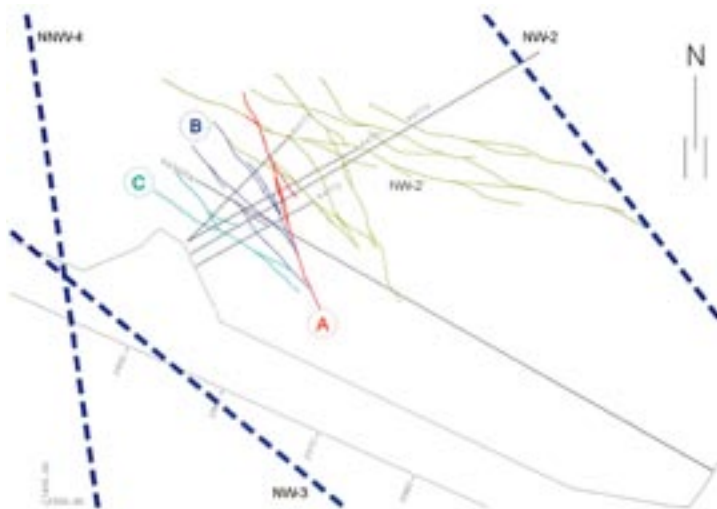
In TRUE-1 much effort was devoted to developing geological and hydrological structural models, both on a macro- and micro-scale. Much of the conceptual understanding presented in Chapter 2 of this report is based on findings in TRUE-1 (and the rest of the TRUE series of projects).

#### Site selection

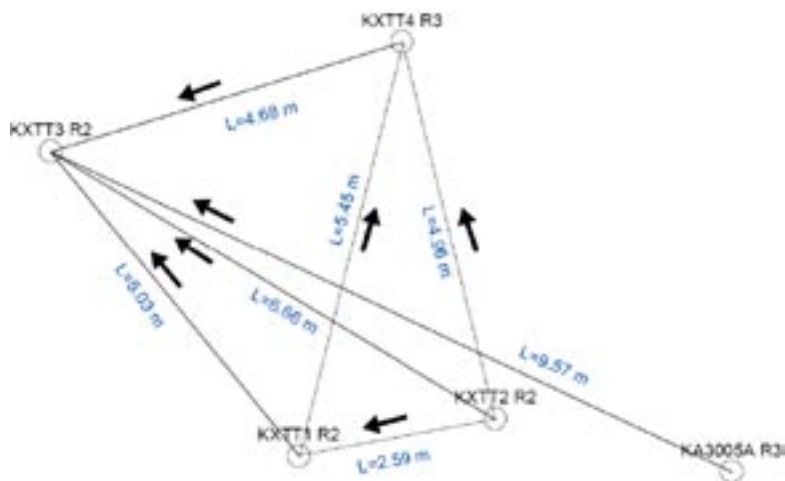
The experimental phase was preceded by the elaborate site selection programme SELECT, in which investigations were performed in a number of 10–70 m long boreholes penetrating the candidate volume. The boreholes and drill cores were subjected to various types of characterisation methods including borehole TV imaging (BIPS), drill core logging, geophysical measurements, single-packer flow logging, multi-packer cross-hole interference tests, and tracer dilution tests.

From the SC data, a number of hydraulically conductive structures were identified within the investigated volume. One of these structures, the so-called “Feature A”, was selected for further studies including the subsequent tracer tests. Figure 3-22 shows the interpreted structures in blue, green, yellow, and red. The dashed blue lines represent larger deformation zones, of which NNW-4 and NW-3 intersect the Äspö HRL tunnel close to the TRUE-1 rock volume.

Feature A was selected on the basis of its relative simplicity (interpreted as a single fracture) and its hydraulic isolation from neighbouring features. In addition its transmissivity was in a suitable range ( $8 \cdot 10^{-9}$ – $4 \cdot 10^{-7}$  m<sup>2</sup>/s). In Figure 3-23, five boreholes are shown that intersect Feature A. Furthermore, the geometric distances between the boreholes where they intersect the plane of Feature A are shown.



**Figure 3-22.** Features and boreholes within the TRUE-1 rock volume. The cross section represents ~ 35×60 m. Image taken from /Winberg et al. 2000/.



**Figure 3-23.** Flowpaths and flow direction between boreholes in Feature A. Image taken from /Winberg et al. 2000/.

### Tracer tests

A total of 18 cross-hole tracer tests were performed within TRUE-1 along seven different flowpaths in Feature A and one in Feature B, where distances ranged from 2.6 to 9.6 m between injection and withdrawal points. Figure 3-23 shows the flow direction of the seven different flowpaths utilised in the tracer tests performed in Feature A.

Both radially converging and unequal dipole configurations were used in the tracer tests. The bulk of the tracer tests used non-sorbing tracers only and were intended to provide information on connectivity and advective travel times for use in designing subsequent tracer tests using a cocktail of sorbing and non-sorbing tracers. Non-sorbing tracers included Uranine, Amino-G acid, Rhodamine WT, Eosin Y, Gd/Eu/Ho/Tb-DTPA, HTO,  $^{82}\text{Br}^-$ , and  $^{131}\text{I}^-$ .

Based on information from preliminary tracer tests, the most suitable sink-source pairs for performing three tracer tests with sorbing radionuclides were identified. The STT-1 and STT-2 tests were performed between boreholes KXTT4 and KXTT3 while STT-1b was performed between boreholes KXTT1 and KXTT3 (see Figure 3-23). The following sorbing tracers were used:  $^{22}\text{Na}^+$ ,  $^{42}\text{K}^+$ ,  $^{47}\text{Ca}^{2+}$ ,  $^{58}\text{Co}^{2+}$ ,  $^{85}\text{Sr}^{2+}$ ,  $^{86}\text{Rb}^+$ ,  $^{99\text{m}}\text{Tc}$ ,  $^{131/133}\text{Ba}^{2+}$ , and  $^{134/137}\text{Cs}^+$ . The tracer tests were, at most, carried on for 15 months. Many of the same tracers were also used in the laboratory for comparative studies on generic samples of intact Äspö rock and rock samples taken from drill cores of the TRUE-1 site.

### Pilot Resin Experiment

As TRUE-1 progressed, it was decided to develop a method where an epoxy resin could be injected in a fracture plane. The Pilot Resin Experiment /Winberg et al. 2000/ was performed at a different location than the rest of TRUE-1, in the F-tunnel close to the TRUE Block Scale site. The experiment had technology development as a prime objective. Upon hardening of the resin, the fracture plane could be overcored and the fracture pore space examined. The intent was that with this technique one could obtain information on the pore space geometry, on flow channelling within a fracture plane, and on the flow aperture.

Prior to the resin injection, nine short boreholes were drilled to intersect a target structure. By performing investigations in these boreholes, and on their drill cores, the site was characterised much in the same way as the TRUE-1 site. In three of the boreholes successful injection of epoxy resins was achieved. This was done by injecting in one hole at a time and using one or a few of the other boreholes for drainage. Each borehole was injected with a resin that had been labelled with a specific colour, in order to facilitate analysis of the resin spread upon excavation.

The resin could be injected over a period of several hours before it hardened. Based on the amounts of resin injected it was estimated that a resin spread on the order of one or a few square metres had been achieved. Upon the hardening of the resin, the overcoring was performed and by examining the drill cores the fracture aperture could be estimated based on the visible resin thickness.

### **Major achievements**

The major achievements of TRUE-1 can be divided into three categories, development of methodology and technology, development/refining of a site descriptive model, and successfully performing and evaluating tracer tests using sorbing and non-sorbing tracers.

As TRUE-1 had development of methodology and technology as one of its primary objectives, major achievements were made in many areas. A technology improvement highlighted in /Winberg et al. 2000/ was the development of an injection method giving rise to reduced tailing of the tracer injection pulse. A sophisticated system was devised to quickly exchange the tracer fluid in the packed-off injection section by non-traced groundwater, upon cessation of the tracer injection. The injection section was also equipped with volume-reducer “dummies” to minimise the residence time effect associated with the volume of the injection well. In this way, a very precisely defined tracer pulse injection curve could be obtained without a long trailing edge (which otherwise complicates modelling interpretation).

Concerning the site descriptive model, much new information resulted from the TRUE-1 Site Characterisation. One major achievement was a refinement of the microstructural model of a fracture, its associated infilling material, and surrounding rock matrix. Another major achievement was the development of a hydrostructural model, where structures intersecting the site could be identified, their extension in space modelled, and their hydraulic connectivity demonstrated.

From the TRUE-1 tracer tests performed with sorbing tracers (STT-1, STT-1b, STT-2), clear indication of retardation was obtained. The solutes were injected simultaneously and with high resolution as a result of the sophisticated injection system, and the retardation of each solute could be quite clearly delineated. Although not all the tracers were fully recovered, the fact that the recovery sequence followed the so-called “Hofmeister series” /e.g. Stumm and Morgan 1996/ ( $\text{Na}^+ < \text{Ca}^{2+} \approx \text{Sr}^{2+} \ll \text{Rb}^+ \approx \text{Ba}^{2+} < \text{Cs}^+$ ) gave indications that ion-exchange sorption was most probably involved in the retardation of those particular solutes and was also consistent with data obtained from the TRUE-1 laboratory programme.

The tracer tests were initially evaluated using the LaSAR model /Cvetkovic et al. 1999/. From the evaluation it was found that the retention capacity of the flowpath was larger than expected based on the retention data obtained in the laboratory on intact rock samples /Winberg et al. 2000/. This was primarily explained as a result of fracture infillings and an alteration rim adjacent to the fracture with higher retention capacities. Also the possibility of an increased flow-wetted surface, compared to the expected, was offered as an explanation. The TRUE-1 tracer tests were modelled by a number of research groups in Tasks 4 and 6 of the Äspö Task Force, whereof Task 6 is summarised in subsection 3.5.5 of this report.

The major achievements of the Pilot Resin Experiment concerned technology development. The obtained mean fracture apertures were on the order of 0.2 mm.

### **3.5.4 TRUE Block Scale**

TRUE Block Scale /e.g. Winberg et al. 2003/ should be seen as a continuation of TRUE-1, with the aim at studying retention of sorbing and non-sorbing tracers in a fracture network on a 10–100 m scale. The general objectives of TRUE Block Scale were to /Winberg 1997/:

- Increase understanding of tracer transport in a fracture network and improve predictive capabilities.

- Assess the importance of tracer retention mechanisms (diffusion and sorption) in a fracture network.
- Assess the link between flow and transport data as a mean for predicting transport phenomena.

### **Site selection**

The tracer tests were preceded by a similar site selection programme as in TRUE-1, using the full range of geoscientific methods for characterisation of the rock volume of interest /Andersson et al. 2002a/. A great number of boreholes were drilled from the tunnel into the TRUE Block Scale rock volume for use in the Site Characterisation and subsequent tracer tests (see Figure 3-24). The characterisation programme in the field included:

- Borehole geophysics such as borehole radar, rock resistivity, etc.
- Drill core logging combined with borehole camera (BIPS) and other geological/mineralogical investigations.
- Single-hole and cross-hole hydraulic testing and studying of hydraulic drilling responses.
- Flow logging with, for example, the Posiva Difference Flow meter.
- Hydrogeochemical investigations, including chemical analyses of groundwater.
- Tracer dilution tests.

Based on the Site Characterisation, elaborate descriptive hydrostructural models were developed for the rock volume of interest /Poteri et al. 2002, Winberg et al. 2003/. Initially only a Discrete Fracture Network (DFN) model was to be developed but later on in the project also Stochastic Continuum (SC) and Channel network (DFN/CN) models were developed.

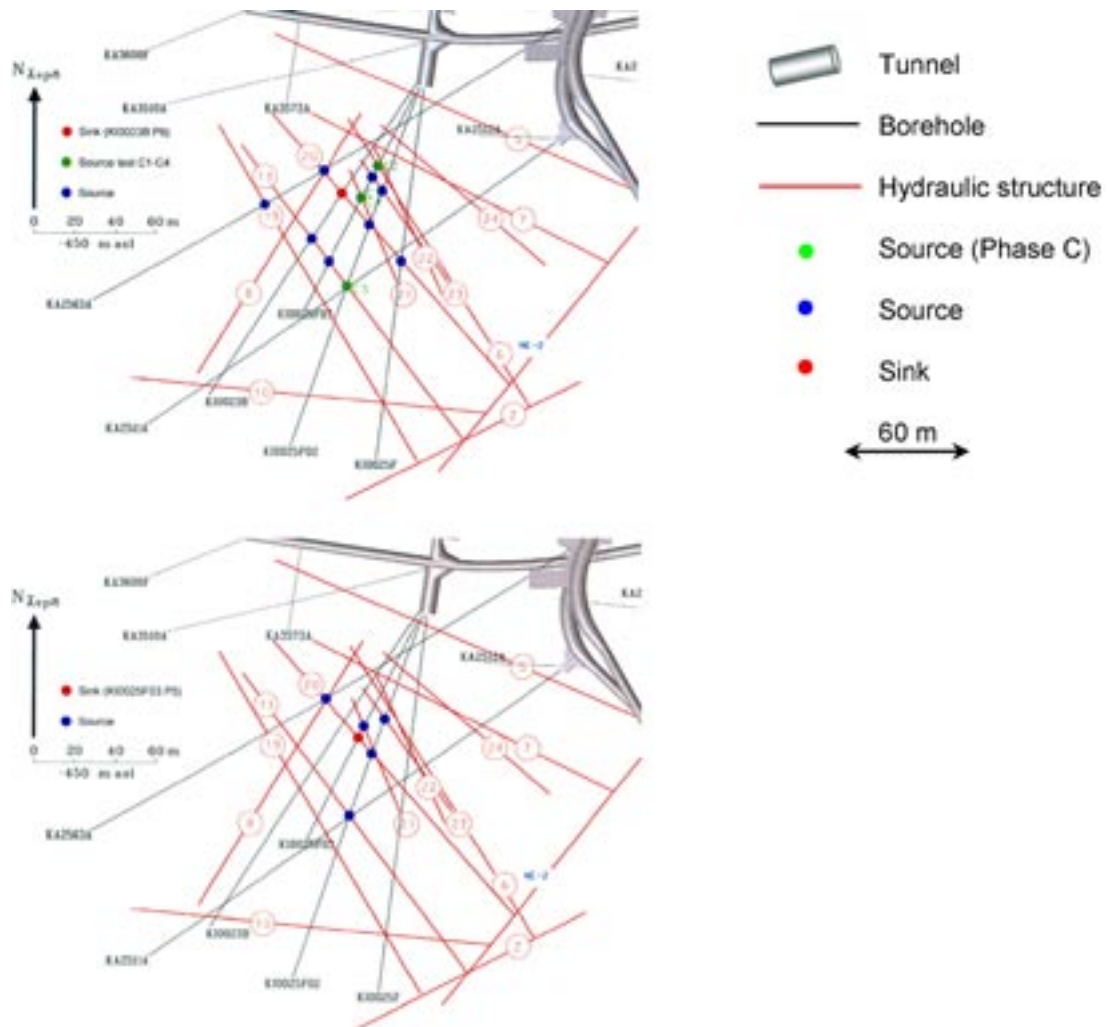
### **Tracer tests**

Based on the available information from the Site Characterisation, target structures were chosen for performing tracer tests. The tracer test stage of TRUE Block Scale was divided into three different phases. In the first phase (Phase A), hydraulic interference tests, tracer dilution tests, and preliminary cross-hole tracer tests were performed with the objective of finding two prioritised sink sections /Andersson et al. 2000a/. In the tracer dilution tests, pumping was performed in a packed-off section that was selected as a sink candidate for the subsequent phases. A great number (up to 17) of surrounding packed-off sections were filled with tracer and the tracer dilution was studied during the pumping. This was repeated for the study of four sink candidates. Out of these sinks, two were tested in cross-hole tracer tests, where tracers were sampled and monitored in the sink. The tracers injected in surrounding sections were Amino-G acid, Rhodamine WT, Naphtionate, and Uranine.

Phase B included 10 different tracer tests and aimed at demonstrating that high enough recovery could be achieved with the selected sink-source pair within a reasonable time period. The outlined objectives for Phase B were /Andersson et al. 2000b/:

- To investigate transport in fracture networks.
- To investigate heterogeneity within single fractures.
- To investigate the effect of fracture intersection zones (FIZ).
- To revisit earlier investigated flowpaths and verify high mass recovery.

The tracer tests were performed in radially converging or (strongly) unequal dipole configuration. The non-sorbing tracers Amino-G acid, Rhodamine WT, Naphtionate, Uranine,  $^3\text{He}$ , Yb-EDTA,  $\text{ReO}_4^-$ , Ho-DTPA, Gd-DTPA, and In-EDTA were used.



**Figure 3-24.** Boreholes, interpreted hydraulic structures, sources and sinks in TRUE Block Scale. Images taken and modified from /Winberg et al. 2003/.

Figure 3-24 shows the drilled boreholes and interpreted hydraulically conductive structures of the TRUE Block Scale rock volume. In addition, locations for tracer test sources and sinks are shown. In total 16 flowpaths (sink-source pairs) were tested and the flowpaths were equally divided in single structures and network structures.

Based on the results in Phase B, three flowpaths were suggested for Phase C. According to /Winberg et al. 2003/ the key design challenge with the tracer test runs on larger length scales using sorbing tracers was to select a test geometry which produced both sufficiently high mass recovery and analysable breakthrough curves within reasonable time frames. When choosing tracers for different tests, the experience gained from TRUE-1 was used and a similar approach was taken.

In the three chosen flowpaths, four different tracer tests with either radially converging or unequal dipole flow geometry, using decaying pulses and finite pulses, were performed /Andersson et al. 2001/. The sink and injection points for the Phase C tests are shown by red and green dots in Figure 3-24. The Euclidian injection-recovery well distance, via deterministic features in the hydrostructural model, ranged from 10–55 m /Andersson et al. 2002b/.

Tracer test C1 was performed between the closest sink-source pair (see Figure 3-24) and the following sorbing tracers were used:  $^{24}\text{Na}^+$ ,  $^{42}\text{K}^+$ ,  $^{47}\text{Ca}^{2+}$ ,  $^{86}\text{Rb}^+$ , and  $^{134}\text{Cs}^+$  together the non-sorbing tracers  $^{82}\text{Br}^-$  and Uranine. The main flowpath between source and sink was predicted to follow two separate structures in series.

The tracers in test C2 were injected in the most northern of the Phase C injection points. The following sorbing tracers were used:  $^{131}\text{Ba}^{2+}$ ,  $^{137}\text{Cs}^+$ , and  $^{47}\text{Ca}^{2+}$  together with the non-sorbing tracers  $^{186}\text{ReO}_4^-$  and Naphtionate. The main flowpath between source and sink was predicted to follow three separate structures in series.

The tracers in test C3 were injected in the most southern of the Phase C injection points. The following sorbing tracers were used:  $^{22}\text{Na}^+$ ,  $^{85}\text{Sr}^{2+}$ ,  $^{83}\text{Rb}^+$ , and  $^{133}\text{Ba}^{2+}$  together with the non-sorbing tracer HTO. The main flowpath between source and sink was predicted to follow a single structure.

The C4 tracer test was performed between the same sink-source pair as in C1. Sorbing tracers used were  $^{47}\text{Ca}^{2+}$ ,  $^{131}\text{Ba}^{2+}$ ,  $^{54}\text{Mn}^{2+}$ ,  $^{57}\text{Co}^{2+}$ , and  $^{65}\text{Zn}^{2+}$  together with the conservative tracers  $^{82}\text{Br}^-$  and  $^{131}\text{I}^-$ .

In the four tracer tests, first tracer arrival for the conservative tracers varied between about 4–100 hours. For the conservative tracers, good recovery (73–100%) was obtained in all tracer tests within the project's official monitoring period of 3,300 hours /Andersson et al. 2001/. For the sorbing tracers the recovery varied from almost complete recovery to no breakthrough, depending on the flowpath and solute.

Figure 3-25 shows the breakthrough curves of the tracers where recovery was obtained. Note the logarithmic scales on the axes and that the tracer concentration is normalised to the total injected activity.

In test C1 there was a breakthrough of Uranine that is not shown in Figure 3-25. However, Uranine was delayed compared to  $^{82}\text{Br}^-$  and it was suspected that this was due to problems in the injection where the pH underwent changes /Andersson et al. 2001/. For some tracer injections, sampling and analysis continued after the official monitoring time for two more years within the programme /Winberg et al. 2003/.

### **Major achievements**

A summary of the major achievements of TRUE Block Scale has been devoted a separate chapter in /Winberg et al. 2003/ to where the reader is directed for further reading.

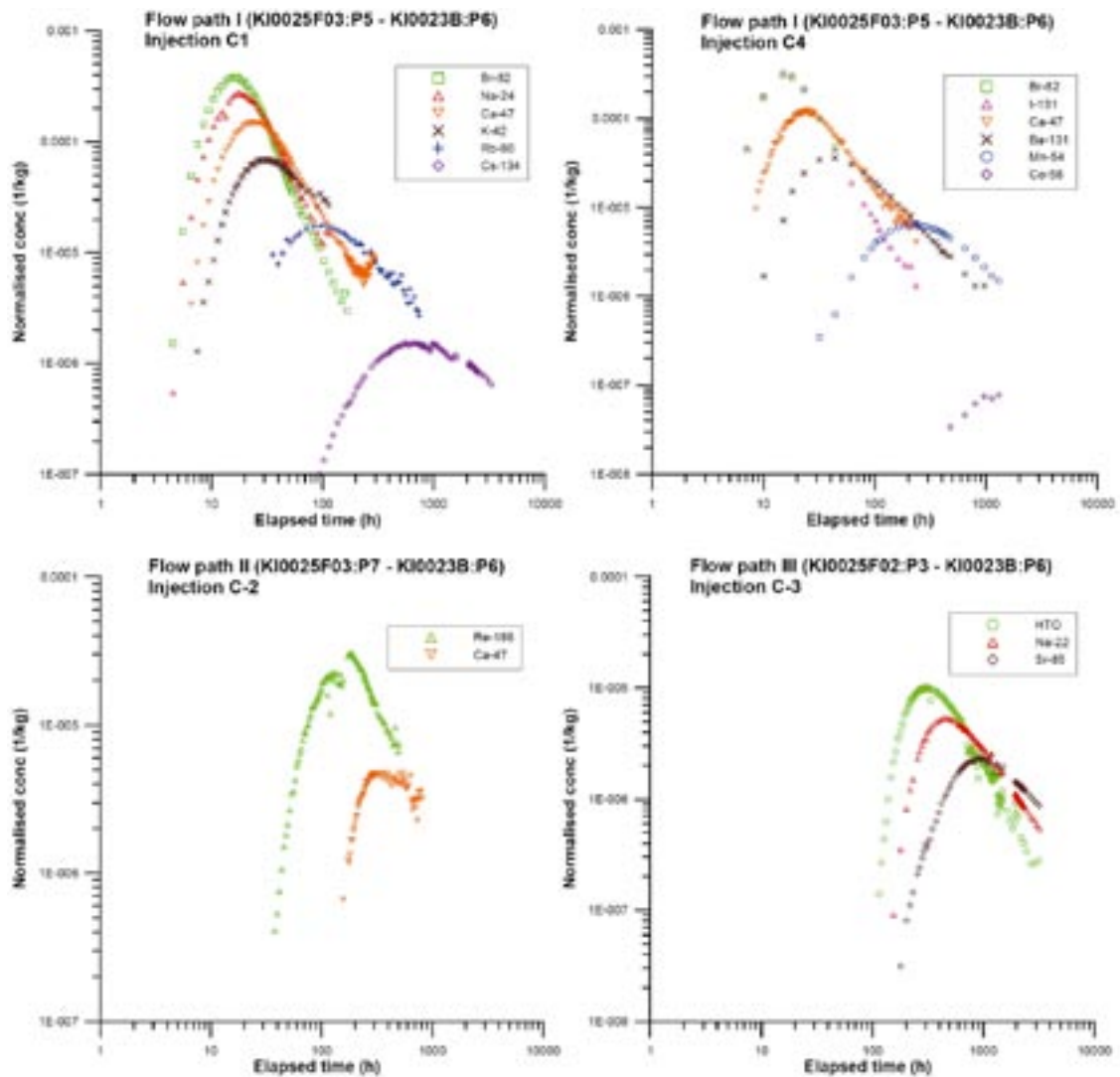
In TRUE Block Scale, techniques and methods for Site Characterisation were further refined. The large-scale network of hydraulically conductive structures was especially put in focus and extensive hydraulic testing was performed. The hydraulic testing, together with Phase A and Phase B tracer tests, gave much useful input to the elaborate hydrostructural model that was developed.

The microstructural model developed within TRUE-1, concerning the pore space and minerals within and around fractures, was further refined within TRUE Block Scale. Also the heterogeneity within a structure was demonstrated in the Site Characterisation, especially for structure #20 that was intersected by all boreholes drilled into the volume.

Phase C of the tracer test campaign gave information on how tracers are transported in fracture network, where a flowpath is constituted of a number of intersecting structures, potentially having different retention properties. Connectivity of the fracture network, where tracers needed to be transported across fracture intersection zones (FIZ), was demonstrated. From the result it appeared that transport in or across fracture intersection zones did not remarkably affect solute transport and retention, at least not on the spatial and temporal scales of the tracer tests. In the zones the hydraulic conductivity may be enhanced but on the other hand additional fracture infillings may give higher retention capacity.

Furthermore, clear evidence of retention was seen that was consistent with that seen in the earlier tracer tests carried out within TRUE-1. For example, in path 1 (used in C1 and C4) the relative order of retention was  $\text{Br}^- < \text{Na}^+ < \text{Ca}^{2+} < \text{Ba}^{2+} \approx \text{Rb}^+ < \text{Cs}^+$  (see Figure 3-25).





**Figure 3-25.** Breakthrough curves from the C1–C4 tracer tests in TRUE Block Scale. Image taken from /Andersson et al. 2001/.

TRUE Block Scale also provided insight into modelling of radionuclide transport. An interesting part of this was that different organisations (ENRESA, JNC, Nirex, Posiva, and SKB) made predictions of the breakthroughs of different tracers in the different Phase C tracer tests. Furthermore, TRUE Block Scale was a basis for modelling within the Äspö Task Force, which is discussed in the following subsection.

### 3.5.5 Äspö Task Force 6 – Understanding TRUE results

The evaluation of the results in the TRUE series of projects was made in close cooperation with the Äspö Task Force (short for The Äspö Task Force on Modelling of Groundwater Flow and Transport of Solutes). The Äspö Task Force was intended to provide a bridge between Site Characterisation and Safety Assessment approaches to solute transport in fractured rock. Two different tasks, Task 4 and Task 6, have had as their main focus the modelling of the TRUE series of projects. It should be carefully noted that Äspö Task Force performed no tracer test or investigation on its own in the field or in the laboratory.

Task 4 /e.g. Marschall and Elert 2003/ was carried out in 1995–2000 and consists of several modelling exercises in support of the TRUE-1 tracer tests, including predictive modelling where experimental results were not made available beforehand (i.e. blind predictions). As the TRUE-1 tracer tests were also revisited in Task 6, we refrain from summarising the finding of Task 4 and instead focus on Task 6 in this report.

The objectives of Task 6 were /Hodgkinson 2007/:

- To assess simplifications used in SA models.
- To determine how, and to what extent, experimental tracer and flow experiments can constrain the range of parameters used in SA models.
- To support the design of SC programmes to ensure that the results have optimal value for SA calculations.
- To improve the understanding of site-specific flow and transport behaviour at different scales using SC models.

The TRUE-1 tracer tests were revisited in Tasks 6A, 6B, and 6B2. In these tasks a number of modelling teams independently attempted to model solute transport in Feature A, based on the information obtained in SC and tracer tests. A review of these sub-tasks has been made by /Hodgkinson and Black 2005/, which is recommended for further information. A few of the major outcomes of the sub-tasks were the following:

- Even if Feature A had been extensively investigated in the SC of TRUE-1, the information delivered to the modelling teams left opportunity for different interpretations of the flowpath geometry. Entities of the flowpath geometry that were open for interpretations included the flow aperture, the degree of channelling, the degree of flow and connectivity in other fractures of the rock volume than Feature A, and the ratio of the flow-wetted surface to flow (or *F*-factor). The value assigned to the *F*-factor was especially identified as a major uncertainty. Moreover, there is currently poor understanding of how the *F*-factor might vary between experimental and SA timescales. The fact that all teams could calibrate their models with the water-residence time and obtain reasonable matches for the breakthrough data of the non-sorbing solutes demonstrated that the hydraulic data available were not sufficient to discriminate between the different conceptual models.
- A consensus was achieved about the general conceptual model for diffusion and sorption of tracers in the immobile zone. Furthermore, there was general agreement that short-term tracer tests only constrain the parameters of the most accessible pore spaces. Conversely, the rock matrix properties required for long-term SA are not significantly constrained by such tests and need to be measured independently.
- Concerning the usefulness of tracer tests it was concluded that as tracer tests only measure short-term behaviour, they should primarily be performed with conservative tracers to establish connectivity, water travel time and dispersivity, rather than to study radionuclide transport processes. However, tests using sorbing tracers have been important for understanding solute transport behaviour in and near fractures. They also reflect on differences in sorption partitioning coefficients obtained in the laboratory and in the field, but without answering the question what data should be used in SA.
- Concerning bridging the gap between SC and SA it was concluded that while laboratory and field characterisation data and associated SC modelling is necessary, it is rarely sufficient. Information or assumptions about long-term processes, for example from natural analogues, are also required.

Task 6C had as its focus the development of a “semi-synthetic” hydrostructural model /Dershowitz et al. 2003/ of the TRUE Block Scale volume and a review has been made by /Black and Hodgkinson 2005/.

Task 6D had as its focus, solute transport in the TRUE Block Scale volume. Task 6E extended the Task 6D transport calculations to a reference set of SA timescales and boundary conditions.

As one of the goals of the Tasks 6D and 6E was the harmonisation of SC and SA modelling, it was deemed that the hydrostructural model of the TRUE Block Scale rock volume produced in Task 6C should be considered to be a “correct” description of reality to be used by different modelling groups (i.e. applied without reinterpretation) in an attempt to understand differences in modelling approaches adopted by different modelling teams.

In Tasks 6F and 6F2, benchmark studies of a modelled singled fracture in the TRUE Block Scale rock volume were performed. Eight modelling teams representing five organisations participated in the exercise. A review of Tasks 6D, 6E, 6F, and 6F2 has been made by /Hodgkinson 2007/ to which the reader is directed for more information. A few of the major outcomes of these tasks, according to /Hodgkinson 2007/ were the following:

- As in Tasks 6A and 6B, it was found that the flowpath geometry was not sufficiently characterised prior to the modelling. Therefore, different modelling teams included channelling phenomena to different degrees. Even if there is evidence for channelling from Äspö and other sites, further research and investigations on the topic are needed before a consensus can be reached on how to implement channelling in transport models. Furthermore, it was identified that the ratio of the transport to hydraulic aperture is a crucial input for modelling solute transport.
- It was noted that the flowpaths of the TRUE Block Scale Phase C tracer tests were chosen based on good connectivity and large hydraulic conductivity that enabled good recovery. Such flowpaths are non-representative for flowpaths of interest for SA. Tracer tests performed in flowpaths of low hydraulic conductivity and connectivity can provide useful information, even if the recovery is reduced.
- For each structure constituting potential flowpaths, a Geological Structure Type was assigned. In addition, the structures were given complexity factors, where a single fracture was given a low complexity factor and a cluster of parallel or sub-parallel fractures (a fracture zone) was given a high complexity factor. Based on this information, different flow-wetted surfaces could be assigned to different structures. This was considered to be a major innovation of the exercise. However, it was also pointed out that the additional flow-wetted surface of a structure featuring closely spaced parallel fractures, may not behave identically on SA timescale as on the experimental timescale owing to saturation of the limited rock matrix. If the timescale is large enough to afford solute penetration depths of the same magnitude as the fracture spacing, some of the flow-wetted surface will become ineffective. It was also noted that some modelling teams assumed equal transmissivities of sub-fractures, even though it is more likely that one or a few fractures in a fracture zone dominate the advective transport.
- Concerning simplifications for SA modelling the following conclusions were drawn: Diffusion and sorption into intact rock are the primary retention processes on SA timescales. Diffusion and sorption into the immobile zone in direct contact with the flowpath provide secondary retention and can be approximated by a retardation factor on SA timescales. Geological Structure Type is a useful concept for classifying features, but does not have a first-order influence on SA.
- Concerning the use of tracer tests for constraining retention parameters for subsequent use in SA the following conclusions were drawn: Immobile zone parameters derived from tracer tests are not first-order contributors to SA retardation. However, tracer tests are invaluable for confirming our understanding of the dominant transport processes relevant for SA. Tracer tests provide useful information for quantifying transport aperture and flow wetted surface. All (even null) tracer test results, and also hydraulic (especially cross-hole) test results, can provide useful constraints on hydrostructural models.

The listed findings of Task 6 above are excerpts of all the findings of the task of particular interest for this present report, and we direct the reader to the original documents of Task 6 for more information, especially the reviews by /Hodgkinson and Black 2005, Black and Hodgkinson 2005, Hodgkinson 2007/.

### **3.5.6 TRUE-1 Continuation**

TRUE-1 Continuation involves complementary investigations at the TRUE-1 site, with the aim of exploring some of the unresolved issues from previous hydraulic investigations and tracer tests performed at the site. Field scale investigations of TRUE-1 Continuation involve, in the first stage, additional tracer tests /Andersson et al. 2002c/ with the aim of confirming or refining the TRUE-1 hydrostructural model. In a later stage injection of an epoxy resin into Feature A was to be performed with the aim of studying the small-scale flowpath geometry /SKB 2006f/. TRUE-1 Continuation also includes a number of other activities not performed in the field that are omitted in this report.

#### ***Tracer tests CX-1 to CX-5***

The tracer test campaign included three tracer dilution tests combined with pressure interference tests (CX-1 to CX-3) and two cross-hole tracer tests (CX-4 and CX-5) in arrays of multiple boreholes. The first three tests aimed at confirming, or delivering data for further refinement of, the TRUE-1 large-scale hydrostructural model. In CX-1, pumping was performed in a borehole section intersecting Feature A and pressure responses in 25 surrounding sections, at distances between 1 and 27 m, could be monitored. Within CX-1, tracer dilution tests were performed in 12 surrounding sections, both under “natural” gradients and during pumping. The measurements were repeated in CX-2 using the same set-up, with the exception that pumping was performed in a section intersecting Feature B. In the CX-3 tests, pumping was once more performed in a section intersecting Feature A. However, due to a higher pumping rate, pressure responses could be measured in 41 borehole sections, at distances up to 166 m. Tracer dilution tests were performed using the same setup as in CX-1 and CX-2, with the exception of the chosen pumping section. As in CX-1 and CX-2, tracer dilution tests were performed both with and without pumping.

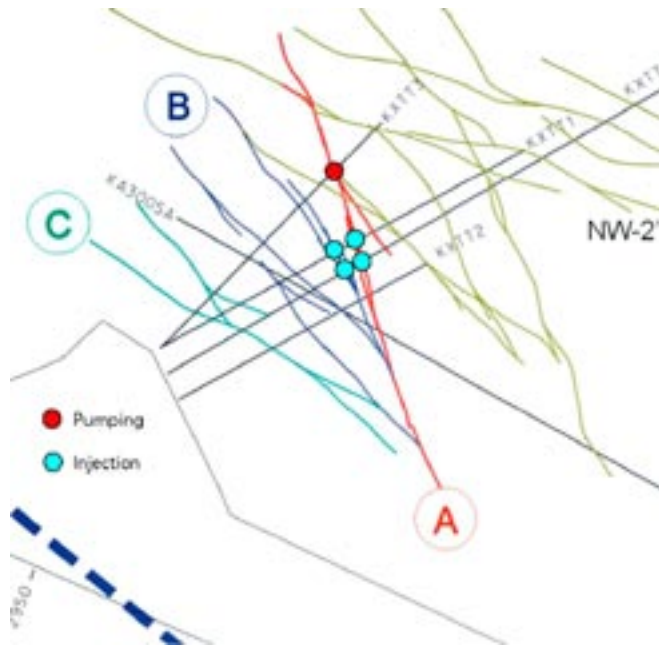
The CX-4 tracer tests aimed at investigating the reason for the dual-peak breakthrough obtained in previously performed tracer tests of STT-2 in Feature A. The hypothesis was that the injection section intersecting Feature A in STT-2 also contained a water conducting splay, denoted A', which gave rise to a second flowpath. The injection section was therefore separated in two sections in CX-4 by installing a short extra packer. In one section Uranine was injected while in the other Amino-G acid was injected. By performing the tracer test the double flowpath hypothesis could be strengthened, as significant peak separation was obtained. However, it should be noted that compared to the STT-2 test less flow between the injection and pumping sections could be achieved when using the same drawdown. This indicates that the hydraulic characteristics of the flowpaths had changed during the years intervening between STT-2 and CX-4. This is considered to be an important result in itself.

Test CX-5 was performed with the purpose of assessing connectivity between Feature B and A. Four sections were used for passive injection; two in Feature A and two in Feature B. Pumping was performed in a section intersecting Feature A at a distance between 4 and 5 m from the injection points (see Figure 3-26).

The non-sorbing tracers used were Uranine, Amino-G acid, and Rhodamine WT. Monitoring and sampling in the pumping section were performed for 650 hours and during this period, only breakthroughs from the injections in Feature A were obtained. This suggested that even if Features A and B may be connected, as indicated from pressure interference tests, short-range connection between the features by means of a three dimensional network of minor fractures in the rock mass sandwiched by the features is not likely (or at least the connection is poor).

#### ***Epoxy injection and pore space characterisation***

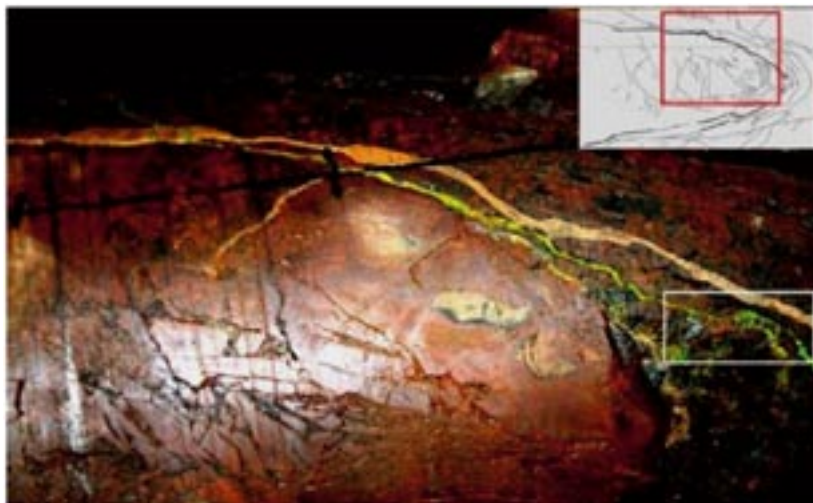
Within the TRUE-1 continuation project there is a sub-project, TRUE-1 Completion, which aims at; 1) performing injection of an epoxy resin into Feature A, 2) performing overcoring and 3) performing subsequent analysis of the injected resin and thus also the pore space geometry of Feature A. Presently the resin injection and overcoring are preformed but results from the analysis are pending. As a result of the overcoring the site has become disturbed (destructively dismantled) to the extent that no further investigation is intended to be made.



**Figure 3-26.** Set-up of the CX-5 tracer test. Image taken from /Andersson et al. 2002c/.

Additional resin injections have been performed within TRUE-1 Continuation, but at an access tunnel and not at the TRUE-1 site. Within the Fault Rock Zones Characterisation campaign /Hansen and Staub 2004/, 16 pilot boreholes with a diameter 76 mm were drilled into candidate structures. By performing BIPS-loggings and hydraulic testing, candidate structures in 7 of these boreholes were chosen for epoxy-resin injection. The technique for performing the injection is described in /Hansen and Staub 2004/. The epoxy resin used had Uranine additive to facilitate subsequent analysis.

Upon hardening of the resin, overcoring with boreholes of 300 mm diameter was performed. 4 of the 7 holes produced cores of high integrity while the remaining 3 produced poor results. After the overcoring the cylindrical surfaces of the cores were covered with transparent film, to facilitate the subsequent analysis. Figure 3-27 shows a structure where some fractures have been filled with the resin (with a greenish colour).



**Figure 3-27.** Drill core with fractures filled with epoxy resin of greenish colour. Image taken from /Hansen and Staub 2004/.

By performing image analysis, valuable information on fracture aperture and the connected pore space geometry could be obtained.

### **SWIW-tests**

Complementary tracer tests were performed at the TRUE-1 site using SWIW (Single-Well Injection Withdrawal) technique. The technique is discussed in subsection 3.6.1 of this report. Except for the injection in section KXTT4, using non-sorbing tracers, four surrounding sections were used for observation. The results show that tracers were propagated in Feature A over distances between 5–10 m during the test. However, when withdrawing the tracer only a slight recovery, 3%, was obtained that was much lower than expected. An explanation offered for the low recovery is the occurrence a larger background hydraulic gradient in Feature A than expected /SKB 2006f/.

### **CEC-test**

As part of TRUE-1 Completion, an attempt to assess the CEC (Cation Exchange Capacity) of fracture surfaces is underway by way of performing an in situ cross-hole tracer test /SKB 2007a, SKB 2005d/. The idea is to continuously inject a solution with relatively high non-radioactive  $\text{Cs}^+$  concentration into Features A and A' and thus saturate the surface sites with  $\text{Cs}^+$ . In the test, tracer injection has been made in borehole KXTT4 with withdrawal in KXTT3 (see Figure 3-23). With the aid of modelling interpretation of the breakthrough results the aim is to assess the CEC capacity of the fracture surfaces. An additional aim is to analyse the withdrawn groundwater for desorbed  $^{137}\text{Cs}^+$  and  $^{134}\text{Cs}^+$ , previously injected within TRUE-1. This is an ongoing project and no results have been reported at this time.

### **Major achievements**

In general, the additional tracer tests confirm the existing hydrostructural model presented in /Winberg et al. 2000/. From the CX-4 tracer test clear indications of separate flow paths hosted in splay fractures to Feature A were obtained, which was an initial hypothesis. From the CX-5 tracer test the hydraulic units Feature A, Feature B+D, and Feature NW-2 seemed to be well separated with respect to transport paths on the TRUE-1 scale, which weakened the modelling assumption of a three dimensional flow in the TRUE-1 tracer tests.

The aim of the epoxy resin injections is to gain insight of the pore geometry of the TRUE-1 site and, in particular, Feature A. Within the TRUE Continuation phase, epoxy resin injection has been performed at the Äspö HRL, which has resulted in technological and methodological improvements to the method and also strengthened the pre-existing conceptual model of pore space heterogeneity.

### **3.5.7 TRUE Block Scale Continuation**

The overall objective of TRUE Block Scale Continuation (BS2) was to improve understanding of transport pathways at block scale, including assessments of effects of geometry, macro-structure and micro-structure /Andersson et al. 2007/. Special focus was put upon transport in background fractures of low hydraulic conductivity. BS2 was divided into two separate phases. The first phase, BS2A concerned complementary modelling, which is not discussed in this chapter, and some additional hydraulic testing. The second phase, BS2B, concerned among other things additional field scale tracer tests /Andersson et al. 2007, SKB 2006f/.

## BS2B

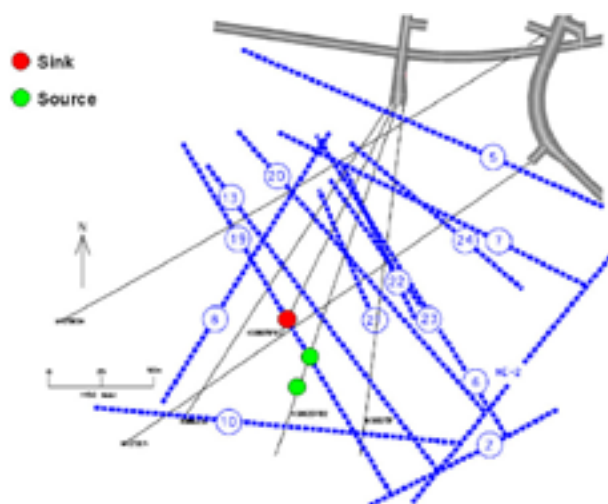
The aim of the BS2B phase was to perform tracer tests over longer distances with sorbing tracers. The tracer tests were to be performed within or nearby Structure #19 of the TRUE Block Scale site (see Figure 3-28). This included investigating minor fractures connected to or sub-parallel to Structure #19.

Prior to the tracer tests using sorbing and radioactive tracers, pre-tests were made with Uranine, Amino-G acid, and Rhodamine WT with the objective of localising suitable sinks and sources, as well as flowpaths with suitable recoveries and groundwater residence times /Andersson et al. 2004/. The pre-test sequence included a series of pressure interference tests combined with tracer dilution tests (CPT-1 to CPT-3) that generally confirmed the existing hydrostructural model presented in /Winberg et al. 2003/. Based on the results, one section was suggested as a sink and six sections were suggested as sources for the subsequent cross-hole tests. In CPT-4a, CPT-4b, and CPT-4c nine tracer tests were performed along the six suggested flowpaths. The tracer tests were performed either as radially converging tests with passive (decaying pulse) injections or as unequal dipole tests.

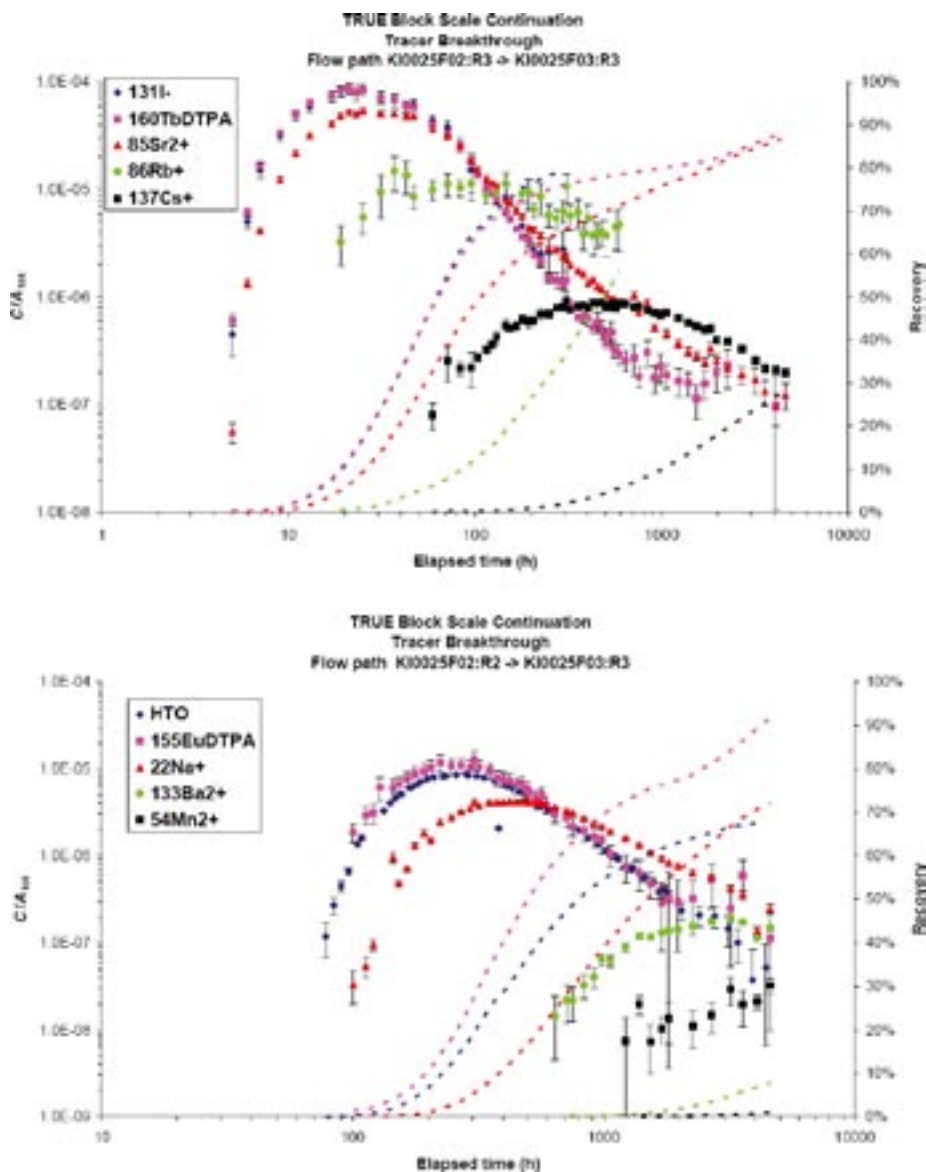
Based on the earlier investigations, a sink and two sources were suggested for the subsequent tests using sorbing tracer. In both of these flowpaths more than 80% recovery had been obtained with conservative tracers. One of the flowpaths (KI0025F02:R3 → KI0025F03:R3) was in the single fault structure #19, had a shorter residence time, and a length of about 19.5 m. The other flowpath (KI0025F02:R3 → KI0025F03:R3) involved a background fracture connected to structure #19, had a longer residence time, and a length of about 22 m.

For the shorter flowpath the non-sorbing tracers  $^{131}\text{I}^-$  and  $^{160}\text{Tb-DTPA}$  were used together with the sorbing tracers  $^{85}\text{Sr}^{2+}$ ,  $^{86}\text{Rb}^+$ , and  $^{137}\text{Cs}^+$ . For the longer flowpath the non-sorbing tracers HTO and  $^{155}\text{Eu-DTPA}$  were used together with the sorbing tracers  $^{22}\text{Na}^+$ ,  $^{133}\text{Ba}^{2+}$ , and  $^{54}\text{Mn}^{2+}$ .

For the non-sorbing and weakly sorbing tracers, high recovery (68–92%) was obtained for both flowpaths while for the moderately sorbing tracers 56% of the  $\text{Rb}^+$  was recovered in the shorter flowpath but only 8% of  $^{133}\text{Ba}^{2+}$  in the longer flowpath within the experimental time. For the strongly sorbing tracers 28% of  $^{137}\text{Cs}^+$  and 1% of  $^{54}\text{Mn}^{2+}$  were recovered. Figure 3-29 shows breakthrough curves and recoveries for the two tracer tests.



**Figure 3-28.** The TRUE Block Scale site with interpreted structures and sink and sources of BS2B. Image taken from /Andersson et al. 2005/.



**Figure 3-29.** Breakthrough curves and recoveries in BS2B for the short (upper) and long (lower) flowpaths. Image taken from /Andersson et al. 2005/.

### Major achievements

In the BS2B tracer tests, clear retention could be seen. The following retention factors were obtained /Andersson et al. 2007/:

- $^{85}\text{Sr}^{2+}$  1.1–1.25
- $^{22}\text{Na}^{+}$  1.3–1.5
- $^{86}\text{Rb}^{+}$  2.9–3.2
- $^{133}\text{Ba}^{2+}$  4.1–4.5
- $^{54}\text{Mn}^{2+}$  6.4–7.4
- $^{137}\text{Cs}^{+}$  12.8–13.2

It was shown when fitting models to the breakthrough curves that matrix diffusion was a significant retardation process in the tests. The breakthrough curves could be fitted relatively well with the advection-dispersion model using matrix diffusion and linear sorption, but not



with the advection-dispersion model disregarding matrix diffusion. Compared to the Phase 3 tracer tests of TRUE Block Scales, many similarities could be observed. However, the retardation was smaller for the weakly sorbing tracers  $\text{Sr}^{2+}$  and  $\text{Na}^+$  in BS2B.

In TRUE Block Scale Continuation it was verified that background fractures in the rock mass have lower retention material properties than fault type structures, which is consistent with the existing microstructural model. On the other hand, the flow in background fractures is generally lower which enables greater contact times and thus the overall retention was higher in the background fractures compared with the fault-type structure in the campaign. It was concluded that background fractures may contribute significantly to solute retention on the block scale.

### 3.5.8 Long Term Sorption Diffusion Experiment – LTDE-SD

Presently SKB is conducting an experiment at the Äspö HRL called LTDE-SD (Long Term Sorption Diffusion Experiment) /Byegård et al. 1999, SKB 2007a/. The tracer test is performed in the vicinity of a hydraulically conductive fracture at a depth of 410 m below sea level in the Äspö HRL. The specific objectives of LTDE-SD are /SKB 2007b/:

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

A small-diameter borehole ( $\varnothing = 36$  mm) was drilled from the Äspö HRL tunnel into the bedrock to intersect a hydraulically conductive fracture, some 10 m from the tunnel wall. The small-diameter borehole penetrates about 11.8 m beyond the fracture plane (see Figure 3-30).

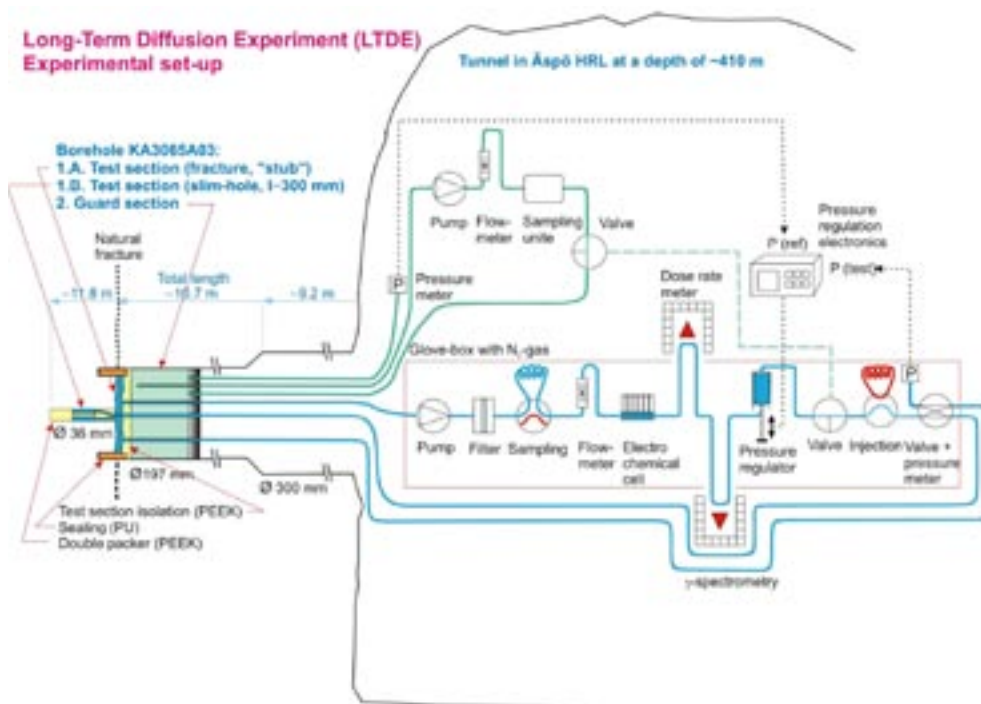


Figure 3-30. Sketch of the LTDE-SD experiment. Image taken from /SKB 2007a/.

The small-diameter borehole was overcored by a large-diameter borehole drilled just long enough to intersect a hydraulically conductive fracture. In the process of doing this, the large-diameter borehole penetrated through the fracture plane by a short distance resulting in a stub at the fracture surface (see Figure 3-31).

In the tracer test the stub is sealed off with a polyurethane cylinder and a peek lid, which contains a “cup-like” packer (see Figure 3-31). Within this reservoir the tracer solution circulates. The remainder of the large-diameter borehole is packed-off with a system of one mechanical and two inflatable packers (see Figure 3-30) in order to simulate the natural pressure at the fracture surface.

The first few meters of the small-diameter borehole that penetrates the fracture plane is sealed-off by a tube, restricting cylindrical diffusion in the stub. From the 3 m long end section of the small-diameter borehole, which is connected to the tracer reservoir, tracers are intended to diffuse into the undisturbed rock matrix.

The tracer solution consists of 22 different tracers, with sorption properties ranging from non-sorbing ( $^{36}\text{Cl}$ ,  $^{35}\text{S}$ ) to strongly sorbing ( $^{175}\text{Hf}$ ,  $^{236}\text{U}$ ). The in situ part of the experiment should be carried out for 5–7 months, whereafter the stub and small-diameter borehole will be overcored, sectioned, and analysed. During the experiment some groundwater chemistry parameters are closely monitored (pH, Eh), as one of the objectives is to maintain natural groundwater chemical conditions. However, from the initial results it has become evident that one can not keep the redox potential at negative values in the test section.

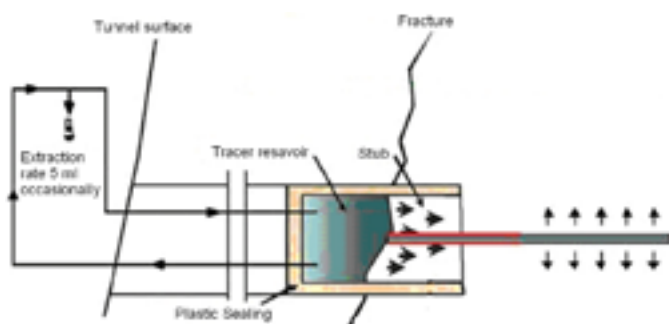
The in situ experiment is supported by laboratory experiments on core specimens from the experimental borehole and various types of mineralogical, geochemical and petrophysical analyses.

### **Expected achievements**

At the time of the writing of this report, no significant data has been presented from the ongoing LTDE-SD campaign. However, there are some expected achievements.

According to project plan of LTDE-SD /Gustafsson 2006/, the project is expected to provide quantitative and qualitative information on sorption and matrix diffusion under in situ conditions, i.e. natural stress, hydraulic pressure and groundwater composition, and is expected to demonstrate the validity of the coupled sorption-diffusion processes adopted in SA modelling.

Here we take the liberty to make some comments of potential value for the subsequent evaluation of LTDE-SD. As for all tracer tests, even if great care is taken to simulate “natural conditions” as closely as possible there will always be deviations from such and the system should be considered to be more, or less disturbed. Such deviations must be considered in future evaluations of the LTDE-SD tracer test and the potential effect on the results must be accounted for. Two issues, possibly out of many, that should be considered are: 1) the rock matrix in the stub will to some extent be de-stressed or mechanically disturbed as a result of the drilling.



**Figure 3-31.** The stub of the LTDE-SD experiment. Image modified from /SKB 2007a/.

This also applies to the rock in the immediate surroundings of the pilot borehole. For most sorbing species, only this rock volume is expected to be utilised for sorption within the experimental time frame, 2) the hydrogeochemical situation is likely to be disturbed as a result of the Äspö tunnel, the borehole, and the tracer test itself. This may affect minerals at the fracture surface of the stub (e.g. fracture coatings) and potentially also sorption partitioning coefficients.

### 3.6 Tracer tests within the site investigations

#### 3.6.1 Single-Well Injection-Withdrawal – SWIW

The Single-Well Injection-Withdrawal (SWIW) tracer test is a relatively new method with the estimation of in situ retention parameters as its main aim. In the site investigations, SWIW tracer tests are performed in boreholes drilled from ground surface. Figure 3-32 shows the set-up of the experimental equipment.

The SWIW test is divided into six different phases subsequent to tool emplacement (i.e. after the packer system surrounding the hydraulically conductive fracture or fracture zone of interest is established).

- In the first phase, groundwater is pumped from the fracture, up through a hose in the borehole and into an airtight vessel at the surface. The vessel headspace is filled with nitrogen gas so that the redox potential of the groundwater is not changed.
- In the second phase, the pre-injection, native groundwater is pumped back into the fracture system. During this phase the pressure stabilises and the hydraulic situation is allowed to reach steady state. This phase may take from a few hours to several days depending on the transmissivity of the test section.

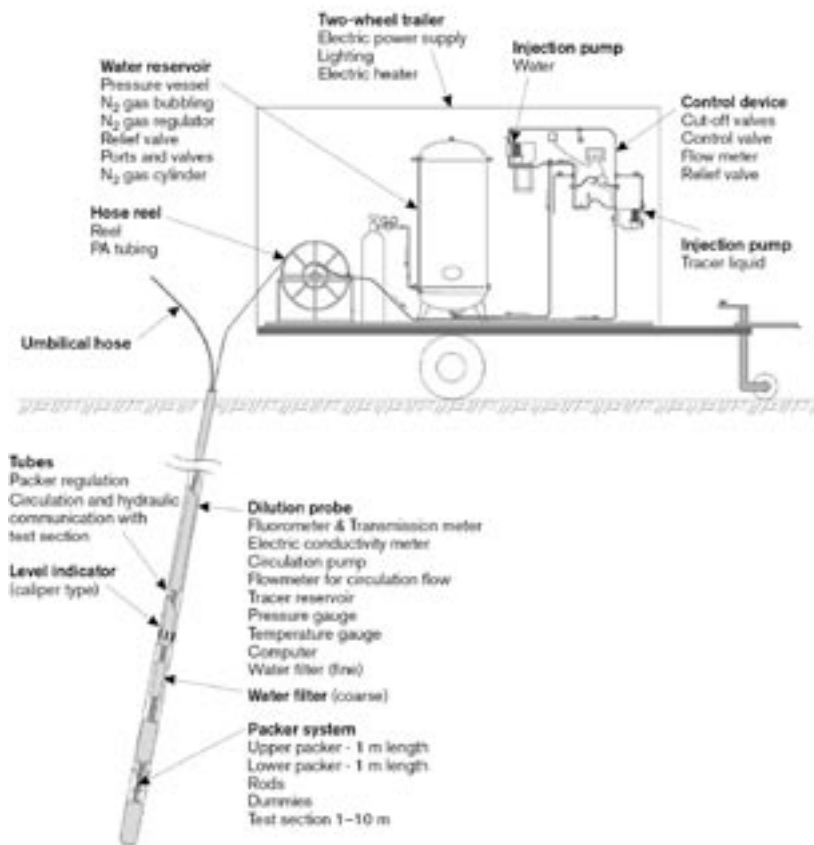


Figure 3-32. Experimental set-up of the SWIW tracer test. Image taken from /Gustafsson and Nordqvist 2005/.

- In the third phase, tracers are injected at ground surface into the hose leading from the vessel down to the packed-off sections and are thereafter carried with the injected groundwater into the fracture. The tracer injection may be carried on for an hour or so.
- In the fourth phase, tracer injection is stopped and native groundwater is used to flush the hose and packed-off section, so that all tracer residues are forced into the fracture system. Furthermore, the flushing is carried on for long enough time so that the tracer-spiked groundwater reaches a suitable distance into the rock. This flushing may be carried on for many hours or even for a few days.
- The fifth phase, which may be omitted, is the waiting phase. In some cases the waiting phase may be short, only a few hours, even if longer waiting phases would be preferable. An important factor limiting the extent of the waiting phase is the natural flow through the fracture. If the flow is large and the waiting phase is too long, the tracers will be carried too far from the borehole resulting in a small subsequent tracer recovery.
- In the sixth phase, the tracer spiked groundwater is withdrawn and pumped up the borehole to the surface. An automatic sampler at ground surface is used to take water samples for analysis of tracers, for example Uranine and Cs, in the withdrawn water. The withdrawal may be carried on for many days.

SWIW testing has been performed in a number of boreholes both within the Forsmark and Oskarshamn site investigations. As an example Figure 3-33 shows recovery phase breakthrough curves for Uranine, Cs<sup>+</sup>, and Rb<sup>+</sup> from a run at a section in KFM04A in Forsmark.

As can be seen from the x-axes in Figure 3-33 the SWIW test was carried out for several hundred hours. Tracer recovery was 42.6%, 53.9% and 32.8% for Uranine, Rb<sup>+</sup>, and Cs<sup>+</sup>, respectively. In this particular SWIW test a retardation factor for Rb<sup>+</sup> ranging from 2.9 to 5.5 was obtained.

As can be seen in /e.g. SKB 2007a, SKB 2006e/ the SWIW equipment has also been used to inject and withdraw tracers in single-hole or cross-hole tracer tests at, for example, Äspö HRL.

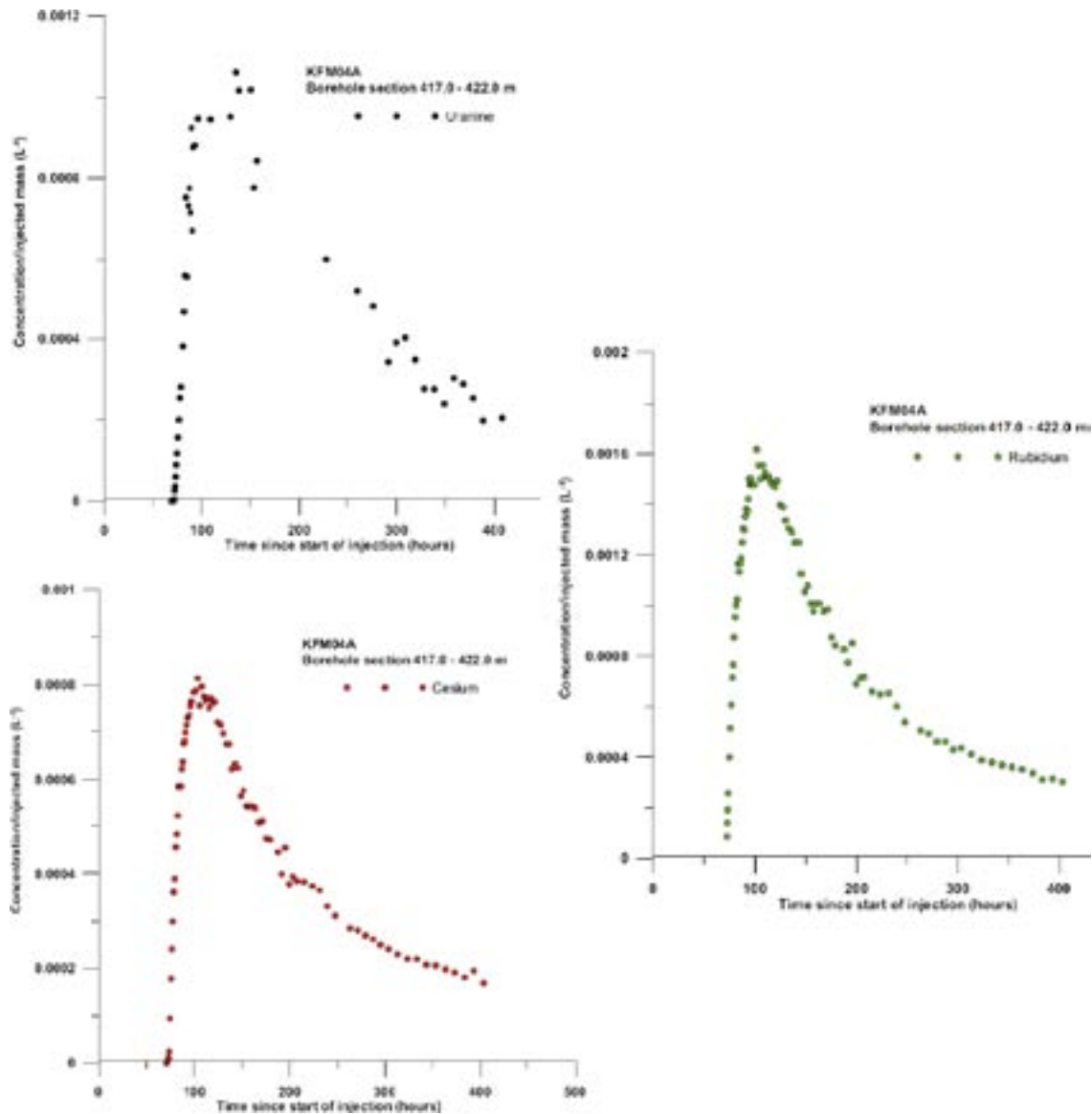
### **Major achievements**

Evaluating results from SWIW test has proven to be a difficult task. Although they deliver qualitative indications of retention, there is a debate on how to robustly extract quantitative retention data from the results. One major reason is that there are numerous processes involved affecting the breakthrough curve in similar ways and that it is difficult to discriminate to what extent the different processes have affected the result. Presently an in-depth publication is being prepared /Neretnieks 2007/ with the aim of surveying the flow and transport mechanisms and processes involved in SWIW tests and the possibility of extracting quantitative retention data from them.

### **3.6.2 Formation factor logging in situ**

By using the physical analogy between diffusion and electromigration, a recently developed method of obtaining diffusive properties of the rock matrix by electrical resistivity measurements has been implemented in the SKB site investigation programme /Löfgren and Neretnieks 2002/. Such measurements are performed in situ from boreholes drilled from ground surface, as well as in the laboratory on drill core samples /e.g. Löfgren and Neretnieks 2005/. Natural tracers are used (ionic groundwater solutes) although it should be noted that while the electric current is propagated over large distances in the rock matrix, the ionic tracers themselves only migrate over minute distances, as alternating current is used in the method.

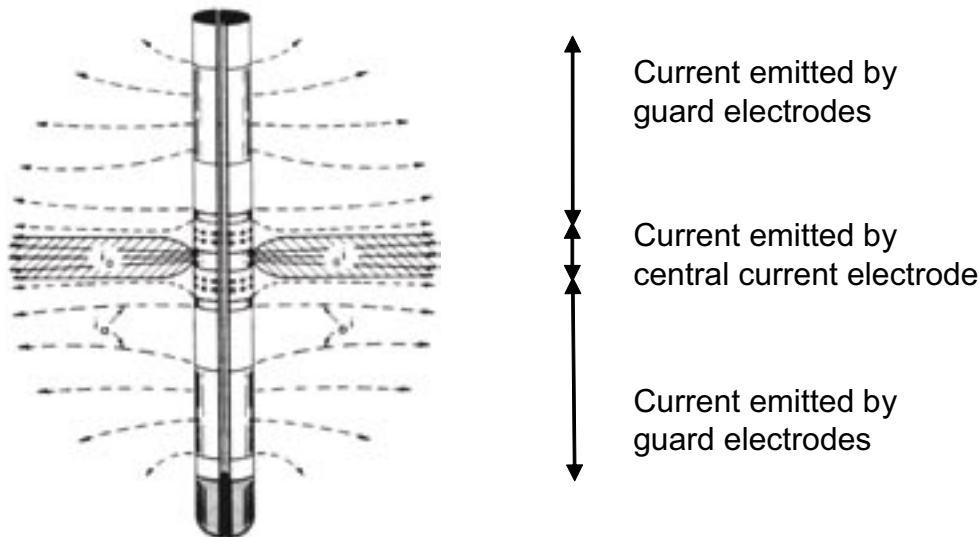
In the in situ method, the resistivity of the rock matrix is measured using standard geophysical logging tools. From such a tool, an alternating current beam is sent out many meters into the rock matrix, perpendicular to the borehole. Figure 3-34 shows an illustration of such a tool and its electrical current field, where a current beam is sent out from the central current electrode.



**Figure 3-33.** Example of SWIW recovery phase breakthrough curves from KFM04A. Images taken from /Gustafsson et al. 2006a/.

By using guard electrodes on each side of the central current electrode, which also emit electrical fields, the shape of the electric field from the central current electrode can be controlled. The current sent out from the electrodes is propagated by ionic solutes in the saturated microporous system of the rock matrix as the mineral grains themselves are too resistive to contribute significantly to the propagation of current. The resistance to propagation of ionic solutes by electromigration through the porous system is proportional to the resistance to diffusive transport through the rock.

By making assumptions concerning the electrical conductivity of the pore water, a proportionality factor for transport through the rock matrix relative to transport through free phase solution can be obtained. The same proportionality factor, the so-called formation factor (see subsection 2.4.2), applies for diffusive transport. The assumption made concerning the pore water is that it is reasonably well equilibrated with, or at least has similar electrical conductivity to, the free groundwater flowing in fractures intersecting the rock at a corresponding depth.



**Figure 3-34.** Illustration of a focused rock resistivity tool. Image modified from figure in /Desbrandes 1985/.

The electrical conductivity of groundwater flowing in fractures can be measured with standard methods after withdrawing it from packed-off sections within boreholes. This also assumes that the sampled water is representative of the “actual” groundwater and is not contaminated by non-representative borehole water from drilling or surface drawdown.

Recently it has been shown that the assumptions made concerning the electrical conductivity of the pore water, based on measurements on freely flowing groundwater, are sufficiently adequate. This has been done by comparing the assessed electrical conductivities with chemistry data obtained on the matrix fluid of drill core samples /e.g. Waber and Smellie 2005/.

By moving the geophysical tools up or down the boreholes, formation factor logs can be obtained. At present, such formation factor logs have been obtained in 20 core drilled boreholes within the site investigations. The boreholes are KFM01A, KFM01D, KFM03A, KFM04A, KFM05A, KFM06A, KFM07A, KFM08A, and KFM08C in Forsmark; KSH01A and KSH02 in Simpevarp; KLX02 (only minor part), KLX03, KLX04, KLX05, KLX06, KLX07A, KLX08, KLX10, and KLX12A in Laxemar.

### **Major achievements**

This method delivers formation factors that directly relate to the effective diffusivity of solutes in the rock matrix. The number of data points obtained by this method is on the order of 100,000, which makes the formation factor by far the most well characterised retention parameter in the site investigations.

A prerequisite for the method to work is that the microporous system is connected over large distances (many meters) in situ. This method gives, together with complementary laboratory work /Löfgren and Neretnikes 2006, Ohlsson 2000/, very clear indications that this is indeed the case.

A drawback of the method is that only rock at some distance from open fractures can be sampled, as free phase groundwater in fractures disturbs the measurements.

### 3.6.3 Large-scale tracer test in Forsmark at drill site 1

In 2005, a large-scale cross-hole tracer test was carried out within the Forsmark site investigation area /Wass and Andersson 2006/. The aim of this study was to increase the understanding of the connection between hydraulic structures in the area rather than to obtain retention parameters. The radially converging tracer test was performed in boreholes drilled from ground surface, where withdrawal was made in a central borehole and tracer injection was made in packed-off sections in three surrounding boreholes. The sketch in Figure 3-20 serves as an illustration of the type of tracer test carried out.

Before performing the tracer test, hydraulic structures in the boreholes were identified by tracer dilution tests /Wass and Andersson 2006/. Thereafter, the non-sorbing tracers Amino-G acid, Uranine, and Rhodamine WT were released as passive decaying pulses from packed-off sections in the boreholes HFM02, KFM01A, and HFM15, respectively.

The tracers were withdrawn from the entire borehole HFM01 (see Figure 3-35) for 380 hours following tracer injection. This relatively short withdrawal time resulted in an absence of tracer breakthrough from borehole KFM01A and HFM15. A breakthrough of Amino-G acid from borehole HFM02 was obtained although the study was terminated before the peak had arrived, thus rendering any transport modelling highly speculative. The distance between HFM02 and HFM01 is 222 m and the tracer was injected at a depth of about 38–48 m.

#### Major achievements

This tracer test suffered from the tight time schedule only allowing for 380 hours of withdrawal time. However, the fact that breakthrough was achieved between boreholes HFM02 and HFM01, at a distance of 222 m, indicate that the utilised structure or structures are very conductive.

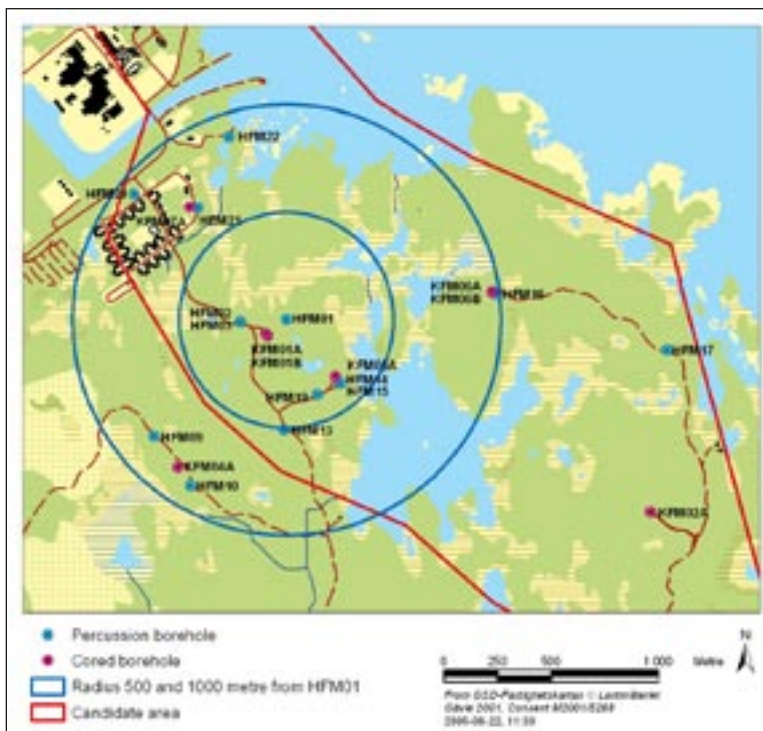


Figure 3-35. Forsmark area with utilised boreholes. Image taken from /Wass and Andersson 2006/.

### **3.6.4 Large-scale tracer test in Forsmark at drill site 2**

Presently the evaluation of a large-scale cross-hole tracer tests at drill site 2 in Forsmark is underway. The tracer tests were carried out in the major fracture zone A2 at Forsmark where borehole KFM02A was used for tracer injection and borehole KFM02B was used for tracer withdrawal and pumping /SKB 2007b/. An unequal dipole flow configuration was used where injection was performed using only a slight overpressure. The distance between the boreholes is about 50 m.

The tracer tests were preceded by pressure interference testing in zone A2 and based on hydrogeological information, injection and withdrawal sections were chosen at about 400 m depth. Before initiating the tracer test with sorbing tracers, a pre-test was made using non-sorbing Rhodamine WT. The tracers used in the subsequent test were Uranine, Li<sup>+</sup>, Rb<sup>+</sup>, and Cs<sup>+</sup>. Sampling and monitoring was carried out for several hundred hours.

At the time of writing this report no data from the tracer test campaign have been reported.

### **3.6.5 Large-scale tracer test in Forsmark 3**

Presently a third large scale tracer test within the site investigations in Forsmark is underway.

### **3.6.6 Large-scale tracer test in Laxemar**

A large scale tracer test has been performed in Laxemar within the site investigations /Gustafsson and Ludvigson 2005/. As no tracer breakthrough was achieved, this test is only described in the briefest way in this present report. However, as even the “no-show” of tracers may give some information the following is mentioned:

The objectives of the activity were to assess the hydraulic connectivity between boreholes KLX02 and HLX10 and to determine transport properties of the flowpaths involved, in the case of a tracer breakthrough. Pumping was performed in borehole HLX10 and the non-sorbing tracer Rhodamine WT was injected in borehole KLX02. The distance between injection and withdrawal sections was about 260 m. The tracer withdrawal was carried on for about three weeks but out of these, the pumps were non-functioning for one week.



## 4 The constraining power of tracer tests

### 4.1 Introduction

In this chapter we discuss the interpretation of tracer tests and, in particular, what kinds of information that can be gained from such tests. At the end of the chapter, possibilities and limitations of tracer tests are tabulated. This table can be used as a short guide of relevant issues to be considered by designers of future field scale tracer tests.

The number of references to specific SKB tracer tests is intentionally kept low in this chapter, as such are already given in given in plenitude in Chapter 3. Instead this chapter focuses on conceptual and modelling issues and refers to a larger extent to the open literature.

Solute transport in geological formations is characterised by complexity on many different levels. The features and mechanisms underlying this complexity and their consequences for solute transport can be explored by modelling in conjunction with field-scale and laboratory experiments. A number of different modelling approaches are used to predict the transport and retention of radionuclides in the geosphere. The different approaches have a lot in common, especially concerning the process description, but also exhibit a number of differences. The choice of modelling approach is to a large extent determined by the objectives and scope of the modelling.

For Safety Assessment (SA) purposes we are interested in reducing the complexity of the system description sufficiently that well-reasoned estimates of risk can be made. Therefore, SA only seeks to capture the main physical and chemical properties of the geological formation. In this context, highly sophisticated models of radionuclide transport and retention, while useful from the point of view of process understanding, may be less relevant for SA purposes.

For Site Characterisation (SC) and Site Descriptive Modelling (SDM), on the other hand, we are interested in understanding the full range of behaviour observed in the natural system. SDM's are multidisciplinary interpretations of the investigated site covering its geology, hydrogeology, hydrogeochemistry, transport properties, etc. For the SDM, it is important to include as much detail as is reasonably possible, even if not relevant for present SA scenarios, in order to provide the possibility for inclusion in subsequent SA scenario modifications. It is also emphasised that in order for a process or system attribute to be downgraded in importance in the SA, it must be sufficiently well characterised and understood that this can be rationalized in a robust fashion.

When interpreting field scale tracer tests, various transport models may be used where the aim is typically to include as much detail and complexity as is reasonably possible. This degree of complexity may be necessary for accurate extrapolation and transferral of results to the SA and SDM. Therefore, substantial efforts are often made to characterise physical and chemical properties of the location of interest before initiating the tracer test. Problems arising with extrapolation of data to SA are often associated with differences in temporal and spatial scales. Problems arising with transferral of data to the SDM may occur as tracer tests are often carried out at locations and under conditions that are atypical for the site.

A full discussion of all the relevant issues associated with modelling of tracer tests and abstraction of data relevant for SA is beyond the scope of this report. For a more detailed discussion of these issues, the reader is directed to the review articles by /Selroos et al. 2002, Berkowitz 2002/ on the subject of hydrogeological modelling of fractured crystalline rock, /Lichtner et al. 1996, Bryant and Thompson 2001, SKB 2004/ for discussions concerning the coupling of transport and reactive chemistry, and /Jenne 1998/ on solute partitioning (mostly sorption). The significant advances in mechanistic sorption modelling that have occurred in the last decade and their use in both SC and SA modelling is also chronicled in some detail in the reports produced within the OECD/NEA Sorption Project /NEA 2001, 2005/.

## 4.2 Interpretation of data at different levels of complexity

Given the level of complexity associated with transport processes that may occur over the timescale of a field scale tracer test, we consider the utility of tracer tests in the context of evidence-based interpretations of data which we characterise here in the form of a sequence of questions of increasing complexity:

- a) *Do the data confirm the extent of connections and identify transport paths between the tracer release and recovery locations?*
- b) *Do the data lend qualitative support to the existence of retention for interacting solutes?*
- c) *Do the data obtained in any way confirm or contradict our process understanding or retention paradigm?*
- d) *Are the data sufficiently well resolved that we can obtain estimates of lumped retardation parameter groups for the specific tracer test and solute within the experimental time frame?*
- e) *Is it possible to distinguish the material properties and flow-related transport parameters, (i.e. MPG and F-factor) in a robust fashion from the experimental data? (Are these estimates physically realistic in the light of independently obtained data? Can these be further decomposed into physically realistic values for their component parts? e.g.  $D_e$ ,  $K_{dp}$ , FWS,  $Q$ , etc.)*
- f) *Is it possible to extrapolate these data to other timescales or flow configurations without introducing additional assumptions that cannot be substantiated?*

Of the questions listed above, a) and b) can be answered without recourse to modelling. Questions c)–f), on the other hand generally require some form of transport modelling in order to answer.

In the following sections we will attempt to discuss the questions listed above in the light of the knowledge evolved from field scale tracer tests that SKB has carried out over the past few decades. For questions a)–c), the discussion is very much based on the SKB tracer tests described in Chapter 3, even if we do not intend to discuss each question in the light of every single tracer test. We have also refrained from excessively repeating information already given in the “major achievements” subsections of Chapter 3. For question d)–f) the discussions become more and more philosophical as the level of complexity increases, and references are to a larger extent made to the open literature and to a lesser extent made to SKB reports and experiments.

A general trend of the SKB field-scale tracer tests is that the more recent they are, the more ambitious they become in the context of obtaining retention parameters. This level of ambition does not only reflect on the attention to detail in the experiments themselves, but also on the reporting and the manner in which results are critically reviewed and concerns brought to light. Many of the earlier tests were primarily intended to assist the development of methods and methodologies for tracer testing.

We have to a greater extent used earlier tracer tests to answer less complex questions, or aspects of questions, and more recent tracer tests to answer more complex questions. In some cases this may give the reader the impression that earlier tracer tests provide more reliable and detailed answers than more recent ones, and that we value them higher. We wish to point out that recent tracer tests may just as well provide answers to less complex questions. Furthermore, we wish to underline that we do not attempt to judge the tracer tests in the discussion below in a review sense, but merely to deal with the questions posed.

### 4.3 Confirmation of flow connectivity

In this section we seek to answer the question:

*a) Do the data confirm the extent of connections and identify transport paths between the tracer release and recovery locations?*

As fractures and fracture zones in sparsely fractured rock are frequently inferred from ambiguous geophysical and drill core data, it is difficult to ascertain whether interpreted “deterministic” features actually span interpreted intercepts in different boreholes. For this reason, different kinds of hydraulic tests are usually carried out to elucidate the properties of the rock volume of interest. Typically, these include pumping tests to assess the transmissivity of packed-off borehole sections where connected flowing features are thought to exist. At the same time, monitoring of groundwater pressure may be carried out in one or more other boreholes some distance away from the test borehole (see e.g. Figure 3-20). This kind of hydraulic testing can be used to estimate transmissivities of interpreted fractures and fracture zones and also can be used to qualitatively establish the existence of hydraulic connections between transmissive sections in spatially separated boreholes.

Although pressure and flow responses from pumping test are a good indication of hydraulic connectivity between different locations, the existence of such connectivity does not necessarily guarantee the existence of a direct transport path between two locations. Furthermore, a transient pressure response in a single borehole resulting from pumping doesn’t necessarily mean that the feature is connected to the wider fracture network as the transient effect can be related to storativity effects in a structure that is otherwise hydraulically isolated. For the confirmation of connectivity to the wider fracture network by hydraulic testing, measurements must be carried out for long enough time to rule out transient effects.

It is also worth mentioning that tracer experiments in the form of dilution tests are also frequently used to identify and characterise flowing features connected to the wider fracture network. An advantage of tracer dilution tests is that flowrates under the detection limit of standard hydraulic test equipment can be measured.

If a tracer is injected into a fracture or fracture zone while pumping is carried out at another location, the tracer may arrive at the pumping well during the timescale of the experiment or it may not. If even a tiny fraction of the tracer is recovered, it can be stated with confidence that there exists a transport path between the two locations. If tracer is not recovered, this does not necessarily mean that there is no connection, but could mean that the advective travel time is longer than allowed for in the experimental monitoring time. Alternatively, it could also indicate the presence of hydraulic gradients that cause the tracer to be dispersed away from the pumping well along different transport pathways. A successful recovery of a tracer can therefore be used to unequivocally identify connecting flowpaths within interpreted individual fractures, networked fracture systems and in major fracture zones. The “no-show” of a tracer, on the other hand, does not necessarily disprove the existence of a connection.

The experience from many campaigns described in Chapter 3 is that the degree of connectivity obtained in hydraulic tests does not always match that obtained in tracer tests. Measuring locations may show hydraulic responses indicative of connectivity, although no tracer recovery is obtained in subsequent tracer tests. Also, tracer tests give only a very general picture of system connectivity, which may be biased to some extent by the objective of the test setup being to maximize recovery. Furthermore, tracer tests do not provide direct information on the detailed characterisation of the flowpaths, for example, flowpath geometry or the branching and mixing that occurs within flowpaths (flow channelling). Evaluation of breakthrough curves can give circumstantial evidence, but this is based on interpretations that most often are ambiguous.

In the field scale tracer tests performed by SKB, the recovery has varied from almost full recovery to a no-show of tracers (see various examples in Chapter 3), depending on the flow-path, flow situation, tracer, and experimental time. One of the most striking examples is the 3D migration experiment carried out in the Stripa mine (see subsection 3.4.5), demonstrating

the complexity of the connected fracture network. In this experiment, tracer recovery locations did not generally correspond to the flow pattern of groundwater entering the tunnel (see Figure 3-16).

#### 4.4 Qualitative confirmation of retention processes

In this section we seek to answer the question:

*b) Do the data lend qualitative support to the existence of retention for interacting solutes?*

Tracer tests performed in fractures with flowing water over length scales from a few to several tens of meters have demonstrated a substantial retention of tracers that in the laboratory have been characterised as moderately sorbing /Winberg et al. 2000, Andersson et al. 2002b, Gustafsson and Nordqvist 2005, Gustafsson et al. 2006b/. More strongly sorbing tracers are frequently retained to such an extent that no breakthrough is obtained /e.g. Birgersson et al. 1992a, Andersson et al. 2002b/. The relative difference in retention between different tracers is consistent with the general knowledge about how the tracers interact with geological media and the postulated mechanisms of retention for the specific tracers concerned.

In many tracer tests described in Chapter 3, clear breakthrough peak separation has been achieved using conservative tracers, such as  $^{82}\text{Br}^-$ , in combination with sorbing tracers, such as  $^{85}\text{Sr}^{2+}$  /e.g. Landström et al. 1978, 1983, Andersson et al. 2001/. Also peak separation between differently sorbing species has been demonstrated. In TRUE-1 /Winberg et al. 2000/, the recovery sequence followed the so-called “Hofmeister series” ( $\text{Na}^+ < \text{Ca}^{2+} \approx \text{Sr}^{2+} \ll \text{Rb}^+ \approx \text{Ba}^{2+} < \text{Cs}^+$ ), which corresponds to the current understanding of the species relative sorption properties.

#### 4.5 Confirmation of process understanding

In this section we seek to answer the question:

*c) Do the data obtained in any way confirm or contradict our process understanding or retention paradigm?*

Although in situ tracer tests in flowing water provide qualitative support for the existence of transport paths in fractures and also retention of tracers, it is more difficult to evaluate the active processes and confirm or contradict our process understanding. However, even if it may not be possible to verify or falsify the retention paradigm on the basis of tracer tests, the results should be at least broadly compatible with the postulated processes considered relevant for solute retardation. Indeed, the retention paradigm could, as a whole, be regarded as a reasonable process description even in the presence of minor deviations. This is a particularly important issue when models used to evaluate data contain deliberately simplified process descriptions.

The breakthrough curves obtained from tracer tests are generally consistent with those predicted from models based on the current understanding of the processes involved and using physically plausible parameters. However, many of the processes governing transport in fissured rock (matrix diffusion and sorption, diffusion into stagnant zones, hydrodynamic dispersion, sorption kinetics, etc) have similar effects on the tracer breakthrough and a discrimination of processes cannot be made from a single breakthrough curve.

Experiments involving several tracers with different retention properties can, to some degree, be used to distinguish between some of the processes. However, due to experimental uncertainty, limited resolution in the tail end of the breakthrough curve, etc, a definite distinction between processes cannot be made and there is a need for supporting information from experiments set up to study individual processes. In the following sections we will detail what we actually can say about our process understanding on the basis of tracer test data interpretation.

#### 4.5.1 Transport of radionuclides along discrete flowpaths

Transport of radionuclides along discrete flowpaths in fractured rock has been shown in tracer experiments. These experiments have also provided evidence that transport can occur in relatively limited parts of the fractures, a phenomenon commonly referred to as “channelling” /Rasmuson and Neretnieks 1986, Abelin et al. 1994, Birgersson et al. 1992a/.

Occasionally, additional information can be obtained by excavation of the presumed transport pathways themselves to measure the distribution of solute retention in the rock. This was done to some extent in the 2D migration experiment at Stripa where the overcored fracture sections exhibiting sorbed solutes gave some indications of preferential flow patterns /Abelin et al. 1985/, although the extent of the channelling could only be observed to a distance of about 0.5 m distant from the injection location. Another possibility is to inject epoxy resin directly into the fracture followed by excavation of the hardened, epoxy-filled fracture to quantify the distribution of fracture apertures. This technique gives indications of where preferential flows might have occurred, although it does not directly demonstrate channelling phenomena.

Another issue is that excavation methods require destruction of the rock volume being studied, thus rendering any conclusions moot if it is not also assumed that the properties of the studied rock volume are applicable to similar rock volumes.

Such an excavation procedure has recently been conducted by SKB for post mortem characterisation of the TRUE-1 site as part of the TRUE-1 Continuation Project. However, results are pending. Previously, a pre-study has been made in four different fault and fracture zones at the Äspö HRL /Hansen and Staub 2004/ with mixed results for quantifying the pore spaces, including the three dimensional connectivity of small-scale structures adjacent to the main feature core. The study involved grouting of the target features with epoxy resin doped with a fluorescent additive (Uranine) followed by a  $\varnothing$  300 mm overcoring.

Other evidence of solute transport along discrete flowpaths comes from observations of tracer breakthrough simultaneously in multiple locations or from interpretation of tracer breakthrough modality.

The 3D migration tests, the 2D migration test in a single fracture, and the channelling experiment performed at Stripa (see Section 3.4) are good examples of simultaneous breakthrough at multiple locations /Abelin and Neretnieks 1981, Abelin and Birgersson 1987, Abelin et al. 1987ab, Abelin et al. 1990, Birgersson et al. 1992ab/.

In many tracer experiments, examination of breakthrough data at a single recovery location has indicated multimodal tracer breakthrough (most notably the radially converging tracer tests at Finnsjön /Gustafsson and Nordqvist 1993/ and the STT2 test in Äspö-TRUE /Winberg et al. 2000/), which is highly suggestive of channelling behaviour or of multiple flowpaths.

#### 4.5.2 Matrix diffusion

Matrix diffusion has been demonstrated in the laboratory in many studies /e.g. Torstenfelt et al. 1982, 1983, Skagius 1986, Ittner et al. 1988, Johansson et al. 1998/ including both non-sorbing and sorbing solutes. In the SKB site investigation programme, matrix diffusion has also been demonstrated in the laboratory on drill core samples from the investigated sites /Byegård et al. 2005, 2006/. A major criticism of the work performed in the laboratory on core samples relates to the possibility of destressing effects and damage to retrieved drill cores. This may lead to substantial overestimates of the extent of in situ connectivity and effective diffusivity. The few in situ studies that have been performed using intentionally introduced tracers, however, have shown clear evidence of matrix diffusion /Birgersson and Neretnieks 1990, Vilks et al. 2003/. Furthermore, in these experiments the solutes were shown to penetrate the rock matrix to at least the depth that would be expected considering the timescale of the experiments. The tracer tests by /Birgersson and Neretnieks 1990/ is described in subsection 3.4.3 of this present report.

Tracer concentration profiles in the rock matrix from in situ experiments can be described by models based on the current process understanding using physically plausible parameters. Concentration profiles, uptake and breakthrough curves from in-diffusion and through-diffusion experiments performed in the laboratory can also be modelled by using physically plausible parameters. It should be acknowledged, however, that the models used are seldom able to reproduce every aspect of the concentration profile or breakthrough curve. This is partly due to the rock heterogeneity and partly due to the fact that the models are based on a simplified process representation. It should also be acknowledged that the present process understanding is not complete. The use of Fickian models, for example, to describe diffusive transport may not be optimal in a reactive system where there are significant electrostatic forces.

For moderately or strongly sorbing species, concentration profiles extending for significant depths (> 1 mm) into the rock matrix are seldom if ever obtained within the available time frame, especially not field scale tracer tests.

There have been arguments against matrix diffusion within the undisturbed rock matrix that would partially contradict the retention paradigm /e.g. Miller et al. 1994/. Such arguments are predominantly based on uranium-series disequilibrium studies involving naturally occurring uranium as a tracer. It has been suggested more recently, however, that such arguments are insufficiently founded, as they have not properly accounted for transport processes /e.g. Rasilainen 1997/. Many of the qualitative conclusions in the earlier studies referred to by /Miller et al. 1994/ were highly speculative and not substantiated by transport modelling. Neither did they always adequately consider boundary conditions. More particularly, many did not consider the timescale of uranium daughter re-equilibration in relation to the timescale of diffusion.

Although not strictly a tracer test, we note that studies of in situ rock resistivity, as described in subsection 3.6.2, lend strong support to the concept of matrix pore connectivity over large distances (metre scale) into the rock matrix. In /Löfgren and Neretnieks 2002, Löfgren 2004/ it is shown how to transfer in situ rock resistivity data to matrix diffusion data and in /Löfgren and Neretnieks 2006/ the long-range pore connectivity of rock at depth is discussed and also demonstrated by using electrical methods.

### 4.5.3 Partitioning of solutes

Partitioning between dissolved radionuclides and mineral surfaces has been previously demonstrated in numerous laboratory investigations /e.g. Torstenfelt et al. 1982, 1983, Ittner et al. 1988, Skagius 1986, Eriksen and Locklund 1987, 1989, Johansson et al. 1998, Byegård et al. 1998/ and has also been demonstrated in situ indirectly by way of the retardation of sorbing solutes as described in the experiments outlined in previous sections.

The relative retardation of solutes seen in tracer tests generally follows predictable patterns established from laboratory investigations. In the tracer tests carried out within the Äspö TRUE-1 and TRUE Block Scale programme, for example, the relative retardation of solutes was observed to approximately follow the Hofmeister series ( $\text{Na}^+ < \text{Ca}^{2+} \approx \text{Sr}^{2+} \ll \text{Ba}^{2+} \approx \text{Rb}^+ < \text{Cs}^+$ ) which is strongly indicative of an ion-exchange sorption mechanism. It is more difficult to establish the exact mechanism of partitioning for other solutes influenced by hydrolysis such as  $\text{Mn}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Co}^{2+}$ ,  $\text{Zn}^{2+}$  as well as the trivalent and tetravalent lanthanides and actinides. In most cases, these solutes are retarded so strongly that it is not feasible to use them to perform tracer tests at all.

The STT1b tracer test from TRUE-1 possibly documented the first breakthrough of  $\text{Co}^{2+}$  obtained in an in situ tracer experiment over a significant transport distance in crystalline rock.  $\text{Co}^{2+}$  was chosen in this experiment to see if it was possible to evaluate the transport of a solute which is known to be affected by hydrolysis and surface complexation processes. The data obtained for  $\text{Co}^{2+}$  indicated a strong retardation effect with a clearly defined peak breakthrough (greater retardation than  $\text{Rb}^+$  and possibly of the same magnitude as  $\text{Cs}^+$ ).

One of the most consistent observations during the modelling evaluation of the TRUE-1 tests overall /e.g. Winberg et al. 2000, Elert and Svensson 2001, Hodgkinson and Black 2005/ was that although the breakthrough order of solutes was as expected, they experienced significantly enhanced retardation compared to what was expected on the basis of independent laboratory sorption and diffusion data, the latter based on laboratory work on generic intact/unaltered material of Åspö rock.

It is noted that the hypothesis of retardation by sorption in tracer experiments performed in situ is largely based upon interpretation of breakthrough data. Occasionally this is coupled with support from laboratory investigations of partitioning processes involving site specific geological materials. Given the very low concentrations of solutes typically used in such laboratory experiments (in most cases well below theoretical solubility limits), partitioning is typically interpreted as a sorption process. Spectroscopic methods of various kinds can be used to directly demonstrate the existence of surface complexes and confirm sorption mechanisms, although generally only at much higher surface loadings where surface precipitation and co-precipitation effects cannot always be neglected as an additional partitioning process /e.g. Bargar et al. 1997, 1999, 2000, Farquar et al. 1997, Strawn and Sparks 1999, Rabung et al. 2000, Sylwester et al. 2000, Stumpf et al. 2001, 2002, Stumpf and Fanghänel 2002, Bostick et al. 2002, Hennig et al. 2002, Dähn et al. 2003/. The hypothesis of retardation by sorption is thereby an assumption that is usually not corroborated by direct measurement. Much of the criticism of the hypothesis of sorptive retardation therefore relates to the fact that sorption cannot be directly observed or confirmed in tracer experiments and that it is generally not observed in natural analogues of radionuclide transport.

The injection of the highly mobile pertechnetate ion,  $\text{TcO}_4^-$  and its subsequent “no show” amongst the recovered tracers in TRUE-1, for example, although being suggestive of its reduction to the immobile Tc(IV) form, does not constitute a proof that this indeed occurred during the experiment. Similarly, the fact that known ion-exchanging solutes appear to be retarded in a sequence corresponding to the Hofmeister series, although consistent with the hypothesis, does not constitute a proof of retardation by ion-exchange. In spite of this, given the level of understanding of physical mechanisms that could potentially be involved, these mechanisms are the most parsimonious explanations for the observed phenomena. If the observed behaviour was shown to strongly contradict these hypotheses, on the other hand, then it would constitute a falsification of the postulated retardation mechanisms.

Most natural analogue studies directly related to radionuclide transport processes typically have had as their focus, natural uranium mineralisations and U-series disequilibria (see e.g. /Miller et al. 1994, 2000/ for an overview). If we neglect interpretation issues related to establishing relevant boundary and initial conditions in natural analogues, we generally see either mobilisation of a solute by way of its absence in a geological matrix or accumulation of a solute in some mineralised form.

In the case of uranium analogues, the stark contrast in mobility of U(VI) and U(IV) under oxidising and reducing conditions gives rise to a very easily observed progression of transport and partitioning processes along flowpaths where oxygen rich surface water has penetrated the subsurface. The accumulation of solute at redox fronts is so pronounced that solubility limits are frequently exceeded and uranium is mineralised in the form of micro-precipitates in the reduced zone. The uranium “roll-fronts” observed at Poços de Caldas are typical examples of this /see e.g. Chapman et al. 1992/. In a similar fashion, analysis of fracture minerals such as calcite typically gives indications of historical co-precipitation phenomena. The absence of specific members of the U-series decay chain within the rock matrix is a strong indicator of solute mobilisation, although it does not say a great deal about the presence or absence of sorption, apart from the general observation that certain members of the chain appear to be more mobile than others.

The point here is that such fossils of past solute migration processes are often easily identified, relatively easy to analyse, and in the case of solute accumulations they are often clearly formed as a result of precipitation or co-precipitation processes. The presence of sorbed solutes on

geological samples, however, is not easily established. Measurement of the sorbed fraction of a solute by leaching or selective extraction only indirectly points towards a sorption mechanism as the immobilised fraction could arguably be sequestered in a soluble mineral form, as a soluble co-precipitate, or even dissolved in the matrix porewater of the sample micro-porosity. If the concentration of the solute were sufficiently high that spectroscopic evidence could be cited there would be substantial risk of surface precipitation and co-precipitation effects anyway. This is the confirmation paradox of the sorption paradigm.

It is also worth noting that the presence of one or more of these additional geochemical processes does not rule out sorption, as all of these processes can operate simultaneously in different parts of the system to give rise to the observed retardation.

It is also of relevance here to point out that the transport and immobilisation processes featured in natural analogues are frequently the result of very strong geochemical gradients being imposed upon the system (oxygenated water intrusion in the case of uranium analogues). Although this is of some relevance for flushing events in the wake of glaciation and deglaciation events, it is thought that transport processes during the bulk of the lifetime of a waste repository will be associated with largely static groundwater chemistry. In these situations, where there should be no large geochemical gradients, we would not expect precipitation and co-precipitation phenomena to be the primary means of solute retardation. During the course of a tracer test, on the other hand, the situation is complicated by the very real risk that pumping and disturbance of the natural flow system will result in a changing water chemistry, which might set these other retardation processes in motion.

Breakthrough curves for solute recovery in tracer tests could be evaluated with one or more of these processes in mind, although the fit to the experimental data can give, at best, only vague support for any postulated mechanism given the physical and geochemical heterogeneity inherent in such systems. The notion that sorption as a retardation mechanism does not exist in the field because it cannot be measured directly, when there is very strong theoretical reasoning and laboratory evidence to suggest that it should occur, is not justified.

The above discussion notwithstanding, the reader should take note that we frequently use the term partitioning instead of sorption in this report to note the fact that it is not strictly possible to demonstrate that the retardation effect is actually due to sorption and not a process such as surface precipitation or co-precipitation. It is likely that during the lifetime of a repository, solutes experience transport and retardation processes representing a mix of all these phenomena.

Although linear sorption is typically assumed in Safety Assessment models, the exact mechanism of retardation and its linearity or non-linearity is not of primary importance for this work as it is possible to include any or all of these processes in transport modelling if deemed appropriate for the scenario at hand. Indeed, sorption of solutes in natural systems generally represents a continuum of behaviour from linear sorption at low concentrations ranging through non-linear sorption at higher surface loadings to surface precipitation and co-precipitation phenomena at high concentrations or when strong geochemical gradients exist. It should be remembered that we seek in this document to outline what can be learned from tracer test experiments rather than defend any particular modelling assumption or approach used in SA, which by necessity is characterised by a much simplified process description.

To our knowledge, substantiated arguments in contradiction of partitioning in the immobile phase have not been made. As described in some detail above, however, it is difficult from in situ tracer tests to differentiate between the processes involved (sorption, precipitation, co-precipitation, etc).

By and large, tracer data qualitatively corroborates our process understanding in a broad sense, although a certain degree of deviation from predicted behaviour can probably always be expected. Much of this deviation is the result of oversimplification of what is an extremely complex system. Such deviations, however, are opportunities for increasing our understanding of retention processes as they give insights into behaviour that may not necessarily be apparent



in laboratory experiments. It should be noted, however, that phenomena that can be observed on the timescale of a tracer experiment may not necessarily be relevant on the scale of SA. This is an issue of great importance for the use of tracer test data in SA.

## 4.6 Abstraction of lumped transport parameters

In this section we seek to answer the question:

*d) Are the data sufficiently well resolved that we can obtain estimates of lumped retardation parameter groups for the specific tracer test and solute within the experimental time frame?*

If the retardation paradigm is applicable, the residence time distribution (RTD) for the tracer can be used to infer properties of both the flowpath and the geological material with which the tracer interacts. With this information it is possible, in principle, to predict tracer breakthrough in tests performed over similar time spans within comparable flow fields.

An important output from a tracer test is the tracer residence time. In many modelling concepts used for both tracer test evaluation and SA, transport along a flowpath is assumed to be retarded both by sorption on external fracture surfaces and diffusion into the rock matrix coupled with sorption on mineral surfaces internally within the rock matrix. In both tracer test evaluation and SA, sorption is typically considered to be a linear, reversible, equilibrium process. As discussed when handling question c), however, this is a modelling simplification and not absolutely central to the data evaluation; other retention mechanisms can always be modelled if sufficiently motivated. If we momentarily neglect the possibility of other partitioning processes, we shall use the hypothesis of linear equilibrium sorption in the following sections to illustrate some of the more important quantitative aspects related to the interpretation of tracer test data.

### 4.6.1 Aspects of transport processes involving matrix diffusion and sorption

For a generalised transport problem involving retardation by sorption and diffusion, the total tracer residence time of a solute can usually be broken down into contributions from several sub-processes:

$$\begin{pmatrix} \text{total} \\ \text{residence} \\ \text{time, } t \end{pmatrix} = \begin{pmatrix} \text{water} \\ \text{residence} \\ \text{time, } t_w \end{pmatrix} + \begin{pmatrix} \text{residence time due to} \\ \text{sorption on external} \\ \text{fracture surface, } t_s \end{pmatrix} + \begin{pmatrix} \text{residence time due to} \\ \text{diffusion and sorption} \\ \text{in rock matrix, } t_m \end{pmatrix} \quad (4.1)$$

Although more sophisticated approaches can be used for the modelling of transport processes, the simple case of a rock matrix of infinite extent with constant material properties can be evaluated using an analytical solution given by /Neretnieks 1980/ for a solute release of constant concentration. This is used here for illustrative purposes only to indicate some of the overarching aspects of tracer test data evaluation. More complex approaches are discussed later, although all of features and mechanisms discussed in the context of this example are also relevant for more complex modelling approaches.

Based upon the analytical solution, the contributions to the solute residence time can be shown to be:

$$t_w = \frac{\delta_f}{2} \times F \quad (4.2)$$

$$t_s = K \times F_a \quad (4.3)$$

$$t_m = \frac{1}{4 (\operatorname{erfc}^{-1}(C/C_0))^2} \cdot (MPG \times F)^2 \quad (4.4)$$

The parameter  $K_a$  is a surface area normalised sorption partitioning coefficient,  $\delta_f$  is the transport aperture of the fracture, and  $F$  is the flow-wetted surface to flow ratio, or F-factor. It should be emphasised that the magnitude of the term  $K_a$  is related to the effective flow wetted surface over which surface sorption is conceptualised to take place and can therefore have a very different numerical value to  $K_a$  values derived on the basis of BET-surface areas, or other definitions. For a non-sorbing tracer,  $K_a = 0$ , although for a fracture coating with substantial porosity, this need not be always so.  $C$  is the recovered downstream tracer concentration (i.e. breakthrough concentration).  $C_0$  is the tracer concentration at the injection location. The material properties group,  $MPG$  is defined as:

$$MPG = \sqrt{D_e (\epsilon_p + K_d \rho_b)} \quad (4.5)$$

The parameters used in the definition of the  $MPG$  are the connected porosity of the rock matrix,  $\epsilon_p$ , the effective diffusivity,  $D_e$ , the linear equilibrium sorption coefficient,  $K_d$ , and bulk density of the rock matrix,  $\rho_b$ .

The effective diffusivity,  $D_e$  is frequently given in terms of the Formation factor,  $F_f$  (not to be confused with the variable  $F$  denoting the F-factor). This is a dimensionless number which is operationally defined as the ratio of the effective diffusivity of a solute in the rock to that of an infinitely dilute solute in free solution (see subsection 2.4.2).

Although simplistic, the analysis described above can give very interesting insights into the retardation process and how different mechanisms may dominate when considering the transition from tracer test to SA timescales.

For low values of  $F$  and a non-sorbing tracer, for example, the tracer residence time distribution (RTD) may be overwhelmingly dominated by  $t_w$  and matrix diffusion effects may be essentially absent from the breakthrough data. For a sorbing tracer, on the other hand,  $t_w$  may be small compared to both  $t_s$  and  $t_m$  and can therefore be neglected. Depending upon the magnitude of  $F$ , matrix diffusion effects may be either absent, partially contributing to the observed RTD, or may be the dominating process governing solute retardation. Moreover, in many cases the same breakthrough characteristics could arguably be reproduced by assuming hydrodynamic dispersion and surface sorption without the need to invoke matrix diffusion at all /Miller et al. 2000/, thereby indicating that the diffusive interaction with the rock matrix over the timescale of a field scale tracer test is typically very weak.

The interpretation of data is complicated, however, by the presence of additional processes that are not strictly accounted for in the model being used for data evaluation. The presence of multiple pathways, multiple immobile zones in the rock matrix and zones with stagnant water, for example, is not included in the simplistic model described by Equations 4.1–4.5 and this could result in an incorrect interpretation of data.

The idea of separating solute-rock interaction into a fracture surface contribution and a matrix contribution is purely a model simplification and the distinction between the two depends on the test conditions. Most of the immobile zone adjacent to fractures is expected to exhibit material properties ( $\epsilon_p$ ,  $D_e$ ,  $K_d$ ) that vary more, or less continuously with depth into the rock matrix from the fracture, as they are largely a result of tectonics, hydrothermal alteration and other weathering processes. On the basis of drill core observations these alteration zones can vary from a fraction of a millimetre at one extreme, to possibly many tens of decimetres depending on the fracture type and its hydrothermal and tectonic history.

The depth-dependent variation of material properties can be very important for evaluating a tracer test since the retardation properties observed, to a large extent, reflect the penetration depth the solute reaches into the rock matrix during the course of the tracer experiment. The contribution of fracture surface equilibrium sorption can be dispensed with if a model is used for data evaluation that specifically considers this depth dependent variation. Owing to a general lack of data and the difficulty of measuring such property variation, however, very few attempts have been made to model the interaction of solute with the rock matrix in this way. Most models therefore consider a much simplified representation of the rock matrix microstructure (usually abstracted into a number of distinct layers).

Although matrix diffusion is characterised here in this simple analysis as a single rate process with some mean effective diffusivity, we note that the system may exhibit “multirate” characteristics, as discussed in subsection 2.4.2. It should be noted that here, the multirate diffusive properties reflect not only variation of material properties and matrix pore connectivity as a function of depth into the rock, but also the effects of multiple, parallel fluxes into immobile zones possessing differing material properties. Indeed some codes used for transport modelling include this possibility (e.g. the Darcy Tools code /Svensson et al. 2004/). Regardless of this feature, the added complexity does not necessarily lead to a more accurate representation of matrix diffusive properties at SA timescales, owing to the typically low effective penetration depth of matrix interaction that is observed during the tracer test. This is discussed in more detail in Section 4.7.

What is important to note from Equations 4.1–4.5, however, is that flow-related transport parameters ( $FWS/Q$ ) are interweaved with material properties ( $MPG$ ) in such a way that it is only possible to observe their effect in the form of lumped parameter groups as they do not individually provide a unique description of the system. Furthermore, both channelling effects (i.e. giving a distribution of  $FWS/Q$  values) and diffusive matrix interaction give rise to strong tailing effects in the solute recovery curve which are difficult to separate in a robust fashion.

Provided that the theoretical model used for data evaluation represents the major processes that actually give rise to the observed retardation, it is only possible to speculate upon the values of the individual parameters comprising these groups using tracer breakthrough data. If the model used for data evaluation neglects additional processes that may be of importance, the data interpretation may also be strongly biased.

The lumped retardation parameter groups that characterise the tracer transport are therefore only strictly applicable for the flow situation and the time frame of the tracer experiment from which they were derived. This is of great relevance for the upscaling of results from tracer tests and will be discussed in more detail in subsequent sections.

To properly resolve the contributions from the water residence time, sorption and matrix diffusion effects, it is important for the tracer pulse to be as close to an instantaneous pulse release as possible. Although not a strict mathematical requirement for data evaluation, in a practical sense this is very important owing to that an imperfect injection pulse with a decaying tail gives rise to features in the breakthrough curves that are essentially indistinguishable from retardation effects for the typically low  $F$ -factors characterising most tracer tests. Perfect solute release pulses are generally not achievable in real tracer experiments and considerable effort must be invested in imitating such a release. Furthermore, if it is desired to obtain information about the rock matrix,  $t_w$  should be sufficiently small in comparison to  $t_s$  and  $t_m$  so that the matrix interaction is tangible. It should be noted though that if achieving the instantaneous pulse by active injection followed by flushing, which is the practical way of doing it, the added pressure may activate flowpaths that would not be active if performing passive injection.

Many of the earlier tracer tests featured poorly constrained tracer release boundary conditions which made it very difficult to distinguish between the effects of the tracer injection characteristic and the transport properties of the flowpaths connecting injection and recovery wells.

This was particularly the case for experiments where a decaying pulse or constant concentration injection was used in systems with relatively short advective travel times ( $t_w$ ). Tracer injection where natural flow through a borehole section of finite volume is used to disperse the solute typically gives rise to a decaying pulse characteristic. This is especially problematic for radially converging tracer tests as a low flow-rate in the injection section gives rise to significant tailing which can mask retardation effects arising due to matrix interaction /e.g. Andersson et al. 2002b/.

Tracer tests where forced injection of solute is followed by a solute-free groundwater chaser can potentially give better resolution of breakthrough curves although with the disadvantage that a very unnatural flow gradient is applied to the system (i.e. an unequal- or full flow dipole depending upon the injection flowrate). This is not to say that a radially convergent flow field is essentially more natural, but that active injection of tracer can activate flowpaths that are otherwise poorly conductive. These paths may not exhibit flow in a radially converging flow field which is often more reliant upon naturally, high-conductive connections within the wider fracture network. In experiments where the active injection is only applied for a brief period this can also have the disadvantage that tracer is initially pushed into portions of the connected fracture system which subsequently become stagnant when the active injection ceases. This phenomenon was thought to have occurred in at least one of the tracer tests carried out within the TRUE Block Scale project /Andersson et al. 2002b/.

It has also been suggested by /Kohl et al. 1997, Kosakowski and Berkowitz 1999/ on the basis of Navier-Stokes flow simulations, that flow dynamics and mixing behaviour observed during tracer tests may differ to that obtainable under more natural hydraulic gradients. /Neretnieks 2007/ also raises this issue in the context of SWIW-test interpretation. In the analysis it is shown that longitudinal Taylor dispersion in flow channels with tapered edges (i.e. Taylor dispersion across the flowpath width rather than flowpath aperture) can add considerable dispersion to breakthrough curves that would be absent at SA timescales where full mixing across the flowpath width should occur due to molecular diffusion.

In the Äspö Task Force, Task 4 evaluation of the TRUE-1 experiments, an approach based upon deconvolution of breakthrough curves for a known and well defined tracer injection curve was used to obtain unit response curves for solute transport over a 5–10 m scale in the STT-1, STT-1b, and STT-2 tracer tests using sorbing solutes /Elert and Svensson 1999, 2000/. The unit response function, in principle, describes the retardation of solutes along a flowpath and can be regarded as a lumped parameter evaluation of transport properties where effects relating to the solute injection boundary condition are filtered out. It essentially corresponds to the breakthrough curve that would be obtained using a perfect Dirac pulse  $\delta(t)$  boundary condition. The method is most useful for identifying multiple flowpaths and distinguishing them from artefacts relating to tracer injection, although it implicitly assumes a linear sorption process. The method generally does not give any indication whether interpreted multiple transport paths result from clusters of interconnected fractures or individual flow channels within a single fracture.

The approach used by /Elert and Svensson 1999, 2000/ was previously used by /Tsang et al. 1991/ to analyse the breakthrough data from the 3D tracer experiment at Stripa. In this case, the existence of between two to four independent flowpaths was strongly indicated by the multiple peaks revealed in the deconvoluted data. In the STT-1 and STT-1b experiments from TRUE-1, no strong indications of multiple flowpaths were observed, although a bimodal pathway was clearly evident in the data for STT-2. Although undoubtedly a very useful tool, deconvolution is an ill-conditioned problem that is difficult to implement numerically and is therefore very sensitive to measurement errors and artefacts. In some situations it is difficult or impossible to obtain reliable results using the technique and some kind of data smoothing is almost always required.

Apart from the requirement for a well-defined tracer injection and recovery curve, there are a number of other issues that must be considered both in the selection of tracers as well as in the analysis of tracer breakthrough data before any serious discussion of transport properties can be considered. Dilution of the tracer over the sampled transport distance can have a significant

impact upon the possibility of obtaining a measurable recovery of tracer. In the TRUE Block Scale experiments, for example, dilution factors on the order of  $10^8$  were estimated for some of the test geometries over a 15–130 m scale.

For a tracer to be useful in such a test it must therefore be detectable in extremely low concentrations. Generally speaking, gamma emitting radioactive tracers tend to have very low detection limits and multiple tracers can be measured simultaneously using solid state detectors. Tritium (usually in the form of HTO) being a beta emitter is typically measured using liquid scintillation counting – a very sensitive method of detection. Other tracers such as non-radioactive metal complexes and dye tracers must be measured using techniques that have less sensitivity (e.g. ICP-MS and spectrophotometry), although this can be partially compensated for by using higher injection concentrations. All measurements are also influenced to varying degrees by interfering substances (gas bubbles, colloidal mineral precipitates, background gamma activity, etc).

Another aspect of some importance is the issue of radiological protection. This sets a limit upon both the activity of radioactive tracer that may be released in a tracer test as well as its half-life. In the TRUE-1 and TRUE Block Scale experiments, the injected radioactive tracers represented a range of half-lives from some tens to hundreds of hours (e.g.  $^{24}\text{Na}^+$ ,  $^{42}\text{K}^+$ ,  $^{186}\text{ReO}_4^-$ ,  $^{47}\text{Ca}^{2+}$ ,  $^{131}\text{Ba}^{2+}$ ) up to years ( $^{22}\text{Na}^+$ ,  $^{134}\text{Cs}^+$ ,  $^{133}\text{Ba}^{2+}$ ,  $^{137}\text{Cs}^+$ ). This also has some bearing on the choice of tracer to be used for different transport distances in a tracer experiment.

## 4.7 Abstraction of individual transport parameters

In this section we seek to answer the question:

*e) Is it possible to distinguish the material properties and flow-related transport parameters, MPG and  $F$  in a robust fashion from the experimental data? (Are these estimates physically realistic in the light of independently obtained data? Can these be further decomposed into physically realistic values for their component parts? e.g.  $D_e$ ,  $K_d\rho_b$ ,  $F$ , etc.)*

As discussed previously, it is generally the influence of lumped parameter groups that are discernible in solute RTD data rather than individual parameters. For a solute that interacts strongly with the rock matrix, for example, it would be the product  $K_d \cdot F$  or  $\text{MPG} \cdot F$  that can be estimated from the solute RTD data rather than the individual entities comprising this parameter group. For a solute whose residence time is largely governed by the water residence time, on the other hand, the RTD would be dominated by  $\delta_f \cdot F$ . It is not possible to say anything concrete about various components of these groups without supporting data or additional assumptions which can be used to deterministically reduce the number of degrees of freedom in the data interpretation.

In principal, there are two options:

- Independent evaluation of transport aperture or material properties group ( $\text{MPG}$ ).
- Independent evaluation of flow geometry and channelling (the distribution of the  $F$ -factor amongst different transport paths).

In practice, both options are combined using assumptions in the modelling that are based on physically realistic parameters and are consistent with independently obtained data.

If assumptions concerning  $F$  are made, the effective transport aperture of the system can be estimated from the tracer residence time of non-sorbing radionuclides. If the water residence time can be estimated or otherwise neglected, the RTD of a sorbing tracer can give information about diffusion and sorption within the rock matrix. Given that matrix interaction is influenced through the action of parameter groups, one or more parameters must be fixed using independently obtained data if it is desired to obtain quantitative data from the tracer RTD concerning other entities comprising the parameter group. The RTD for a single sorbing tracer by itself does not contain any information that can be used to separate the effects of the different components

of  $K_a \cdot F$  or  $MPG \cdot F$ . Provided that the  $F$ -factor can be deterministically fixed, the effective  $MPG$  can be estimated from the RTD data via a calibration procedure or inverse modelling of some kind. However, estimates of  $F$  are difficult to make and we need to make some kind of assumption about flowpath geometry and flow channelling in order to fix  $F$ . As has already been briefly touched upon in Section 4.6, it is also important to remember that the spreading of recovered solute and late time slope behaviour actually represents a combination of effects relating to matrix diffusion heterogeneity, advective flow channelling and other hydrodynamic dispersion phenomena.

Independent measurements of matrix properties can be obtained from immobile zone tracer tests. In the present stage of the SKB site investigations, information from tracer tests in the immobile zone has been obtained in the laboratory. Diffusion studies have been carried out on a few dozen drill core samples using the non-sorbing tracer HTO /Börjesson and Gustavsson 2005ab/. As mentioned previously, however, such measurements carried out in the laboratory have been criticised owing to that they may substantially overestimate the effective diffusivity. In situ tracer tests in the immobile zone have only been carried out in a limited number of investigations. At present, SKB is performing such a tracer test (LTDE-SD) at the Äspö HRL within the Oskarshamn site investigation area /Byegård et al. 1999/. An important supporting investigation has previously been made by SKB in the Stripa mine /Birgersson and Neretnieks 1990/. Other important tests worth mentioning have been carried out by AECL in Canada /Vilks et al. 2003/ and by Nagra in Switzerland /Möri et al. 2003/.

As a part of the SKB site investigations, information concerning the formation factor and pore connectivity of the in situ rock matrix has been obtained by electrical methods. By measuring the rock and groundwater resistivities in situ and using the physical analogue between diffusivity and ionic mobility, the formation factor can be assessed. Such measurements have delivered on the order of 100,000 data points /e.g. Löfgren and Neretnieks 2005/. By using a similar method in the laboratory, the formation factor of hundreds of drill core samples has been obtained /e.g. Löfgren and Neretnieks 2005, Thunehed 2005/. Supporting information on the immobile zone has been obtained in the laboratory concerning the rock matrix porosity and sorption properties /e.g. Byegård et al. 2005, 2006/. Additional supporting measurements have been carried out by /Löfgren and Neretnieks 2006/, where tracer migration was studied on drill core samples by through-electromigration experiments using the anionic tracer iodide.

The  $MPG$  can be decomposed into  $D_e$ ,  $F_f$ ,  $K_d$ ,  $\varepsilon_p$  in some cases where non-sorbing or weakly sorbing species are used. By modelling, for example, a breakthrough curve from a through-diffusion experiment,  $D_e$  can be separated from the parameter group  $\varepsilon_p + K_d \rho_b$ . Furthermore, independent experiments can give  $\varepsilon_p$ ,  $K_d$ ,  $\rho_b$  etc. It should be cautioned, however, that knowledge obtained from non-sorbing or weakly sorbing species is not necessarily transferable to more strongly sorbing species that may have more complex interactions with the rock matrix. It is also noted that due to damage incurred during drill core sampling and issues related to geochemical heterogeneity, data obtained from laboratory measurements on site specific materials may not be representative of the in situ retention properties of the rock comprising a transport path.

In the Äspö Task Force, Task 4, different modelling teams attempted to simulate tracer breakthrough data from the TRUE-1 series of experiments using only independently obtained data from laboratory measurements, the tracer injection curves, and the residence time distribution of a non-sorbing tracer. To avoid confirmation biases, the breakthrough data for sorbing tracers were kept secret from the modelling teams during the task. Generally, the blind predictions did not agree well with the measured breakthrough data for sorbing tracers and in many cases matrix interaction effects were under-predicted by a factor of 30–50 /Marschall and Elert 2003/ possibly due to enhanced retention within the fracture rim or diffusion into stagnant zones. In a later analysis, /Neretnieks and Moreno 2003/ suggested that much of the difference could be explained by considering a three-dimensional flow geometry rather than the simple flowpath geometry assumed for the flow structures involved in the test. To this end the TRUE-1 Continuation project (see subsection 3.5.6) was conceived as a means of attempting to clarify

some of these issues /Andersson et al. 2002c/. However, the outcome did not lend weight to the proposed three-dimensional flow geometry.

One means that has been suggested for fixing individual transport parameters uniquely is to use a cocktail of tracers with differing diffusive or sorptive characteristics and to evaluate the multi-tracer RTD data concurrently. As for all inverse modelling procedures it is therefore subject to the danger of confirmation bias of the kind where model parameters are tweaked and adjusted until good agreement is obtained with the data, thus introducing circularity in the argumentation.

Inverse modelling in this fashion requires at least two tracers with sufficiently different matrix interaction properties that the contributions of matrix interaction, water residence time and flow-channelling effects can be separated clearly. There are good examples in the literature where this approach has been used to distinguish advective flow channelling effects from matrix diffusion (see, for example, the review by /Bodin et al. 2003/). Using non-sorbing tracers, the effective diffusivities should differ sufficiently that the peak arrival times are quantifiably distinguishable from each other. This method was used by /Becker and Shapiro 2000/ to show that matrix diffusion effects were of subordinate importance relative to flow channelling for tracer tests carried out in crystalline rock. It is important to note that this does not show that matrix interaction was absent, but rather that it was of less importance for breakthrough data interpretation of the particular tracer test considered and the timescale of monitoring.

To give good quantitative details of the  $F$ -factor and its distribution, it is more realistic to use a cocktail consisting of a number of sorbing tracers in addition to one or more non-sorbing tracers which are used to estimate the water residence time. For the data to be effectively resolved the sorption mechanism should be identical for the tracers and they should have sorption  $K_d$  values that differ sufficiently that the peak arrival times are quantifiably distinguishable from each other. In the TRUE STT-1 experiment, for example, the ion exchanging solutes  $^{137}\text{Cs}^+$ ,  $^{132}\text{Ba}^{2+}$ , and  $^{86}\text{Rb}^+$  were used as part of the tracer cocktail. In this particular experiment, clearly different peak arrival times were obtained for each tracer. If the relative sorptivities are known from laboratory experiments under representative conditions, it is possible in principle, to estimate in situ values for  $K_d$  (or  $MPG$ ) and the  $F$ -factor for the tracer test to an accuracy of perhaps an order of magnitude using a fitting procedure. In practice, this is not as clear-cut as the above description implies owing to that laboratory determined relative sorption properties of solutes do not necessarily translate well to in situ conditions. If, on the other hand, laboratory data are not used to estimate the relative sorption properties of the solutes, the system will be under-constrained and other assumptions need to be used to, for example, fix  $F$ .

A similar approach was used by /Cvetkovic and Cheng 2002/ to simultaneously estimate both  $F$  and  $MPG$  parameters for tracer tests carried out in the TRUE Block Scale experiments. In this case, a model-derived, linear functional relation was assumed to exist between the  $F$ -factor distribution and the distribution of water residence times estimated from deconvolution of the breakthrough curves for a non-sorbing tracer (i.e.  $F \approx k \times t_w$ ). Assuming also that the water residence time distribution could be roughly described as an inverse Gaussian distribution, the “actual”  $F$  distribution and  $k$  parameter were estimated from the breakthrough data for a non-sorbing tracer using a calibration procedure (physically, the  $k$  parameter can be considered to be the inverse of the fracture half-aperture and has units  $\text{m}^{-1}$ ). From the calibrated  $F$ , the strength of matrix interaction could then be estimated for each of the sorbing tracers used. The further separation of the lumped parameters estimated in this fashion to give individual estimates of  $D_e$ ,  $\epsilon_p$ , and  $K_d$  was not uniquely constrained and ranges of parameters were derived that could fit the measurement data equally well. In this particular case, estimates of individual  $K_d$  values for solutes used in the experiments were given based on the assumption of Archie’s law to describe the relation between matrix porosity and effective diffusivity.

As highlighted by the procedural description given above, the possibility of estimating in situ numerical values for transport parameters is highly dependent upon assumptions made during modelling and the quality and representativity of supporting data. Supporting data can include those obtained from laboratory experiments, hydraulic measurements, and invasive procedures

such as migration path overcoring in combination with solute penetration depth measurements, or even resin injection characterisation of transport apertures.

As already discussed in previous sections, the data evaluation may in unfortunate cases be beset by poorly defined or non-ideal tracer release boundary conditions, noisy measurement data, and weak matrix interactions to such an extent that the data evaluation is ambiguous and the individual parameters governing retardation are poorly constrained.

In the best possible case scenario, however, approximate estimates can be made of individual parameters influencing retardation processes for the specific flow situation and migration path sampled during the experiment.

## 4.8 Extrapolation of numerical data to Safety Assessment

In this section we seek to answer the question:

*f) Is it possible to extrapolate these numerical data to other timescales or flow configurations without introducing additional, non-testable assumptions?*

The scaling of results from tracer experiments to other temporal and spatial scales, with special concern for the scale of interest for SA, involves several issues:

- a) How can the parameters determining the hydrodynamic control of retention ( $F$ -factor) be scaled to different flow situations?
- b) How can the matrix properties be scaled to different timescales?

At SA timescales,  $F$  will be much higher than during a tracer test. This is mostly because relatively large hydraulic gradients are required to obtain any kind of solute recovery in tracer tests, whereas natural hydraulic gradients are much smaller (typically by several orders of magnitude). The flow field obtained in tracer tests is influenced by forced pumping which may also change the geometry of the flowpaths and the overall distribution of  $F$  in the system, particularly in the presence of channelling phenomena /e.g. Tsang and Tsang 1989, Moreno and Tsang 1991/. The existence of flow channels, their distribution, and persistence in time is dynamic and strongly dependent upon the externally imposed flow regime /Neuman 2005/. This also means that flow channels arising in a given tracer test under the influence of pumping may not necessarily be the same as those identified in other tests probing the same fracture or flow features if the hydraulic conditions are not identical in both cases.

In short, the  $F$  obtained in a tracer test has essentially no bearing on the  $F$  that is appropriate for SA timescales (even in the same rock volume being studied) owing to the very different magnitude and geometry of the applied hydraulic boundary conditions. Tracer tests can provide qualitative information concerning connectivity of the fracture network and some indications of flow porosity for the sampled flowpath. The  $F$  appropriate for SA conditions, however, can only be estimated by theoretical consideration of the fracture network connectivity and channelling phenomena using hydrogeological information obtained during Site Characterisation. Its estimation is thereby substantially stochastic in nature and highly dependent upon hydrogeological modelling assumptions that are not easily verifiable.

The scaling of  $F$  also indirectly influences the effective matrix properties “experienced” by a transported solute. This question can be addressed, at least partially, by consideration of solute penetration depths into the rock matrix.

For a tracer experiment, the effective penetration depth,  $\eta$ , can be obtained from the solution of the diffusion equation /Crank 1975/ and is related to the square root of the product of rock matrix apparent diffusivity  $D_a$  and contact time  $t_c$ . For a dipole, or radially converging tracer test, the effective penetration depth associated with solute retardation can be shown in a relative frame to be independent of the sorption properties of the solute (at least for approximately linear sorption). Simply put, a pulse of a more strongly-sorbing solute will have a longer transport



time, as compared to a pulse for a less sorbing solute. This additional time will allow the more strongly-sorbing solute, although having lower apparent diffusivity, to penetrate the rock matrix to the same depth as the less sorbing solute. In this case, the effective penetration depth can be shown to be proportional to the product of effective diffusivity and  $F$  /e.g. Crawford 2006/:

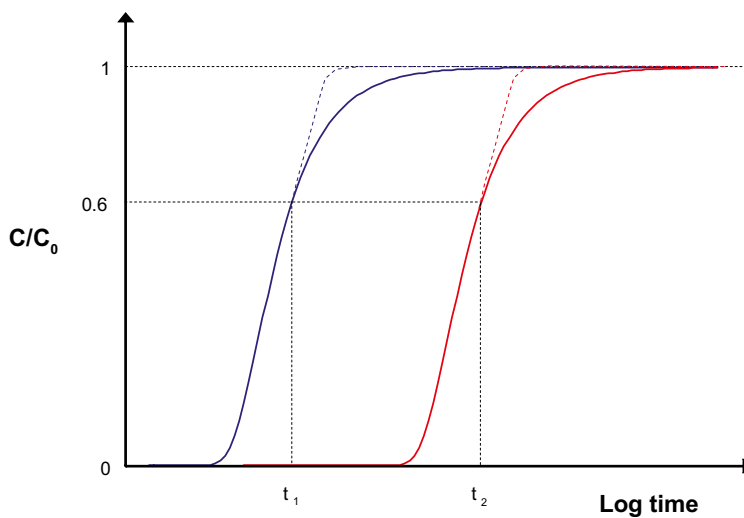
$$\eta \propto \sqrt{D_a t_c} = D_e \times F \quad (4.6)$$

To explain the concept of effective penetration depth, we can make a simple example using two tracers of differing sorption strength transported along a flowpath characterised by some arbitrary, although temporally constant  $F$ . The breakthrough curve obtained from such an experiment for a constant concentration solute release boundary condition is depicted in Figure 4-1 below where the blue curve corresponds to the less strongly sorbing tracer and the red curve is for the more strongly sorbing tracer.

In this simple example, both tracers exhibit the classic sigmoidal breakthrough curve with the weakly sorbing tracer appearing early and the more strongly sorbing tracer appearing at a later time. What is meant by effective penetration depth is that for a given breakthrough (say,  $C/C_0 = 0.6$  as depicted in the figure) both tracers experience the same depth of penetration.

This can be demonstrated more clearly by carrying out simulations for a rock matrix of limited extent. In such simulations it can be shown (see e.g. /Crawford 2006/) that both tracers start to “sense” the presence of the limited matrix boundary at identical locations on the breakthrough curve. This is also depicted schematically in the figure where the broken coloured curves start to exhibit matrix saturation characteristics at the same recovery fraction (i.e.  $C/C_0 = 0.6$ ), although at different absolute times ( $t_1$  and  $t_2$ ). The only way for this to occur is if both tracers experience the same depth of penetration at these locations.

Although this appears counter-intuitive to standard notions of penetration depth, it should be remembered that we are speaking in relative terms with the recovery curve itself as the reference frame. In an absolute reference frame, however, (i.e. at a specified time such as  $t_1$  or  $t_2$ ) both tracers are subject to very different depths of penetration. At the time corresponding to  $t_1$  in the figure, for example, the weakly sorbing solute will have penetrated to a significant depth into the rock matrix, perhaps many metres, and may even experience matrix saturation effects if the matrix connectivity is of limited extent. At the same time, the strongly sorbing tracer may only have penetrated a very small distance into the rock, perhaps as little as a few  $\mu\text{m}$  or mm.



**Figure 4-1.** Illustration of the impact of effective penetration depth concept for two tracers of differing sorption strengths. Red (strongly sorbing solute) and blue (weakly sorbing solute) solid curves correspond to the solution for an infinite matrix; broken red and blue curves correspond to the corresponding solution for a matrix of limited extent.

In safety analysis, additional consideration must also be given to the timescale of radionuclide release and the decay time of the radionuclide (and its daughters) with respect to the travel time.

A typical mean  $F$ -factor for an in situ tracer experiment is perhaps on the order of 1–100 y/m, depending on the hydraulic characteristics of the flow system. For such  $F$ -factors and for effective diffusivities typical of the rock matrix, we would expect effective penetration depths ranging from perhaps 0.1  $\mu\text{m}$  for rock representative of unaltered rock to as much as 100  $\mu\text{m}$  for a rock matrix typical of fault gouge. Thus, only the parts of the rock closest to the fracture surface contribute to the observed retardation.

Owing to that the  $F$ -factor on SA timescales is thought to be some orders of magnitude larger than that characteristic of a typical tracer test, we would expect the effective penetration depth of solute to be considerably larger under SA conditions than in a tracer test (i.e. more deeply lying rock matrix actively participates in retardation at SA timescales). Since there is evidence that rock properties vary considerably with depth into the rock matrix from flowing fractures, we would expect the retardation observed in a tracer experiment to not be representative of that at SA timescales. Given that the outer layers of rock matrix (alteration rim) adjacent to flowpaths typically (although not always) exhibit a higher effective diffusivity and sorptivity than deeper lying rock, the retardation experienced by a solute under tracer test conditions is likely to be substantially greater than that appropriate for parameterisation of SA models.

A more important concern, however, is that owing to the typically low penetration depths achieved in tracer tests, any particulate material or fracture surface roughness encountered along the transport path will appear to be part of the “apparent” flow-wetted surface in the data evaluation (see Figure 2-21). This feature will not be present at SA timescales where particulate matter and surface roughness are likely to appear as equilibrium storage capacities and most likely not contribute appreciably to the solute transport times. Depending upon how individual entities comprising the matrix interaction parameter groups are separated in the RTD data evaluation, these effects could have a large impact on the data obtained.

Notwithstanding the above, the relative importance of the outermost layers of the altered zone relative to the more deeply lying rock matrix is entirely dependent upon the part of the RTD under consideration. While the results from the first order analysis of penetration depth (Equation 4.6) suggest that tracer test data say very little about, say the median (i.e. 50% recovery) travel time for released radionuclides under SA conditions, the data do probe the rock layers of relevance for the arrival time of the first, say 0.05% of released radionuclides if the  $F$ -factor under SA conditions is roughly 1,000 times greater than that achieved during a tracer test.

Concern has been raised about the long range connectivity of the porous system of in situ rock. As already discussed in previous sections, there are objections against demonstrations of pore connectivity in the laboratory, as the rock becomes damaged in the excavation process and de-stressed when unloaded by drilling and taken to the surface. In situ experiments performed in the Stripa mine (where great care was taken to avoid de-stressing effects) have previously demonstrated long-range connectivity of at least 40 cm /Birgersson and Neretnieks 1990/. However, those consisted of tests at only three locations and concern a very small rock volume. Recently, the fact that an electrical current generally can be propagated through the porous system in situ has been interpreted as strong evidence for long-range pore connectivity in the rock matrix /Löfgren and Neretnieks 2006/.

Another concern is whether one can extrapolate retention due to partitioning in field observations to SA when the partitioning processes are not completely understood and are described in a simplified way in models. At present, linear sorption using the  $K_d$ -approach is used within SA-modelling. The justification for this is partly that radionuclide concentrations undergoing transport are projected to be sufficiently low that non-linear effects relating to surface loading will be absent. Some of the radioactive solutes present in spent nuclear fuel also have naturally occurring counterparts in groundwater at concentration levels higher than that expected to be released from the repository (e.g. Cs(I), Sr(II), Ra(II), U(IV/VI)). The natural background level

of these solutes therefore establishes the relevant concentration range under which sorption will take place. The assumption of linear sorption is also rationalised in terms of the notion that for a groundwater of constant composition it is not necessary to consider multicomponent effects and other non-linearities if the  $K_d$  value is applicable for the prevailing conditions. The use of such models in SA is contingent upon scenarios of repository evolution involving constant groundwater composition. We note, however, that more elaborate models must be used for coupling of geochemistry and transport if scenarios are considered where groundwater composition changes over time in such a way that the  $K_d$  approach is not appropriate. This may be important for remobilisation scenarios involving changing water composition in the wake of glaciation and deglaciation episodes.

In the face of the poorly constrained complexity of natural systems, the goal of SA has generally not been to model solute transport and retardation processes exactly as they would be observed in nature. Indeed, it has previously been stated by /McKinley and Scholtis 1993/ that “the primary objective is to establish a theoretically based picture of potential radiological risk associated with different scenarios of waste repository evolution. When applying linear  $K_d$  data to such analyses, even when such an approach may not be strictly valid, approximate  $K_d$ -values should be chosen to ensure that they conservatively overpredict radiological consequences”. This, of course, should also be balanced against the requirement to provide realistic estimates that can contribute to overall system understanding and confidence building with regard to SA. Data and models used in SA should therefore be chosen judiciously to be appropriate for the task at hand and the timescales considered. In this sense, a clear distinction must be drawn between the objectives of Site Characterisation and Safety Assessment.

As our knowledge of site specific conditions and understanding of sorption processes becomes more sophisticated, the data and concepts used in SA should be continuously reviewed with the aim of simulating the repository system in as much detail as is reasonably possible given the prevailing level of characterisation and system understanding. This will naturally include internalisation of tracer test data in the process understanding and resulting site descriptions.

## 4.9 Possibilities and limitations of tracer tests

In this section we summarise the discussions in the previous chapters and give recommendations. The table below could be used as a guide for use during the initial phase of designing a tracer test.

**Table 4-1. A guide to the possibilities and limitations of tracer tests.**

<b>Confirmation of flow connectivity</b>	
A1	By performing single-hole hydraulic testing one can obtain clear indications whether the investigated borehole sections are hydraulically connected to the wider conductive fracture network. This strengthens the case for interpreted flowing fractures or fracture zones thought to intersect the borehole section where the test is carried out. Similarly, a non-response weakens the argument for hydraulic connections via such features.
A2	By performing cross-hole hydraulic testing one can obtain indications whether there is a hydraulic connection between the test boreholes.
A3	By performing cross-hole tracer tests one can demonstrate unequivocally that the injection and recovery wells are connected by at least one advective transport path if tracer recovery is obtained. If, however, there is a demonstrated hydraulic connection and no recovery of tracer is obtained, no definite conclusion can be drawn concerning transport connectivity.
A4	If recovery is obtained in cross-hole tracer tests, only little information about the flowpaths themselves can be obtained from the tracer recovery data alone. This applies for both small-scale and large-scale tracer tests. At best, a multimodal solute breakthrough is indicative of multiple transport paths, although no specific details concerning flowpath geometry can generally be drawn from such results.

---

**Qualitative confirmation of retention**

---

- B1 If two or more tracers representing differing sorptivity/diffusivity characteristics are injected in the same run at the same injection point (with identical, artefact-free injection characteristics), withdrawn at the same withdrawal point, and there is good peak resolution and separation in the breakthrough curves (i.e. the maximum peak breakthrough is clearly resolved and peak arrival times differ), this lends strong qualitative support of retention. This applies to cross-hole tests of the dipole, unequal-dipole, or radially-converging kind. It is not clear yet whether this conclusion also applies unequivocally to SWIW tests where the dynamics of tracer recovery may be more complex and injected tracers do not necessarily sample identical advective transport pathway trajectories owing to the flow reversal and possible presence of background hydraulic gradients which complicate interpretation.
- B2 If two or more tracers in a single run are injected at the same injection point, withdrawn at the same withdrawal point, and there is poor peak resolution and separation in the breakthrough curves, this does not lend evidence for or against the existence of retention. It merely indicates that the species were subjected to similar “apparent retention” processes (or lack thereof), hydrodynamic dispersion of various kinds, or possibly that the data are corrupted by experimental artefacts.
- B3 If two or more tracers in different runs (one tracer in each run) are injected at the same injection point, withdrawn at the same withdrawal point, and their residence times differ, this does not lend unequivocal support for retention. Other factors than retention may have influenced the tracer residence time, such as uncontrolled changes in hydraulic gradients. The certainty with which one can say that the different runs were subjected to identical, or at least defined, conditions reflects the level of confidence that can be ascribed to the comparative information obtained for tracer recovery.
- B4 If the same tracer is injected at different locations in a cross-hole test, the residence times do not necessarily correlate with the geometrical distance between injection and withdrawal points. Furthermore, the residence times do not necessarily correlate with the flowpath length, groundwater residence time, or any easily evaluated parameters. Comparing residence times from tracer tests performed at different locations is not a robust way of investigating retention in a quantitative sense.
- B5 It is generally not possible to demonstrate retention unequivocally using only one tracer. This is because it is not possible to distinguish between transport aperture effects (i.e. water residence time) and retention processes in an unambiguous fashion. It is also noted that the slope of late time arrival, tracer recovery is not a robust indicator of matrix interaction owing to artefacts relating to multi path advection, stagnant zone diffusion, and other dispersion effects.
- B6 Tracer tests with focus on the rock matrix itself (in situ diffusion) can provide support for the existence of retention under field conditions arising from both matrix diffusion and partitioning. When transferring information from such tests to advective radionuclide transport, however, some assumptions need to be made. Such assumptions are generally based on sound physical reasoning, such as that matrix diffusion will always occur if there is a concentration gradient and a connected pore volume. Much of the apparent retention demonstrated in, for example, cross-hole tests can be shown to occur at the rock/fracture interface. This interface has so far not been sufficiently well addressed in immobile zone tracer tests and neither is it entirely relevant for retention processes at Safety Assessment timescales.

---

**Confirmation of process understanding**

---

- C1 Detailed process understanding generally cannot be obtained solely from tracer test breakthrough data interpretation. The reason is that many processes of relevance are characterised by similar timescales and will give similar responses in the obtained breakthrough data. Separating these different processes on a detailed level is not feasible in the absence of independent corroborating data. More complex tracer test concepts involving, for example, remobilisation of tracers in a systematically altered chemical environment have not been tested at this time. This is an avenue for possible future research.
- C2 Tracer tests giving results that are broadly compatible with the process understanding lend weight to the retention paradigm even if it is not directly confirmed. In many cases, the degrees of freedom in tracer test interpretations can be decreased by using additional, independently obtained information from the laboratory, in situ geophysical measurements, etc.
- C3 Tracer tests giving results that are to some extent incompatible with the process understanding and retention paradigm, and where explanations such as faults in methodology or measuring errors can not be offered, deserve special attention. In a best case scenario the process understanding can be evolved also to explain such results. It cannot be excluded, however, that such results will weaken the current process understanding in the absence of alternative explanatory mechanisms. It is noted that some deviation of tracer test results from theoretical models can almost always be expected owing to simplifications that must be made to expedite modelling efforts. The reasons for these deviations, however, should be demonstrable in some sense or at least shown to be consistent with the available data.
-

---

**Abstraction of lumped transport parameters**

---

- D1 From cross-hole tracer tests, one can abstract lumped transport parameters (e.g.  $MPG \cdot F$  in the current diffusive-sorptive retention paradigm) from the residence time distribution if tracer recovery is obtained. These data, however, are only valid for the specific tracer test and experimental set up concerned.
- D2 The lumped parameter ( $MPG \cdot F$ ) obtained in one tracer test can not be extrapolated to other tracer tests or Safety Assessment, without including additional assumptions that are difficult to substantiate.
- D3 If either the  $MPG$  or  $F$  can be deterministically fixed by some means, information concerning the other parameter can be abstracted from the lumped parameter data. When fixing the  $MPG$ , however, material properties relating to the fracture/rock interface are needed as tracers may not penetrate into the rock matrix more than a short distance over the timescale of the test.
- 

**Abstraction of individual transport parameters**

---

- E1 Direct abstraction of constituent parameters comprising the  $MPG$ , such as  $D_e$ ,  $K_d \rho_b$ , etc is not feasible from data obtained in tracer tests in the absence of detailed corroborating data.
- E2 For in situ diffusion tests performed in the immobile zone, transport properties of the rock matrix ( $D_e$ ,  $K_d \rho_b$ , etc) can be obtained if overcoring and subsequent analysis are performed. The resolution of individual variables in such tests, however, is also usually subject to a certain degree of modelling interpretation (e.g. by way of diffusion profile interpretation, etc).
- 

**Extrapolation of numerical data to Safety Assessment**

---

- F1 Extrapolation of  $F$  from tracer tests to SA scale is generally not advisable. This is due to the fact that the hydraulic gradients and flow geometries characterising tracer tests are atypical for SA, and that much of the small-scale structure of the fracture/rock interface that may appear to be flow-wetted surface in the interpretation of a typical tracer test can be shown to become equilibrium storage capacity at longer SA timescales.
- F2 Extrapolation of the rock matrix  $MPG$  from tracer tests to SA scale is generally not advisable. This is due to that the retention properties of the rock that are sampled in tracer tests (i.e. at the immediate fracture/rock interface) is of lesser importance on an SA-scale where the retention properties of the more deeply lying rock matrix may be of greater importance for solute retardation. The apparent  $MPG$  obtained from tracer tests, if correctly evaluated, can be used to parameterise the equilibrium sorption properties of the fracture interface which is relevant for the arrival time of the very leading edge of the solute breakthrough curve in SA (i.e. estimates of "first arrival" times).
- F3  $MPG$  data abstracted from in situ diffusion tracer tests can be broadly extrapolated to SA scale, provided that the penetration depth of solute is comparable to that which would give rise to the retardation effect at the relevant SA timescale. In the case of sorbing species that are sensitive to groundwater speciation, this also requires that the future groundwater speciation is comparable to that used in the present day tracer test.
-

## 5 Conclusions

Tracer tests have played, and still play, a central role in investigations relating to the understanding of retention processes in the field. Over the last few decades, SKB has made a substantial effort to investigate radionuclide retention in, for example, the TRUE-1, TRUE Block Scale, and Stripa projects. Furthermore, progress towards developing an adequate process understanding and ways to model this has been made in the RETROCK project and Äspö Modelling Task Force on Groundwater Flow and Radionuclide Transport. At present, SKB is performing site investigations resulting in much new information of relevance for understanding of retention processes.

Much of the work performed during the 1970's and up until the early 1980's had, as a primary focus, the development of test methods rather than being strictly concerned with process understanding and data acquisition. The work carried out today owes much to this early development, which although of limited quantitative utility today, pointing the way towards the methods that are now routinely used in site investigation.

In this report we have summarised the current level of process understanding concerning radionuclide retention. Much of the knowledge has been obtained in the multidisciplinary task of Site Characterisation and Site Descriptive Modelling. Sufficient knowledge concerning radionuclide transport and retention to perform a sound Safety Assessment can only be obtained by incorporating information from all fields available. Doing this is probably as important as repeating the tracer tests carried out during the last few decades at new locations.

In situ tracer tests can either be performed in flowing systems in hydraulically conductive fractures or in the rock matrix surrounding boreholes (in situ diffusion). Complementary information about material properties can be obtained in the laboratory on drill core samples. Given the level of complexity associated with solute transport and retention that may occur over the timescale of a tracer test, we consider the utility of tracer tests in the context of evidence-based interpretations of data. In this report we have formulated a sequence of questions of increasing complexity concerning what can be done, and what cannot be done, with tracer tests. We have concluded that:

- a) Tracer tests performed in flowing systems can give a general picture of the hydrodynamic system including flow path connectivity. However, they do not provide direct information on the detailed characteristics of the flowpaths themselves;
- b) Tracer tests do lend qualitative support to the existence of retention of radionuclides in the natural barrier. The relative difference in retention between different tracers observed in *in situ* investigations is consistent with the general knowledge about how the tracers interact with geological media and the postulated mechanisms of retention for the specific tracers concerned;
- c) Tracer tests performed in flowing systems give results that are broadly compatible with the postulated processes considered relevant for solute retardation. Tracer tests performed in the rock matrix have verified the concept of matrix diffusion. Tracer tests in combination with complementary investigations have demonstrated that partitioning between dissolved radionuclides and mineral surfaces occurs. However, limited information concerning the partitioning mechanism can be extracted directly from tracer tests.

Much improvement in conceptualising solute transport during the last few decades has been a result of performing tracer tests and interpreting their results. It is likely that future tracer tests will continue to aid improvement of our present process understanding of solute transport. It should be kept in mind that parallel to in situ tracer tests, investigations concerning geology, geophysics, hydrology, etc should preferably be performed at the site at hand.

In addition, a programme for laboratory investigations of transport properties of geological material from the site should be carried out. The combined information from tracer tests and complementary investigations lend more weight to the arguments for and against postulated processes.

- d) Tracer tests performed in flowing systems may be sufficiently well resolved to make estimations of lumped retardation parameters ( $MPG \cdot F$ ). However, the lumped retention parameters obtained are only applicable to the specific tracer test over the specific timescale it is performed.
- e) From tracer tests in flowing systems it is not possible to distinguish material properties ( $MPG$  or  $K_d$ ) from flow-related transport parameters ( $F$ -factor) in a robust fashion without substantial complementary information, and neither can parameter groups be decomposed into their constituent components.

In situ diffusion tracer tests in the rock matrix are capable of determining parameters such as  $D_e$  and  $\varepsilon_p + K_d \rho$ . By combining information obtained from tracer tests performed in both flowing systems and the rock matrix, an approximate conceptualisation of the transport and retention processes involved in tracer tests can be made.

- f) Numerical results from inverse modelling of tracer tests in flowing systems cannot be extrapolated to the scale of interest for Safety Assessments without a substantial amount of complementary information. When studying retention in such tracer tests, the retention will often occur within fracture coatings, fault gouge, stagnant water volumes in the fracture plane, etc. However, on the timescale of a tracer test the penetration into the rock matrix may only be very limited for non-sorbing species and sorbing species may actually never interact with the interior of the rock matrix. The bulk of the retention of importance for SA will probably not occur in the fracture and its various components, but rather in the rock matrix. Even if one is able to obtain well-resolved and accurate retention data in a tracer test conducted in a hydraulically conductive fracture (a very ambitious task in itself), there would be no guarantees that these data would be representative, in a quantitative sense, for the retention capacity of the rock matrix in the long term. Furthermore, the flow-situation in a field tracer test is by design atypical for the flow situation under natural conditions, so it is hard to draw robust conclusions concerning the nature of SA flowpaths from these tests.

By performing diffusion tracer tests in the rock matrix, away from hydraulically conductive fractures, there is a better chance of obtaining retention properties that at least broadly represent those of the rock matrix of interest for retention in Safety Assessments. However, such tracer tests give no direct information on interactions between solutes in flowing and stagnant water.

The  $F$ -factor applicable to SA conditions will differ significantly from that obtained in tracer tests where very different hydraulic situations and flow geometries are encountered. For this reason  $F$ -factor data obtained from the evaluation of tracer test data cannot be used directly for SA purposes. Neither will the  $MPG$ -group assessed from in situ tracer tests be directly applicable in SA due to the differences in penetration depths of solutes into the rock matrix as a result of the different timescales. Therefore, the specific task that must be handled by SKB, and is being handled, is to adequately assess the parameter groups  $F$  and  $MPG$  for use in the Safety Assessment by other or complementary means and to use tracer testing as a means of evolving process understanding.

When assessing the  $F$ -factor, detailed hydraulic test data coupled with hydrogeological modelling should be used for making estimates that are relevant for the hydraulic situations and flow geometries applicable to SA conditions.

When assessing the  $MPG$ , data obtained from both laboratory and in situ studies (where appropriate) for porosity, formation factor, and  $K_d$  over a representative range of groundwater compositions should be used. It may be considered as conservative to only make use of the retention capacity of the undisturbed rock matrix and if so, this question can largely be handled

by laboratory investigations. A prerequisite is that great care is taken to control the laboratory conditions, especially for  $K_d$  measurements. By taking this conservative approach, one avoids relying on radionuclide retention in the presently poorly characterised altered rock and alteration products present at the fracture surface. However, before doing this it is important to demonstrate that neglecting retention in altered rock indeed is conservative. It has to be demonstrated that retention generally is larger in altered rock compared to the unaltered rock. It must also be demonstrated that this is conservative during major changes in groundwater chemistry, hydraulic gradients, etc, for example in the wake of a glacial period. This may also be achievable in the laboratory if the laboratory conditions are (very) well controlled and the laboratory experiments are sufficiently well thought through.

An additional objective of this report has been to highlight a number of elaborate in situ tracer tests that SKB has performed and reported which may not be widely known to the general reader. These works, which are referred to in this report, represent often extraordinarily detailed and unique investigations carried out in an international collaborative setting. If the reader is interested in further studying these tracer tests, he or she is encouraged to turn to the SKB library for hard copies. Here, a great number of SKB reports can be found that cannot be downloaded or ordered from the SKB-website.

Finally we want to state our opinion that even though field-scale tracer tests have limitations, when combined with a complete geoscientific programme enough information can be extracted to defend postulated processes in SA. Furthermore, considering all information available, data quantification can be made to a sufficient degree to demonstrate repository safety.



## Notation list

Parameter	Unit	Explanation
$C$	mol/m <sup>3</sup>	Tracer concentration of breakthrough
$C_0$	mol/m <sup>3</sup>	Tracer concentration of injection
$D_a$	m <sup>2</sup> /s (or m <sup>2</sup> /y)	Apparent diffusivity
$D_e$	m <sup>2</sup> /s (or m <sup>2</sup> /y)	Effective diffusivity
$F$	m/y	Flow-wetted surface to flow ratio (F-factor)
$F$ -factor	m/y	Flow-wetted surface to flow ratio
$F_f$	–	Formation factor
$FWS$	m <sup>2</sup>	Flow wetted surface
$k$	m <sup>-1</sup>	Inverse of the fracture half-aperture
$K_a$	m	Surface area normalised sorption partitioning coefficient
$K_d$	kg/m <sup>3</sup>	Sorption partitioning coefficient
$MPG$	m/y <sup>1/2</sup>	Material property group
$Q$	m <sup>3</sup> /y	Flow rate
$t$	y	Total residence time
$t_c$	y	Contact time
$t_m$	y	Residence time due to diffusion and sorption in the rock matrix
$t_s$	y	Residence time due to sorption on external fracture surfaces
$t_w$	y	Water residence time
$\eta$	m	Penetration depth
$\delta_f$	m	Transport aperture of a fracture
$\varepsilon_p$	–	Connected porosity of the rock matrix
$\rho_b$	kg/m <sup>3</sup>	Bulk density of the rock matrix
$\varnothing$	m	Diameter

## References

- Abelin H, Neretnieks I, 1981.** Stripa Project. Migration in a single fracture. Preliminary experiments in Stripa. SKB Stripa Project Report 81-03, Svensk Kärnbränslehantering AB.
- Abelin H, Gidlund J, 1985.** Stripa Project. Migration in a single fracture. Instrumentation and site description. SKB Stripa Project Report 85-02, Svensk Kärnbränslehantering AB.
- Abelin H, Neretnieks I, Tunbrant S, Moreno L, 1985.** Stripa Project. Final report of the migration in a single fracture – experimental results and evaluation. SKB Stripa Project Report 85-03, Svensk Kärnbränslehantering AB.
- Abelin H, Birgersson L, 1987.** Stripa Project. 3-D migration experiment – Report 1. Site preparation and documentation. SKB Stripa Project Report 87-19, Svensk Kärnbränslehantering AB.
- Abelin H, Birgersson L, Gidlund J, 1987a.** Stripa Project. 3-D migration experiment – Report 2. Instrumentation and tracers. SKB Stripa Project Report 87-20, Svensk Kärnbränslehantering AB.
- Abelin H, Birgersson L, Gidlund J, Moreno L, Neretnieks I, Widén H, Ågren T, 1987b.** Stripa Project. 3-D migration experiment – Report 3, Part I. Performed experiments, results and evaluation. SKB Stripa Project Report 87-21, Svensk Kärnbränslehantering AB.
- Abelin H, Birgersson L, Widén H, Ågren T, Moreno L, Neretnieks I, 1990.** Channeling. SKB Stripa Project Report 90-13, Svensk Kärnbränslehantering AB.
- Abelin H, Birgersson L, Widén H, Ågren T, Moreno L, Neretnieks I, 1994.** Channeling experiments in fractured crystalline rocks. *J. Contam. Hydrol*, 15, pp. 129–158.
- Allard B, Karlsson M, Tullborg E-L, Larson S Å, 1983.** Ion exchange capacities and surface areas of some major components and common fracture filling materials of igneous rock. SKB TR 83-64, Svensk Kärnbränslehantering AB.
- Almén K-E, Ekamn L, Olkiewicz A, 1978.** Försöksområdet vid Finnsjön. Beskrivning till berggrunds- och jordartskartorna. SKBF-KBS Teknisk Rapport 79-02.
- Andersson P, Klockars C-E, 1985.** Hydrogeological investigations and tracer tests in a well-defined rock mass in the Stripa mine. SKB TR 85-12, Svensk Kärnbränslehantering AB.
- Andersson J-E, Ekman L, Gustafsson E, Nordqvist R, Tirén S, 1989a.** Hydraulic interference tests and tracer tests within the Brändan area, Finnsjön study site. The fracture zone projekt – phase 3. SKB TR-89-12, Svensk Kärnbränslehantering AB.
- Andersson P, Andersson P, Gustafsson E, Olsson O, 1989b.** Investigation of flow distribution in a fracture zone at the Stripa mine, using the radar method, results and interpretations. SKB TR-89-33, Svensk Kärnbränslehantering AB.
- Andersson P, Nordqvist R, Persson T, Eriksson C-O, Gustafsson E, Ittner T, 1993.** Dipole tracer experiment in a low-angle fracture zone at Finnsjön – results and interpretation. The Fracture Zone Project – Phase 3. SKB TR-93-26, Svensk Kärnbränslehantering AB.
- Andersson P, 1995.** Compilation of tracer tests in fractured rock. SKB internal document PR-25-95-05, Svensk Kärnbränslehantering AB.
- Andersson P, Ludvigsson J-E, Wass E, Holmqvist M, 2000a.** Interference tests, dilution tests and tracer tests, Phase A. Äspö Hard Rock Laboratory. SKB IPR-00-28, Svensk Kärnbränslehantering AB.

- Andersson P, Wass E, Holmqvist M, Fierz T, 2000b.** True Block Scale Project. Tracer tests, Phase B. Äspö Hard Rock Laboratory. SKB IPR-00-29, Svensk Kärnbränslehantering AB.
- Andersson P, Byegård J, Holmqvist M, Skålberg M, Wass E, Widestrand H, 2001.** True Block Scale Project. Tracer tests, Phase C. Äspö Hard Rock Laboratory. SKB IPR-01-33, Svensk Kärnbränslehantering AB.
- Andersson P, Byegård J, Dershowitz B, Doe T, Hermanson J, Meier P, Tullborg E-L, Winberg A, 2002a.** Final report of the TRUE Block Scale projekt 1. Characterisation and model development. SKB TR-02-13, Svensk Kärnbränslehantering AB.
- Andersson P, Byegård J, Winberg A, 2002b.** Final report of the TRUE Block Scale project 2. Tracer tests in the block scale. SKB TR-02-14, Svensk Kärnbränslehantering AB.
- Andersson P, Wass E, Gröhn S, Holmqvist M, 2002c.** Äspö Hard Rock Laboratory. TRUE-1 continuation project. Complementary investigations at the TRUE-1 site – Crosshole interference, dilution and tracer tests, CX-1–CX-5. SKB IPR-02-47, Svensk Kärnbränslehantering AB.
- Andersson P, Gröhn S, Nordqvist R, Wass E, 2004.** TRUE Block Scale continuation project. BS2B pretests. Crosshole interference, dilution and tracer tests, CPT-1–CPT-4. Äspö Hard Rock Laboratory. SKB IPR-04-25, Svensk Kärnbränslehantering AB.
- Andersson P, Byegård J, Nordqvist R, Wass E, 2005.** TRUE Block Scale continuation project. BS2B tracer tests with sorbing tracers. Äspö Hard Rock Laboratory. SKB IPR-05-01, Svensk Kärnbränslehantering AB.
- Andersson P, Byegård J, Billaux D, Cvetkovic V, Dershowitz W, Doe T, Hermanson J, Poteri A, Tullborg E-L, Winberg A (ed), 2007.** TRUE Block Scale Continuation Project. Final Report. SKB TR-06-42, Svensk Kärnbränslehantering AB.
- Arthur D Little Inc, 1962.** Electrical conductivity, compressibility, and viscosity of aqueous electrolytical solutions. Project Trident Technical Report S-7001-0307. Department of the Navy, Bureau of Ships, USA.
- Axe L, Trivedi P, Anderson P, 2002.** Diffusion at oxide and related surfaces. Encyclopedia of surface and colloid science. Marcel Dekker, Inc.
- Bargar J, Brown G, Parks G, 1997.** Surface complexation of Pb(II) at oxide-water interfaces: I. XAFS and bond valence determination of mononuclear and polynuclear Pb(II) sorption products on aluminium oxides. *Geochim. Cosmochim. Acta*, 61(13), pp. 2617–2673.
- Bargar J, Reiteyer R, Davis J, 1999.** Spectroscopic confirmation of uranium(VI)-carbonate adsorption complexes on hematite. *Environmental Science & Technology*, 36(5), pp. 2481–2484.
- Bargar J, Reitmeyer R, Lenhart J, Davis J, 2000.** Characterization of U(VI)-carbonato ternary complexes on hematite: EXAFS and electrophoretic mobility measurements. *Geochim. Cosmochim. Acta*, 64(16), pp. 2737–2749.
- Becker M W, Shapiro A M, 2000.** Tracer transport in fractured crystalline rock: Evidence of nondiffusive breakthrough tailing. *Water Resour. Res.*, 36(7), pp. 1677–1686.
- Benson D A, Wheatcraft S W, Meerschaert M M, 2000.** Application of a fractional advection-dispersion equation. *Water Resour. Res.*, 36(6), pp. 1403–1412.
- Berkowitz B, Kosakowski G, Margolin G, Scher H, 2001.** Application of continuous time random walk theory to tracer test measurements in fractured and heterogeneous porous media. *Ground Water*, 39(4), pp. 593–604.
- Berkowitz B, 2002.** Characterizing flow and transport in fractured geological media: A review. *Adv. Water Resour.*, 25, pp. 861–884.

- Birgersson L, Neretnieks I, 1990.** Diffusion in the matrix of granitic rock: Field test in the Stripa mine. *Water Resour. Res.*, Vol. 26 (11), pp. 2833–2842.
- Birgersson L, Ågren T, 1992.** Stripa Project. Site characterization and validation – Tracer migration experiment in the validation drift, Report 1: Instrumentation, site preparation and tracers, SKB Stripa Project Report 92-02, Svensk Kärnbränslehantering AB.
- Birgersson L, Widén H, Ågren T, Neretnieks I, 1992a.** Stripa Project. Tracer migration experiments in the Stripa mine 1980–1991. SKB Stripa Project Report 92-25, Svensk Kärnbränslehantering AB.
- Birgersson L, Widén H, Ågren T, Neretnieks I, Moreno L, 1992b.** Stripa Project. Site characterization and validation – Tracer migration experiment in the validation drift, Report 2, Part 1: Performed experiments, results and evaluation, SKB Stripa Project Report 92-03, Svensk Kärnbränslehantering AB.
- Black J, Hodgkinson D, 2005.** Äspö Task Force on Modelling of Groundwater Flow and Transport of Solutes: Review of Task 6C. SKB R-05-33, Svensk Kärnbränslehantering AB.
- Bodin J, Delay F, de Marsily G, 2003.** Solute transport in a single fracture with negligible matrix permeability: 1. fundamental mechanisms. *J. Hydrogeol.*, 11, pp. 418–433.
- Bostick B, Vairavamurthy M, Karthikeyan K, Chorover J, 2002.** Cesium adsorption on clay minerals: An EXAFS spectroscopic investigation. *Environ. Sci. Technol.*, 36, pp. 2670–2676.
- Bradbury M H, Green A, 1986.** Investigations into the factors influencing long range matrix diffusion rates and pore space accessibility at depth in granite. *J. Hydrol.*, 89, pp. 123–139.
- Brantley S L, Mellott N P, 2000.** Surface area and porosity of primary silicate minerals, *American Mineralogist*, 85(11–12), pp. 1767–1783.
- Bryant S L, Thompson K E, 2001.** Theory, modeling and experiment in reactive transport in porous media. *Curr. Opin. Colloid Interface Sci.*, 6, pp. 217–222.
- 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, Johansson H, Andersson P, Hansson K, Winberg A, 1999.** Äspö Hard Rock Laboratory. Test plan for the long term diffusion experiment. SKB IPR-99-36, Svensk Kärnbränslehantering AB.
- Byegård J, Gustavsson E, Tullborg E-L, 2005.** Bedrock transport properties: Preliminary site description Simpevarp subarea – version 1.2, SKB R-05-05, Svensk Kärnbränslehantering AB.
- Byegård J, Gustavsson E, Tullborg E-L, 2006.** Bedrock transport properties: Preliminary site description Laxemar subarea – version 1.2, SKB R-06-27, Svensk Kärnbränslehantering AB.
- Börjesson S, Gustavsson E, 2005a.** Oskarshamn site investigation: Laboratory data from the site investigation programme for the transport properties of the rock. Data delivery for data freeze Laxemar 2.1. SKB P-05-106, Svensk Kärnbränslehantering AB.
- Börjesson S, Gustavsson E, 2005b.** Forsmark site investigation: Laboratory data from the site investigation programme for the transport properties of the rock. Data delivery for data freeze Forsmark 2.1. SKB P-05-109, Svensk Kärnbränslehantering AB.
- Chapman N, McKinley I, Penna Franca E, Shea M, Smellie J, 1992.** The Pocos de Caldas project: an introduction and summary of its implications for radioactive waste disposal. *J. Geochem. Explor.*, 45(1–3), pp. 1–24.

- Cortis A, Gallo C, Scher H, Berkowitz B, 2004.** Numerical simulation of non-Fickian transport in geological formations with multiple-scale heterogeneities, *Water Resour. Res.*, 40, pp. 1–16.
- Crank J, 1975.** *The Mathematics of Diffusion*, 2 ed, Oxford University Press, UK
- Crawford J, 2006.** Modelling in support of bedrock transport property assessment. Preliminary site description Laxemar subarea – version 1.2. SKB R-06-28, Svensk Kärnbränslehantering AB.
- Crawford J, Neretnieks I, Malmström M, 2006.** Data and uncertainty 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.
- Cvetkovic V, Selroos J. O, Cheng H, 1999.** Transport of reactive tracers in rock fractures. *J. Fluid Mech.*, 378, pp. 335–356.
- Cvetkovic V, Cheng H, 2002.** Äspö Hard Rock Laboratory. TRUE Block Scale Project. Evaluation of block scale tracer retention understanding experiments at Äspö HRL. SKB IPR-02-33, Svensk Kärnbränslehantering AB.
- Dentz M, Cortis A, Scher H, Berkowitz B, 2004.** Time behaviour of solute transport in heterogeneous media: transition from anomalous to normal transport. *Adv. Water Resour.*, 27, pp. 155–173.
- Dershowitz W, Winberg A, Hermanson J, Byegård J, Tullborg E-L, Andersson P, Mazurek M, 2003.** Äspö Hard Rock Laboratory. Äspö Task Force on modeling of groundwater flow and transport of solutes. Task 6C. A semi-synthetic model of block scale conductive structures at the Äspö HRL. SKB IPR-03-13, Svensk Kärnbränslehantering AB.
- Desbrandes R, 1985.** *Encyclopedia of well logging*. Translated by Brace G. Graham & Trotman Ltd.
- Dähn R, Scheidegger A, Manceau A, Schlegel M, Baeyens B, Bradbury M, Chateigner D, 2003.** Structural evidence for the sorption of Ni(II) atoms on the edges of montmorillonite clay minerals: A polarized X-ray absorption fine structure study. *Geochim. et Cosmochim. Acta*, 67(1), pp. 1–15.
- Elert M, Svensson H, 1999.** Äspö Hard Rock Laboratory. Deconvolution of breakthrough curves from TRUE-1 tracer tests (STT-1 and STT-1b) with sorbing tracers. Äspö Task Force, Task 4E. SKB IPR-99-35, Svensk Kärnbränslehantering AB.
- Elert M, Svensson H, 2000.** Äspö Hard Rock Laboratory. Deconvolution of breakthrough curves from TRUE-1 tracer tests (STT-2) with sorbing tracers. Äspö Task Force, Task 4F. SKB IPR-00-22, Svensk Kärnbränslehantering AB.
- Elert M, Svensson H, 2001.** Evaluation of modelling of the TRUE-1 radially converging tests with sorbing tracers. The Äspö Task Force on modelling of groundwater flow and transport of solutes. Tasks 4E and 4F. SKB TR-01-12, Svensk Kärnbränslehantering AB.
- Eriksen T, Locklund B, 1987.** Radionuclide sorption on granitic drill core material. SKB TR-87-22, Svensk Kärnbränslehantering AB.
- Eriksen T, Locklund B, 1989.** Radionuclide sorption on crushed and intact granitic rock. Volume and surface effects. SKB TR-89-25, Svensk Kärnbränslehantering AB.
- Fairhurst C, Ferruccio G, Gnirk P, Gray M, Stillborg B, 1993.** *Stripa Project Overview Report. Vol. I: Executive summary*. OECD/NEA International Stripa Project 1980–1992. ISBN: 91-971906-2-4.

- Farquar M, Vaughan D, Hughes C, Charnock J, England K, 1997.** Experimental studies of the interaction of aqueous metal cations with mineral substrates: Lead, cadmium, and copper with perthitic feldspar, muscovite, and biotite. *Geochim. et Cosmochim. Acta*, 61(15), pp. 3051–3064.
- Gelhar L W, Welty C, Rehfeldt K R, 1992.** A critical review of data on field-scale dispersion in aquifers. *Water Resour. Res.*, 28(7), pp. 1955–1974.
- Gustafsson E, Klockars C-E, 1981.** Studies on groundwater transport in fractured crystalline rock under controlled conditions using nonradioactive tracers. SKBF/KBS Technical Report 81-07, Svensk Kärnbränslehantering AB.
- Gustafsson E, Klockars C-E, 1984.** Study of strontium and cesium migration in fractured crystalline rock. SKBF/KBS Technical Report 84-07, Svensk Kärnbränslehantering AB.
- Gustafsson E, Andersson P, 1991.** Groundwater flow conditions in a low-angle fracture zone at Finnsjön, Sweden. *Journal of Hydrology*. Vol 126, pp. 79–111.
- Gustafsson E, Nordqvist R, 1993.** Radially converging tracer test in a low-angle fracture zone at the Finnsjön site, central Sweden. The Fracture Zone Project – Phase 3. SKB TR-93-25, Svensk Kärnbränslehantering AB.
- Gustafsson E, Ludvigson J-E, 2005.** Combined interference test and tracer test between KLX02 and HLX10. Oskarshamn site investigation. SKB P-05-20, Svensk Kärnbränslehantering AB.
- Gustafsson E, Nordqvist R, 2005.** Groundwater flow measurements and SWIW tests in boreholes KLX02 and KSH02. Oskarshamn site investigation. SKB P-05-28, Svensk Kärnbränslehantering AB.
- Gustafsson E, 2006 (in prep.)** Äspö Hard Rock Laboratory: Project Plan for LTDE Sorption Diffusion Experiment (LTDE-SD). Svensk Kärnbränslehantering AB.
- Gustafsson E, Nordqvist R, Thur P, 2006a.** Groundwater flow measurements and SWIW test in borehole KFM08A. Forsmark site investigation. SKB P-06-90, Svensk Kärnbränslehantering AB.
- Gustafsson E, Nordqvist R, Thur P, 2006b.** Groundwater flow measurements and SWIW test in borehole KFM04A. Forsmark site investigation. SKB P-06-141, Svensk Kärnbränslehantering AB.
- Gnirk P, 1993.** Stripa Project Overview Report. Vol. II: Natural barriers. OECD/NEA International Stripa Project 1980–1992. ISBN: 91-971906-3-2
- Haggerty R, Gorelick S M, 1995.** Multiple-rate mass transfer for modelling diffusion and surface reactions in media with pore-scale heterogeneity. *Water Resour. Res.*, 31(10), pp. 2383–2400.
- Haggerty R, 1999.** Application of the multirate diffusion approach in tracer test studies at Äspö HRL. SKB R-99-62, Svensk Kärnbränslehantering AB.
- Haggerty R, 2002.** Matrix diffusion: heavy-tailed residence time distribution and their influence on radionuclide retention. Radionuclide retention in geologic media, workshop proceedings. Nuclear Energy Agency. ISBN 92-64-19695-1.
- Hansen L M, Staub I, 2004.** Äspö Hard Rock Laboratory. TRUE-1 Continuation Project. Fault rock zones characterisation. Overcoring (300 mm) of impregnated fault rock zones at chainages 2/430, 2/545, 2/163 and 1/600m. SKB IPR-04-10, Svensk Kärnbränslehantering AB.
- Heath M J, Montoto M, Rodriguez Rey A, Ruiz de Argandoña V G, Menendez B, 1992.** Rock matrix diffusion as a mechanism of radionuclide retention: a natural analogue study of El Berrocal granite, Spain. *Radiochimica Acta* 58–59, 379–384.

- Hennig C, Reich T, Dähn R, Scheidegger A, 2002.** Structure of uranium sorption complexes at montmorillonite edge sites. *Radiochimica Acta*, 90(9–11), pp. 653–657.
- Hermanson J, Doe T, 2000.** Äspö Hard Rock Laboratory. March'00 structural and hydraulic model based on borehole data from KI0025F03. SKB IPR-00-34, Svensk Kärnbränslehantering AB.
- Hodgkinson D, Black J, 2005.** Äspö Task Force on modelling of groundwater flow and transport of solutes. Review of Tasks 6A, 6b and 6B2. SKB TR-05-14, Svensk Kärnbränslehantering AB.
- Hodgkinson D, 2007.** Äspö Task Force on Modelling of Groundwater Flow and Transport of Solutes: Review of Tasks 6D, 6E, 6F and 6F2. SKB TR-07-XX, Svensk Kärnbränslehantering AB.
- Hölttä P, Siitari-Kauppi M, Huikuri P, Lindberg A, Hautojärvi A, 1997.** The effect of specific surface area on radionuclide sorption on crushed crystalline rock, *Mat. Res. Soc. Symp. Proc*, 465, pp. 789–796.
- 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.
- Jenne E A (ed), 1998.** Adsorption of metals by geomedial. Variables, mechanisms, and model applications. Academic Press, New York.
- Johansson H, Siitari-Kauppi M, Skålberg M, Tullborg E-L, 1998.** Diffusion pathways in crystalline rock – examples from Äspö-diorite and fine-grained granite. *J. Contam. Hydrol*, 35(1–3), pp. 41–53.
- Kohl T, Evans K F, Hopkirk R J, Jung R, Rybach L, 1997.** Observation and simulation of non-Darcian flow transients in fractured rock. *Water Resour. Res*, 33(3), pp. 407–418.
- Kosakowski G, Berkowitz B, 1999.** Flow pattern variability in natural fracture intersections. *Geophys. Res. Lett*, 26(12), pp. 1765–1768.
- Krishna R, Wesselingh J A, 1997.** The Maxwell-Stefan approach to mass transfer. *Chemical Engineering Science*, vol. 52, no. 6, pp. 861–911.
- Laaksoharju M, Smellie J, Gimeno M, Auqué L, Gómez J, Tullborg E-L, and Gurban I, 2004.** Titel Hydrogeochemical evaluation of the Simpevarp area, model version 1.1. SKB R-04-16, Svensk Kärnbränslehantering AB.
- Landström O, Klockars C-E, Holmberg K-E, Westerberg S, 1978.** In situ experiments on nuclide migration in fractured crystalline rocks. KBS Technical Report 110, Svensk Kärnbränslehantering AB.
- Landström O, Klockars C-E, Persson O, Tullborg E-L, Larson S Å, Andersson K, Allard B, Torstenfelt B, 1983.** Migration experiments in Studsvik. SKBF/KBS Technical Report 83-18, Svensk Kärnbränslehantering AB.
- Lichtner P C, Steefel C I, Oelkers E H, 1996.** Reactive transport in porous media. *Reviews in Mineralogy Volume 34*, Mineralogical Society of America, Washington, DC.
- 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, 2004.** Diffusive properties of granitic rock as measured by in-situ electrical methods. Doctoral thesis at the Royal Institute of Technology, Stockholm, Sweden. ISBN 91-7283-935-X.

- Löfgren M, Neretnieks I, 2005.** 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.
- Löfgren M, 2006.** Forsmark site investigation. Formation factor logging in-situ by electrical methods in KFM07A and KFM08A. SKB P-06-91, Svensk Kärnbränslehantering AB.
- Löfgren M, Neretnieks I, 2006.** Through-electromigration: A new method of investigating pore connectivity and obtaining formation factors. *J. Contam. Hydrol*, Vol. 87(3–4), pp. 237–252.
- Margolin G, Berkowitz B, 2000.** Application of continuous time random walks to transport in porous media. *J. Phys. Chem. B*, 104, pp. 3942–3947.
- Marimon M M, 2002.** On sorption and diffusion of radionuclides in bentonite clay. Doctoral thesis at the Royal Institute of Technology, Stockholm, Sweden. ISSN: 0349-6465.
- Marschall P, Elert M, 2003.** Overall evaluation of the modelling of the TRUE-1 tracer tests – Task 4. The Äspö Task Force on modelling of groundwater flow and transport of solutes. SKB TR-03-12, Svensk Kärnbränslehantering AB.
- McKinley I G, Scholtis A, 1993.** A comparison of radionuclide sorption databases used in recent performance assessments. *J. Contam. Hydrol*, 13(1–4), pp. 347–363.
- Miller W, Alexander R, Chapman N, McKinley I, Smellie J, 1994.** Natural analogue studies in the geological disposal of radioactive wastes, *Studies in Environmental Science* 57, 395 pp, Elsevier, Amsterdam.
- Miller W, Alexander R, Chapman N, McKinley I, Smellie J, 2000.** Geological disposal of radioactive wastes and natural analogues. *Lessons from nature and Archaeology*, 316 pp, Pergamon Press, Elsevier, Amsterdam.
- Milnes A G, 2002.** Swedish deep repository siting programme Guide to the documentation of 25 years of geoscientific research (1976–2000). SKB TR-02-18, Svensk Kärnbränslehantering AB.
- Moreno L, Neretnieks I, Klockars C-E, 1983.** Evaluation of some tracer tests in the granitic rock at Finnsjön. SKBF/KBS Technical Report 83-38, Svensk Kärnbränslehantering AB.
- Moreno L, Tsang C F, 1991.** Multiple-peak response to tracer injection tests in single fractures: A numerical study. *Water. Resour. Res.*, 27(8), pp. 2143–2150.
- Moreno L, Neretnieks I, 1993.** Fluid flow and solute transport in a network of channels. *J. Contam. Hydrol*, 14(3–4), pp. 163–192.
- 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.
- NEA, 2001.** Using thermodynamic sorption models for guiding radioelement distribution coefficient ( $K_d$ ) investigations. OECD-NEA.
- NEA, 2005.** NEA Sorption Project Phase II. Interpretation and prediction of radionuclide sorption onto substrates relevant for radioactive disposal using thermodynamic sorption models, OECD-NEA.
- Neretnieks I, 1980.** Diffusion in the rock matrix: An important factor in radionuclide retardation. *J. Geophys. Res.*, 85(8), pp. 4397–4379.
- Neretnieks I, 1993.** Solute Transport in Fractured Rock – Applications to Radioactive Waste Repositories. Chapter 3 book. Ed Bear J, de Marsily G, Tsang C-F. Academic Press, pp. 39–127.



- Neretnieks I, Moreno L, 2003.** Prediction of some in situ tracer tests with sorbing tracers using independent data. *J. Contam. Hydrol*, 61(1–4), pp. 351–360.
- Neretnieks I, 2006.** Channeling with diffusion into stagnant water and into a matrix in series. *Water Resour. Res.*, 42(W11418), pp. 1–15.
- Neretnieks I, 2007** (in prep.) Single Well Injection Withdrawal Tests (SWIW) in Fractured Rock – Some Aspects on Interpretation. Department of Chemical Engineering and Technology, Royal Institute of Technology, Stockholm, Sweden.
- Neuman S P, 2005.** Trends, prospects and challenges in quantifying flow and transport through fractured rocks. *J. Hydrogeol*, 13, pp. 124–147.
- Nielsen U T, Ringgaard J, Fris Dahl J, 2005.** Forsmark site investigation. Geophysical borehole logging in the boreholes KFM07A, KFM08A and KFM08B. SKB P-05-159, Svensk Kärnbränslehantering AB.
- Nordqvist R, Gustafsson E, 2002.** Single-well injection-withdrawal tests (SWIW). Literature review and scoping calculations for homogeneous crystalline bedrock conditions. SKB R-02-34, Svensk Kärnbränslehantering AB.
- Ohlsson Y, Neretnieks I, 1998.** Some evidence for surface ion mobility in rock. *J. Contam. Hydrol*, Vol. 35, pp. 91–100.
- Ohlsson Y, 2000.** Studies of Ionic Diffusion in Crystalline Rock. Doctoral thesis at the Royal Institute of Technology, Stockholm, Sweden. ISBN 91-7283-025-5.
- Olsson O, Andersson P, Gustafsson E, 1991.** Site characterization and validation – monitoring of saline tracer transport by borehole radar measurements, final report. SKB Stripa Project Report 91-18, Svensk Kärnbränslehantering AB.
- Petersson J, Skogsmo G, Berglund J, Wängnerud A, Danielsson P, Stråhle A, 2005.** Boremap mapping of telescopic drilled borehole KFM08A and core drilled borehole KFM08B. Forsmark site investigation. SKB P-05-203, Svensk Kärnbränslehantering AB.
- Poteri A, Billaux D, Dershowitz W, Gómez-Hernández J-J, Cvetkovic V, Hautojärvi A, Holton D, Medina A, Winberg A, 2002.** Final report of the TRUE Block Scale project, 3. Modelling of flow and transport, SKB Technical Report TR-02-15. Svensk Kärnbränslehantering AB.
- Press F, Siever R, 1998.** *Understanding earth*, 2nd ed. W. H. Freeman and Company. ISBN 0-7167-2836-2.
- Rabung T, Stumpf T, Geckeis H, Klenze R, Kim J, 2000.** Sorption of Am(III) and Eu(III) onto  $\alpha$ -alumina: experiment and modelling. *Radiochim. Acta*, 88(9/11), pp. 711–716.
- Rasilainen K, 1997.** Matrix diffusion model: In-situ tests using natural analogues. Technical Research Center of Finland. VTT Publication 331. ISBN 951-38-5209-1.
- Rasmuson A, Neretnieks I, 1986.** Radionuclide transport in fast channels in crystalline rock. *Water Resour. Res.*, 22(8), pp. 1247–1256.
- Rhén I, Svensson U, Andersson J-E, Andersson P, Eriksson C-O, Gustafsson E, Ittner T, Nordqvist R, 1992.** Äspö hard rock laboratory: Evaluation of the combined longterm pumping and tracer test (LTP2) in borehole KAS06. SKB TR-92-32, Svensk Kärnbränslehantering AB.
- Saadatfar M, Sahimi M, 2002.** Diffusion in disordered media with long-range correlations: Anomalous, Fickian, and Superdiffusive transport and log-periodic oscillations, *Phys. Rev. E*, 65, pp. 1–8.

- Sahimi M, 1995.** Flow and transport in porous media and fractured rock: From classical methods to modern approaches. John Wiley & Sons, New York.
- Sandström B, Tullborg E-L, 2005.** Forsmark site investigation. Fracture mineralogy. Results from fracture minerals and wall rock alteration in boreholes KFM01B, KFM04A, KFM05A and KFM06A. SKB P-05-197, Svensk Kärnbränslehantering AB.
- Selroos J-O, Walker D D, Ström A, Gylling B, Follin S, 2002.** Comparison of alternative modelling approaches for groundwater flow in fractured rock. *J. Hydrol*, 257, pp. 174–188.
- SICADA, 2006.** Site Characterisation Data Base, Svensk Kärnbränslehantering AB.
- Skagius K, 1986.** Diffusion of some dissolved species in the matrix of some Swedish crystalline rocks. Ph.D. Thesis, Department of Chemical Engineering and Technology, Royal Institute of Technology, Stockholm.
- SKB, 1989.** Executive summary of phase 2. Stripa Project Technical Report 89-01. Svensk Kärnbränslehantering AB.
- SKB, 2004.** RETROCK Project. Treatment of geosphere retention phenomena in Safety Assessments. Scientific basis of retention processes and their implementation in Safety Assessment models (WP2). Work Package 2 report of the RETROCK Concerted Action. SKB R-04-48, Svensk Kärnbränslehantering AB.
- SKB, 2005a.** Preliminary site description. Simpevarp subarea – version 1.2. SKB R-05-08, Svensk Kärnbränslehantering AB.
- SKB, 2005b.** Preliminary site description. Forsmark area – version 1.2. SKB R-05-18, Svensk Kärnbränslehantering AB.
- SKB, 2005c.** Äspö Hard Rock Laboratory. Annual Report 2004. SKB TR-05-10, Svensk Kärnbränslehantering AB.
- SKB, 2005d.** TRUE-1 Continuation Project, TRUE-1 Completion, Complementary tracer tests, SWIW and CEC, at the TRUE-1 site. SKB internal controlling document AP TD F83-05-039, Svensk Kärnbränslehantering AB.
- SKB, 2006a.** Preliminary site description. Laxemar subarea – version 1.2. SKB R-06-10, Svensk Kärnbränslehantering AB.
- SKB, 2006b.** 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, 2006c.** Data report for the Safety assessment SR-Can. SKB TR-06-25, Svensk Kärnbränslehantering AB.
- SKB, 2006d.** Long-term safety for KBS-3 repositories at Forsmark and Laxemar – a first evaluation. Main report of the SR-Can project. SKB TR-06-09, Svensk Kärnbränslehantering AB.
- SKB, 2006e.** Preliminary safety evaluation for the Laxemar subarea. Based on data and site descriptions after the initial site investigation stage. SKB TR-06-06, Svensk Kärnbränslehantering AB.
- SKB, 2006f.** Äspö Hard Rock Laboratory. Annual report 2005. SKB TR-06-10, Svensk Kärnbränslehantering AB.
- SKB, 2006g.** The Äspö Hard Rock Laboratory. Information brochure. Svensk Kärnbränslehantering AB.

- SKB, 2007a.** Äspö Hard Rock Laboratory. Planning Report for 2007. SKB IPR-07-06, Svensk Kärnbränslehantering AB.
- SKB, 2007b.** Interferenstest och spårämnesförsök med sorberande spårämnen i zon A2 på borrhälsplats 2. SKB internal controlling document AP PF 400-07-013, Svensk Kärnbränslehantering AB.
- Snodgrass M F, Kitandis P K, 1998.** A method to infer in situ reaction rates from push-pull experiments. *Ground Water* 36, no. 4: 645–650.
- Sokolnicki M, Rouhiainen P, 2005.** Difference flow logging in borehole KFM08A. Forsmark site investigation. SKB P-05-43, Svensk Kärnbränslehantering AB.
- Strawn D, Sparks D, 1999.** The use of XAFS to distinguish between inner- and outer-sphere lead adsorption complexes on montmorillonite. *J. Colloid Interface Sci*, 216(2), pp. 257–269.
- Stumm W, Morgan J, 1996.** *Aquatic Chemistry: Chemical Equilibria and Rates in natural Waters*, 3 ed, Wiley, New York.
- Stumpf T, Bauer A, Coppin F, Kim J, 2001.** Time-resolved laser fluorescence spectroscopy study of the sorption of Cm(III) onto smectite and kaolinite. *Environ. Sci. Technol*, 35(18), pp. 3691–3694.
- Stumpf T, Fanghänel T, 2002.** A time-resolved laser fluorescence spectroscopy (TRLFS) study of the interaction of trivalent actinides (Cm(III)) with calcite. *J. Colloid Interface Sci*, 249(1), pp. 119–122.
- Stumpf T, Bauer A, Coppin F, Fanghänel T, Kim J, 2002.** Inner-sphere, outer-sphere, and ternary surface complexes: a TRLFS study of the sorption process of Eu(III) onto smectite and kaolinite. *Radiochimica Acta*, 90(6), pp. 345–349.
- Svensson U, Kuylenstierna H-O, Ferry M, 2004.** DarcyTools, Version 2.1. Concepts, methods, equations and demo simulations. SKB R-04-19, Svensk Kärnbränslehantering AB.
- Suzuki K, Oda M, Kuwahara T, Hiramata K, 1995.** Material property changes in granitic rock during long-term immersion in hot water. *Engineering Geology*, Vol. 40, pp. 29–39.
- Sylwester E R, Hudson E A, Allen P G, 2000.** The structure of uranium(VI) sorption complexes on silica, alumina, and montmorillonite. *Geochim. Cosmochim. Acta*, 64(14), pp. 2431–2438.
- Thunehed H, 2005.** Forsmark site investigation: Resistivity measurements on samples from KFM01A and KFM02A. SKB P-05-26, Svensk Kärnbränslehantering AB.
- Torstenfelt B, Ittner T, Allard B, Andersson K, Olofsson U, 1982.** Mobilities of radionuclides in fresh and fractured crystalline rock. SKB TR-82-26, Svensk Kärnbränslehantering AB.
- Torstenfelt B, Eliasson T, Allard B, Andersson K, Höglund S, Ittner T, 1983.** Radionuclide migration into natural fracture surfaces of granitic rock. In: D. G. Brookins ed, *Scientific Basis for Nuclear Waste Management – VI*, Elsevier, New York.
- Tsang Y W, Tsang C F, 1989.** Flow channeling in a single fracture as a two-dimensional strongly heterogeneous permeable medium. *Water Resour. Res*, 25(9), pp. 2076–2080.
- Tsang C F, Tsang Y W, Hale F V, 1991.** Tracer transport in fractures: Analysis of field data based on a variable-aperture channel model. SKB Stripa Project Report 91-14, Svensk Kärnbränslehantering AB.
- 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. *Journal of Contaminant Hydrology*. Vol. 61, pp. 191–202.

- Waber H N, Smellie J A T, 2005.** Forsmark site investigation. Borehole KFM06A. Characterisation of pore water. Part I: Diffusion experiments. SKB P-05-196, Svensk Kärnbränslehantering AB.
- Wahlgren C-H, Ahl M, Sandahl K-A, Berglund J, Petersson J, Ekström M, Persson P-O, 2004.** Oskarshamn site investigation. Bedrock mapping 2003 – Simpevarp subarea. Outcrop data, fracture data, modal and geochemical classification of rock types, bedrock map, radiometric dating. SKB P-04-102, Svensk Kärnbränslehantering AB.
- Wass E, Andersson P, 2006.** Forsmark site investigation. Groundwater flow measurements and tracer tests at drill site 1. SKB P-06-125, Svensk Kärnbränslehantering AB.
- Westall J, 1995.** Modeling of the speciation of metal ions with heterogeneous environmental sorbents. *Mat. Res. Soc. Symp. Proc.*, 353, pp. 937–950.
- Widestrand H, Byegård J, Ohlsson Y, Tullborg E-L, 2003.** Strategy for the use of laboratory methods in the site investigations programme for the transport properties of the rock. SKB R-03-20, Svensk Kärnbränslehantering AB.
- Winberg A, 1997.** Test plan for the TRUE Block Scale Experiment. Swedish Nuclear Fuel and Waste Management Company. Äspö Hard Rock Laboratory. SKB ICR 97-02, Svensk Kärnbränslehantering AB.
- Winberg A, Andersson P, Hermanson J, Byegård J, Cvetkovic V, Birgersson L, 2000.** Äspö Hard Rock Laboratory. Final report of the first stage of the tracer retention understanding experiments. SKB TR-00-07, Svensk Kärnbränslehantering AB.
- Winberg A, Andersson P, Byegård J, Poteri A, Cvetkovic V, Dershowitz W, Doe T, Hermanson J, Gómez-Hernández J, Hautojärvi A, Billaux D, Tullborg E-L, Holton D, Meier P, Medina A, 2003.** Final report of the TRUE Block Scale project. 4. Synthesis of flow, transport and retention in the block scale, SKB TR-02-16, Svensk Kärnbränslehantering AB.