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Single well injection withdrawal tests (SWIW) in fractured rock

Some aspects on interpretation

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Summary

Single-Well-Injection-Withdrawal, SWIW, tests are used to try to extract information on fracture apertures, sorption and diffusion properties and dispersion information in individual fractures. It is done by injecting a given amount of traced water into an isolated fracture. After a waiting period water is withdrawn from the fracture and the tracer concentration is measured. The concentration time curve is fitted to a model and the parameter values quantifying the different interaction mechanisms are determined. A number of different mechanisms influence the recovery of the tracer. One or more of the following mechanisms are considered. They include: dispersion due to velocity differences, sorption on fracture surface and on infill, diffusion in rock fragments in the fracture, diffusion between “streamlines”, diffusion into rock matrix and other stagnant water volumes, sorption kinetics and slow drift of the plume caused by the natural gradient.

Many of the interaction mechanisms can influence the recovery curve in a similar way. For example, diffusion into rock matrix water and into stagnant water in the fracture adjacent to the flowing channels cannot be distinguished if only one tracer is used. Tracers with different properties can in principle be used but they will encounter different parts of the fracture, the sorbing tracer will move out less from the injection point than a nonsorbing tracer will. Diffusion and sorption in small particles in the flowpath can influence the recovery curve in a similar way as rock matrix diffusion does.

Dispersion caused by diffusion between “streamlines”, Taylor dispersion, can give very different results in channels of different shapes. Such dispersion effects can be difficult to distinguish from matrix diffusion effects. Dispersion coefficients obtained in a SWIW test may have little relation to dispersion of a tracer moving from A to B. This is partly due to the different mechanisms and partly due to different time scales.

The short duration of a SWIW test can not be expected to have been influenced in the same way by the different mechanisms that are active over the long time scales of interest for safety analysis.

Because many different mechanisms influence the recovery curve in a similar way any parameter values extracted from SWIW tests must be treated with considerable caution and afterthought.

Sammanfattning

Så kallade Single-Well-Injection-Withdrawal, SWIW, tester används för att söka experimentellt bestämma sprickvidder samt sorptions-, diffusions- och dispersionsdata hos enskilda sprickor. I försöken injiceras vatten med ett spårämne i en spricka. Efter en väntetid pumpas vattnet tillbaka och halten spårämne mäts. Spårämnehaltens ändring med återpumpad mängd vatten används för att bestämma parametervärden i någon modell som beskriver hur spårämnet påverkas av olika mekanismer. Ett antal olika mekanismer påverkar spårämnets uppförande. En eller fler av följande mekanismer brukar modelleras: dispersion förorsakad av hastighetsskillnader, sorption på sprickfyllnadsmaterial och sprickytor, diffusion in och ur bergfragment i sprickan, diffusion mellan ”strömlinjer”, diffusion in och ut ur bergmatrisen och andra stagnanta vatten, sorptionskinetik och inverkan av naturligt flöde i sprickan under försöket.

Många av dessa mekanismer påverkar hur spårämnehalten i återpumpat vatten varierar med tiden på ett snarlikt sätt. Det leder till att t ex diffusion i bergmatrisen och i andra stagnanta vatten inte kan särskiljas om endast ett spårämne använts. Spårämnen med olika diffusions- och sorptionsegenskaper kan i princip användas för att särskilja mekanismerna men de kommer att ha varit i kontakt med olika delar av sprickan; det sorberande spårämnet kommer att ha varit i kontakt med sprickytan närmast injiceringshålet medan det icke sorberande kommer att ha nått längre ut. Diffusion och sorption i små partiklar och bergmatris kan påverka hur mycket spårämne som återfås i tillbakapumpat vatten på ett snarlikt sätt, vilket gör det svårt att utskilja om det var partiklar eller bergmatris som påverkade resultatet.

Dispersion förorsakad av diffusion mellan ”strömlinjer”, s k Taylordispersion kan vara mycket olika i kanaler med olika form. Denna typ av dispersion kan också ge upphov till snarlika effekter som matrisdiffusion. Dispersionskoefficienter utvärderade ur ett SWIW försök kan ha liten relation till vad som skulle erhållas vid transport mellan punkt A och B i samma spricka under andra förhållanden. Detta beror dels på att olika mekanismer kan ha orsakat dispersionen, dels på att tidsskalorna kan vara mycket olika.

SWIW försök görs ofta under relativt kort tid för att undvika att spårämnet förs iväg av det naturliga vattenflödet. Det innebär att andra mekanismer kan påverka spårämnets uppförande än de mekanismer som kommer att ha störst inverkan för tider av intresse för en säkerhetsanalys för radionuklidtransport.

Eftersom många olika mekanismer kan ha snarlik inverkan på spårämnena måste parametervärden som bestämts med SWIW försök användas med yttersta försiktighet och eftertanke.

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1 Introduction

1.1 Background

The long term safety of a final repository for nuclear waste in deep lying crystalline rocks may be influenced by the capability of the fractured rock to retard any potentially escaping nuclides. One of the main retardation mechanisms is the uptake of the nuclides into the porous rock matrix and the sorption on the internal surfaces in the matrix. It has been found that the diffusion and sorption properties of the rock as well as the contact surface between the flowing water and the rock, the so called Flow Wetted Surface, FWS, are essential for assessing the retardation. It is of considerable interest to be able to verify that these retardation mechanisms are active in the deep lying rocks. Tracer tests in situ are used for this purpose. The main thrust of this report will be on discussing what information actually can be extracted from in situ tracer tests and what other mechanisms could possibly disturb the interpretation. It is also highly important to distinguish mechanisms that will be active over time scales of thousands to hundreds of thousands of years from those mechanisms that may be active in short time tests but will not have much effect over longer times. The discussion in the following will therefore point to a number of mechanisms and relate them to the time scales where they may be of importance.

Models of solute transport in fractured rocks are used to simulate contaminant transport in the complex three dimensional fracture networks that constitute the flowpaths. Most solutes interact in various ways with the fracture surfaces and with the underlying rock. Laboratory measurements are commonly used to study the different interaction mechanisms and processes. However, it is necessary to verify and validate the models by solute migration experiments in real fractures and networks of fractures in their natural environment i.e. by in situ experiments. Numerous experiments have been performed by different organisations. Very often between hole tests are used where tracers are injected in one borehole and their arrival monitored in another borehole. From the breakthrough curves information can be extracted on at least some of the solute-rock interaction mechanisms. Such between-hole experiments are very costly and difficult to perform in deep lying rocks from surface boreholes, which need to be accurately positioned. Practically nothing is known of the fracture properties along the flowpath where the test will be performed.

Some of the difficulties can be avoided by performing single well (borehole) experiments where a tracer solution is injected into an isolated fracture or in a network of fractures and later pumped back. The idea is that the tracer recovery curve could be used to extract information on at least some of the interaction mechanisms.

This report discusses the flow and transport mechanisms and processes that influence solute transport in fractured rocks with an aim to assess what information could be extracted from Single-Well-Injection-Withdrawal, SWIW, tests, and what obstacles and difficulties should be considered when planning, performing and interpreting such tests.

Below is a short description of SWIW tests and what information has been attempted to extract from such tests. Thereafter follows a short summary description of flow and transport in fractured rock systems, of interaction mechanisms and of rock properties related to the present problem. This will give the background for the rest of the report.

1.2 Basic principles of a SWIW test and some uses

In a SWIW test performed in a borehole a fracture is isolated between two packers. A given volume of traced water is rapidly injected into the fracture. After a waiting period water is pumped back and the tracer concentration in the pumped water is recorded. Much more water

is pumped back than was injected because the tracer has reacted in various ways with the material and water “pockets” in the fracture and is only slowly released back to the pumped water. The concentration time curve is analysed with the aim to extract information on the different mechanisms that caused the recovery curve to be spread out in time. This is typically done by fitting the recovery curve to a model by adjusting the parameters of the model to obtain a best fit in some sense.

For such a fitting to be meaningful the model must account for all important mechanisms that influence the recovery curve. It will be found that many interaction mechanisms have a similar influence on the recovery curve, which implies that independent observations or assumptions are needed to separate the effects of the different mechanisms. Variants of the test can be conceived e.g. slow injection, no or a short waiting period and slow back pumping. In such a test tracer interaction mechanisms caused by slow flow must also be considered in addition to those active in a test where the waiting period essentially determines the interaction. Then even more parameters must be determined.

SWIW tests are made in fractures connected to the flowing network and the natural flow in the rock must be considered to avoid unacceptable loss of tracer solution due to the natural flow in the rock. Commonly days to weeks of waiting time is used.

SWIW tests have been used for a variety of purposes. Some recent investigations are mentioned here. Several investigations using SWIW tests were aimed at obtaining in situ reaction rates for microbially mediated reactions. /Haggerty et al. 1998/ used a simplified method of “push-pull” test data analysis for determining in situ reaction rate coefficients, /Snodgrass and Kitanides 1998/ described a method to infer in situ reaction rates from push-pull experiments. /Burbery et al. 2004/ found that the models used often do not account for some relevant and important processes and they discussed the limitations of SWIW for these purposes. /Istok et al. 2001/ used push-pull tests for in situ determination of subsurface microbial enzyme kinetics. /Istok et al. 2002/ used partitioning tracers in single-well “push-pull” for NAPL detection in the subsurface. /Kim et al. 2004/ used push-pull tests for assessing in situ aerobic co-metabolism. /Davis et al. 2002/ investigated the use of radon and a non-interacting tracer to detect and quantify presence of non-aqueous phase liquid, NAPL, contamination in the subsurface, utilizing the retardation effect on radon by the presence of the NAPL.

SWIW tests have been performed to study dispersion of non-sorbing and interaction of sorbing tracers with the rock fractures assuming single porosity systems, /Snodgrass and Kitanides 1998, Altman et al. 2002, Ghergut et al. 2005, Gustafsson et al. 2006, 2005, Novakowski et al. 1998, 2004, Schroth et al. 2001, Wacker and Berg 2004, Cassini et al. 2005/. In many of the cited papers methods of evaluating and parameterizing the flow and transport properties are discussed. Literature reviews on design and interpretation of SWIW tests are reported in /Nordqvist and Gustafsson 2002, 2004/.

Tests have also been performed and interpreted assuming interaction by diffusion and sorption in a porous matrix of fractured rocks /McKenna et al. 2001, Haggerty et al. 2000, 2001/. /Meigs and Beauheim 2001/ discuss various processes and mechanisms that make it difficult to unequivocally determine transport parameters because they give rise to similar behaviour of the recovery curves.

2 Aims, approach and scope

This report sets out to survey the flow and transport mechanisms and process that influence solute transport in fractured rocks with an aim to assess what information could be extracted from Single-Well-Injection-Withdrawal, SWIW, tests and what obstacles and difficulties should be considered when planning, performing and interpreting such tests.

The mechanisms deemed to be most important are described and scoping calculations are used to explore when they can noticeably influence the solute rock interaction. Simple conceptual and mathematical models are used to assess which parameters and parameter groups contained in the models could potentially be determined by SWIW tests. It is also attempted to assess which mechanisms and their parameter values are likely to be difficult to determine. Furthermore it is attempted to point out information that can not be derived from such tests without resorting to various assumptions, which may be difficult to substantiate by independent measurements or observations.

A specific aim is to assess whether SWIW tests performed over a period of days, weeks or months can give quantitative information on mechanisms that will be of importance for simulating radionuclide transport over safety assessment timescales for repositories for nuclear waste, meaning times longer than thousands to ten thousands of years or even longer.

Central data needed for long term prediction include rock diffusion and sorption properties such as matrix diffusion coefficients and sorption distribution coefficients as well as the magnitude of the Flow Wetted Surface, FWS, over which interaction between solutes in the water and the rock take place. Purely hydrodynamic dispersion effects are deemed to be of secondary importance for the long term but are important to understand when evaluating tests.

3 Flow and transport in fractured rocks

Some recent well referenced papers on flow and solute transport mechanisms in fractured rocks summarize much of what is known today and also point to areas where there is more to learn /Bodin and Marsily 2003ab/. Another valuable source is the paper by /Neuman 2005/ where trends, prospects and challenges in quantifying flow and transport through fractured rocks are discussed. Below is a short summary of the processes and mechanisms that are deemed to be of special importance for the aims and scope of this report.

For the subsequent discussion a simplified conceptualisation of the fracture system in fractured crystalline rocks is given below. Much more detailed descriptions of the internals of fractures fracture zones can be found in e.g. /Mazurek et al. 2003/.

Fractures in crystalline rocks range in size from micro fractures between and in the crystals that make up the rock matrix to major fracture zones that can extend for several km. Fractures with sizes up to tens of meters or even up to 100 m can be made up of individual fractures or an echelon of individual fractures. These are often called background fractures. They can be oriented seemingly randomly in some rocks or in several dominating directions in other rocks. These fractures are sometimes mapped individually but are treated stochastically in fracture network models.

The surfaces of the background fractures are in contact with each other in some places but can also have open parts that are connected and allow water to flow through the fracture. Most of the flowing water is often seen to flow in a minor part of the fracture plane. This is called channelling. Channelling is deemed to mostly be due to the water seeking out the easiest paths under the hydraulic gradient that exists at any time. However, channelling is also known to exist in paths that have been generated by long time chemical weathering of a flow path or by precipitation of secondary mineral closing off section of the fracture.

Figures 3-1 and 3-2 show some channels observed on the walls in tunnels. Water with dissolved ferrous iron emerges from some parts of fractures and the iron is oxidised, mediated by micro-organisms, when it comes in contact with air in the tunnel. This leaves “curtains” of the iron precipitate mixed with micro organisms. Fracture intersections have been observed to form conduits for water. Flow in fracture intersections has also been observed in large scale flow experiment in Stripa underground laboratory /Abelin et al. 1991/. Channelling has been observed in many experiments and observations in fractured crystalline rocks /Abelin et al. 1983, 1991, 1994, Neretnieks 1988, Birgersson et al. 1993/. /Moreno and Neretnieks 1989/ discussed the impact of channelling in fracture zones on the transport of radionuclides. /Moreno et al. 1990/ discussed some anomalous features arising from fracture aperture variability. /Rasmuson and Neretnieks 1986/ showed that narrow channels can strongly influence the retardation effects due to matrix diffusion. /Grenier et al. 1998/ made numerical simulation of the influence of heterogeneities on the transport of non-sorbing solutes. /Tsang and Neretnieks 1998/ reviewed observations on channelling in heterogeneous fractured rocks. They found that this is a common phenomenon and may strongly influence tracer migration. /Neretnieks 2002a/ analysed some tracer experiments using a channelling model finding that uptake into stagnant water in the fracture plane may have a noticeable effect on interpretations of tracers experiments.

Larger fractures or fracture zones are often made up of a number of smaller fractures aligned in essentially a 2-dimensional feature with a thickness that can vary from a few cm to many meters or tens of meters. The zones contain large and small rock fragments, chemically altered rock and clay formed from weathered rock. The hydraulic conductivity within the fracture zones can vary considerably also permitting preferential flow paths to develop.



Figure 3-1. Channelling seen in the Bolmen tunnel as precipitates of iron oxides.



Figure 3-2. Channelling in the Poços de Caldas uranium mine seen as precipitates of iron oxides in the rock matrix into which oxygen has diffused and reacted with pyrite. White lines have been drawn around the oxidized red rock. The grey rock has not been invaded by oxidizing waters.

Figure 3-3 shows a stylised example of flowpaths and channels in a block of rock containing background fractures as well as a fracture zone.

This report will mainly discuss flow and transport properties in background fractures and minor fracture zones with transmissivities typically less than 10^{-6} m²/s. It is the flow and solute transport properties of the hydraulically connected fractures and the “channels” in this network that will be studied in this report.

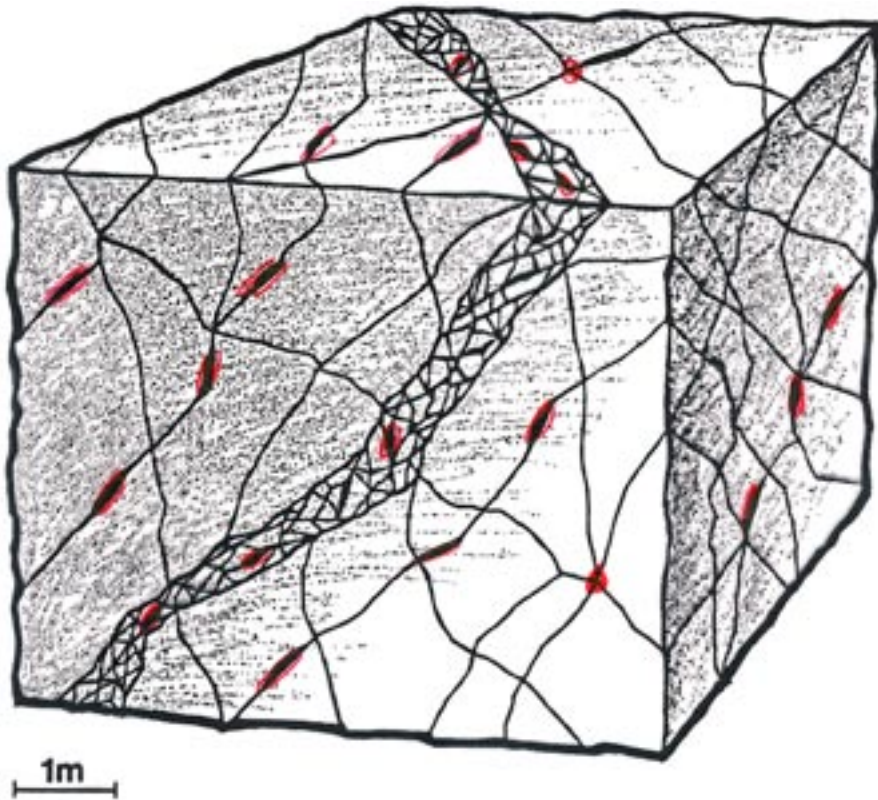


Figure 3-3. Flow takes place in background fractures as well as a fracture zone. Channelling is indicated by the red spots.

Water that flows in the network of channels will have a residence time distribution governed by the volume of the conductive network and the flow rate of the water. The differences in flowrate in different flowpaths will give rise to a Residence Time Distribution, RTD, of the water. Solutes carried by the water will have a RTD that is also influenced by different mechanisms including those caused by interaction with the rock surfaces. Some solute interaction processes with the rock can dramatically influence the RTD of the solutes. This is discussed in the next section.

4 Interactions of solutes with rock

Solutes carried by the water may interact with the walls of the fractures by physical and by chemical mechanisms. The rock matrix is porous and has a connected porosity extending at least tens of centimetres, possibly extending throughout the whole rock. The rock nearest the fracture surface is sometimes altered and can have a higher porosity and different chemical properties /Masurek 2003/. Many fractures have infill and coatings of secondary minerals which can have a different porosity and different chemical properties than the bulk rock.

There will be less or practically no flow in the coatings, secondary mineralization or the rock matrix compared to the flow in the fracture. However, solutes will access the pores by molecular diffusion. The penetration depth will be small for short contact times and larger for long.

Some sorbing solutes will reversibly attach to the outer as well as inner surfaces and be retarded in their migration. A sorbing species can have different sorption properties on the different minerals. There will thus be several mechanisms that influence the RTD of tracers. Different tracers will be influenced in different ways. The sorbing tracers will be retarded compared to the water velocity and during a given time a nonsorbing tracer will move a longer distance than a sorbing and the two tracers will therefore contact different parts of the fracture.

4.1 Interaction mechanisms between rock and dissolved ionic species

The main chemical interaction mechanisms considered for long term migration of radionuclides in fractured rock is adsorption and ion-exchange. This takes place on the mineral surfaces of the rock. Precipitation in various forms could contribute in addition. However, this is rarely accounted for and will not be discussed further. The short section that follows gives the background to why linear reversible sorption is commonly used in the models.

The main minerals of the rock matrix of granitic rocks are alumino-silicates, iron and other oxides and some alteration products such as clays. Table 4-1 shows examples of some common minerals found in granitic rock

Table 4-1. Examples of some common minerals found in granitic rock.

Rock forming minerals – Alumino-silicates (very low solubilities)	
Quarz	SiO ₂
K-Feldspar	KAlSi ₃ O ₄
Biotite	KMgFe ₂ AlSi ₃ O ₁₀ (OH) ₂
Plagioclase	Na _{0.62} Ca _{0.38} Al _{1.38} Si _{2.62} O ₈
Secondary (reactive) minerals	
Carbonates	CaCO ₃ (Secondary mineral in fractures)
Sulphide minerals	FeS ₂
Iron oxides	Fe ₃ O ₄ , Fe(OOH)
Clays (mostly in fractures formed by weathering)	
Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄
Montmorillonite	Al ₂ Si ₄ O ₁₀ (OH) ₂
Gibbsite	Al(OH) ₃

Some of these minerals have a permanent negative charge due to isomorphous substitution, which attracts positive ions in the water. Such charged minerals will have a capacity for cation exchange. In addition some oxide surfaces can be protonated and de-protonated depending on the local pH. This implies that the surface S = OH groups can become positively or negatively charged as S = OH²⁺ or S = O⁻ groups and attract oppositely charged ions. The S = OH groups also can bind cations by what is called surface complexation to form for example an americium complex S = OAm²⁺ with Am³⁺ ions in water. In addition precipitation and co-precipitation can occur when the concentrations of a species comes near its solubility. The sorption affinity of a specific cationic tracer (radionuclide) can be very different on the different minerals. The sorption of a specific cation will also depend on the presence and concentration of other ions, which compete for the sorption sites. The sorption takes place on the surfaces in and on the mineral grains. Therefore the magnitude of the accessible surface of the minerals in the fracture will influence the sorption capacity of a given area or volume of a fracture. Later the matter of accessible surface within porous particles and intact rock matrix will be discussed in the context of time dependence of solute interaction.

The above serves to illustrate that the sorption on a fracture surface and any infill in the fracture will be strongly influenced by the minerals present in the fracture and on the accessibility of sorbing surfaces. Provided that the fracture minerals in and on the fracture surface is reasonably constant in a flowpath some average sorption capacity and affinity can be defined.

4.1.1 The K_d concept and reversible sorption

Ion exchange and surface complexation are usually rapid processes once the ions have come near to the surface of the mineral grain. The processes are also reversible, which means that the ions can detach from the surface and enter the water as free ions again. Attachment and detachment is constantly occurring. Equilibrium may be attained when the two rates are equal. The ratio of the concentration of the species on the solid to that in the water is called distribution coefficient and is denoted K_d . Often it is given in units as shown in Equation (1)

$$K_d = \frac{c_s}{c_w} = \frac{\text{mass of ion} / \text{g solid}}{\text{mass of ion} / \text{l water}} \quad (1)$$

When the concentration in the water is very low i.e. trace concentration, K_d is not much influenced by the concentration and is often taken to be constant. This is usually deemed to be an acceptable approximation for most nuclides. It is inherently assumed that all the sites on the surfaces of a mass of the mineral are accessible and have been accessed in the experiments used to determine the distribution coefficient. Such experiments are commonly made on crushed rock and with contact times ranging from hours to months, sometimes more.

The assumption of linear reversible equilibrium and K_d concept is very useful because the mathematical treatment of the models(s) is much simpler than when nonlinear effects and reaction kinetics must be accounted for. In this report we assume that the K_d concept is applicable. /Crawford et al. 2006/ have recently discussed K_d measurements and compiled data for crystalline rocks.

4.1.2 Reaction kinetic effects

Even though the local sorption-desorption on a sorption site may be quick and reversible there sometimes may be changes of the surfaces due to e.g. slow weathering. It has been noted in laboratory experiments that equilibration with crushed rock can be a slow process taking weeks to months or more. If such effects are suspected in an in situ experiment this will influence the recovery curve of a SWIW test and may not be easy to distinguish from other effects /Haggerty et al. 1998/.

4.2 Diffusion kinetic effects

4.2.1 Diffusion in rock matrix

/Neretnieks 1980/ suggested that matrix diffusion could have a dominating impact on radionuclide transport in fractured rocks, especially for time scales of interest for safety analysis for nuclear waste repositories. It has been found to be the most important retardation mechanism for nuclides migrating in fractured rock formations /Moreno et al. 1993, 1997, Zuber and Motyka 1994, Smith et al. 2001, Reimus et al. 2003, Retrock 2005/. For that reason this report lays much emphasis on matrix diffusion and how it can be observed and quantified in SWIW tests.

Observed slow kinetics and dispersion in tracer tests in fractured rocks are often due to slow diffusion in larger particles where the tracer has to access the interior sites of larger particles and intact rock itself. This is one of the most important kinetic effects. The rock matrix, altered rock, secondary mineral coatings on fracture surfaces and infill particles are more or less porous but the flow resistance in the micropore network is so large that flow is negligible. A solute that comes in contact with rock and rock fragments will move by molecular diffusion into the connected pore network of the material to reach the inner sorption sites. The inner sorption sites have vastly larger surface than that of the flowing fracture. The longer distance the solute has to migrate the longer it will take to equilibrate the interior of the particle. In large particles the solute may not have time to penetrate the entire sample or rock matrix during the time of a test. During the back pumping stage in a SWIW test the recovery of the tracer must be expected to be only partial. This, in theory, could be used to assess the diffusion properties of the flowpath.

Sorbing tracers will be retarded as they diffuse into the material because they have to supply the sorption sites underway and equilibrate these before the tracer can move deeper into the sample. The penetration depth as it evolves with time can be illustrated by the simple expression in Equation (4). If a rock fracture consisting of only the rock matrix is exposed to a solute and the concentration at the surface is kept constant the tracer slowly diffuses into the rock.

Concentration profiles in a rock matrix with constant properties can be determined by solving the instationary diffusion equation, Fick's second law. The solution for unidirectional diffusion such as into the rock from the surface is given by Equation (2), for a material exposed to a constant concentration at the surface from time zero /Bird et al. 2002, p 621/.

$$\frac{c}{c_o} = \text{Erfc} \left(\frac{x}{2 \sqrt{\frac{D_p}{R_p} t}} \right) \quad (2)$$

D_p is the pore diffusion coefficient, which is smaller than that in unconfined water because of tortuosity and constrictivity effects. It can be measured or approximately estimated from the porosity of the material. The rock matrix diffusivity can also be estimated from electrical conductivity measurements performed in situ in boreholes even down to large depths /Löfgren and Neretnieks 2003/.

R_p is the retardation factor for the solute caused by the reversible sorption described earlier, Equation (3). ρ is the density of the rock material and ε_p is its porosity. These entities are measured in the laboratory

$$R_p = 1 + \frac{K_d \rho (1 - \varepsilon_p)}{\varepsilon_p} \quad (3)$$

The leading edge of the tracer where the concentration is arbitrarily set to 1% of that at the surface, after time t is at a location $\eta_{0.01}$. The rate of movement slows down with time. The distance is proportional to the square root of time /Bird et al. 2002, p 621/.

$$\eta_{0.01} = 4 \sqrt{\frac{D_p}{R_p} t} \quad (4)$$

Figure 4-1 illustrates penetrations depth into a porous, somewhat altered, rock such as can be found at the fracture surface. $D_p = 10^{-10} \text{ m}^2/\text{s}$ and porosity $\varepsilon_p = 1\%$, sorption coefficients $K_d\rho = 0, 0.1$ and 1 , (Retardation factors $1, 11$ and 101).

The left figure shows the penetration depth for times typical for a SWIW test and the right hand figure for the low end of typical for SA times.

It is seen that in a SWIW experiment the tracer would access up to a few tens of mm at most whereas for Safety analysis, SA, times the properties of the rock on the order of metres or more would influence the tracer migration. A tracer test performed over a short time may thus not give results that are representative for much longer times if there is reason to believe that the properties of the rock near the fracture surface differs from that further in. Another point to consider is that if there is infill consisting of particles in the flowpath these may add considerably to the sorption during the test but will contribute little during SA times when they have saturated early on already. This was one of the main conclusions also of /Haggerty et al. 2001/.

Another way to explore how far from the fracture surface a tracer penetrates and how this is influenced by an increased porosity near the fracture surface is presented below for illustrative purposes

4.2.2 Penetration into small particles in the flowpath

Similarly one can estimate the time for a solute to penetrate and equilibrate with a porous particle in the fracture. Particle sizes can vary from extremely small clay particles up to rather large blocks in fracture zones. In the type of fractures considered in this report for SWIW tests the fracture apertures are deemed to be smaller than a few centimetres for the more transmissive fractures and down to less than 0.1 mm for the less transmissive fractures, The larger fractures are typically filled with rock fragments, small particles and clay whereas the smaller fractures consist of two more or less closely fitting surfaces.

Particle sizes and the porosity of the infill will influence the transmissivity of a fracture with infill. A low porosity and small particle size will exert a high resistance to flow and will have a low local transmissivity. Water will tend to flow in regions with less such infill. The pore space of porous infill with low transmissivity will act similarly to a porous rock or coating where solutes will diffuse in the nearly stagnant water in the infill more rapidly than they are transported by flow. There may thus be regions with practically stagnant water in the fracture plane between flowing paths into which regions solutes will gain access by diffusion. The infill may also sorb the sorbing tracers.

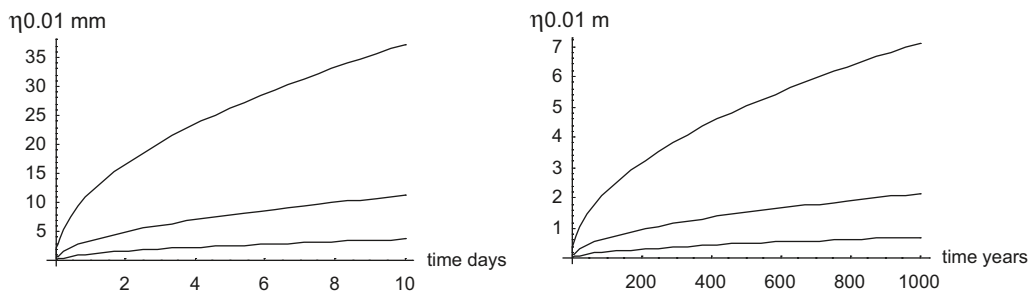


Figure 4-1. Penetration depth into a porous matrix as a function of time for retardation factors $1, 11$ and 101 , curves from top down.

When the penetration depth $\eta_{0.01}$ approaches the half dimension of the particle Equation (4) is no longer valid. Then even the mid parts of the particle become equilibrated. Then another illustrative entity can be useful. One can estimate the time it takes for the porewater in a particle of a given size to become nearly equilibrated with the water outside from diffusion theory. We call this the characteristic time for equilibration t_c . For a spherical particle the average concentration has reached 92% (arbitrarily chosen to get a simple constant in the equation) of the equilibrium concentration at a time /Carslaw and Jaeger 1959, p 102/.

$$t_c = 0.05 \frac{R_p}{D_p} d_p^2 \quad (5)$$

For a slab, d_p thick, the constant 0.05 is 0.2 instead.

Figure 4-2 shows the characteristic time t_c for $D_p = 10^{-10}$ m²/s and $R = 1, 11$ and 101 as a function of particle diameter. It is seen that particles smaller than one mm in diameter will be equilibrated after less than a day for the values used for diffusivity and retardation factor.

This implies that small particles can accommodate noticeable amounts of a sorbing tracer if exposed to it. Furthermore, very small particles will be quickly equilibrated and deplete the injected water rapidly if there is much infill of small particles in the part of the fracture injected with the traced water. Larger particles also can take up the tracer but will not be equilibrated during a day or so. They will, however, be in a transient state of uptake similar to the rock matrix or thick coating.

4.3 Possible scale effects of matrix diffusion

Recently several papers have been published regarding interpretations of tracer tests by models including the effects of matrix diffusion where this seemingly has been found to increase with tracer travel distance. /Becker and Shapiro 2000/ could obtain similar fits based on models with advection only without accounting for diffusion effects. /Shapiro 2001/ concluded from kilometre scale observations that matrix diffusivities must be orders of magnitude larger than that in free water to fit a transport model that included matrix diffusion effects and concluded that this must be caused by major flow heterogeneities and not by matrix diffusion. /Liu et al. 2007/ summarize a large number of papers that discuss these effects. In their paper they also investigated a number of potential mechanisms that could cause such increase. By simulations they show that hydraulic heterogeneities in the form of local flowpaths can give rise to the seeming increase in diffusion with scale. They also suggest that there may be further mechanisms, as yet not explored, that also could cause such effect. These observations suggest that solute interaction effects similar to those caused by diffusion into matrix pore water can be caused by other mechanisms, which may lead to difficulties to unequivocally determine diffusion parameters from tracer tests.

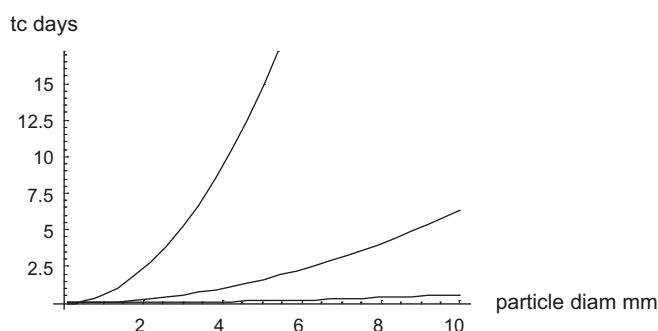


Figure 4-2. Characteristic time for saturation of particles as a function of particle diameter for $R_p = 1, 11$ and 101 , curves from bottom and up.

4.4 Other diffusion effects

Stagnant or nearly stagnant water is also found in regions with infill in the fractures and fracture zones and in regions adjacent to the flowing channels. Solutes in the flowing water will diffuse in and out of such regions. The effects on tracer transport with the flowing water will be very similar to those caused by matrix diffusion and cannot readily be distinguished from the solute concentration time curves. However, the use of tracers with different diffusion and sorption properties can be used to separate such effects. This is also possible for other diffusion effects discussed below, at least in principle.

In this section we consider the effects molecular diffusion has on spreading the solute between and across “streamlines” in a fracture. First, for illustrative purposes, a case is considered where there is no diffusion across streamlines. Thereafter the effects of molecular diffusion are considered and it is shown that this can increase the spreading of a pulse in some circumstances and to narrow the pulse in other.

4.4.1 Parabolic velocity profile in a slit for laminar flow

Water that flows in a slit moves faster in the middle than near the walls where it has zero velocity. The velocity profile is parabolic /Bird et al. 2002, p 63/. Equation (6) shows the velocity distribution in a channel. \bar{v} is the mean velocity and b is the half aperture of the fracture. z in this case is the distance from the middle of the slit.

$$v(z) = \bar{v} \frac{3}{2} \left(1 - \left(\frac{z}{b}\right)^2\right) \quad (6)$$

This is shown in Figure 4-3 below.

The velocity in the centre of the channel is 1.5 larger than the mean velocity. This has implications on how a tracer solution spreads out when it is injected into a fracture displacing the water without tracer. The water in the centre of the fracture will carry the tracer a given distance 50% faster than the average flow. The first arrival will come earlier than the mean of the tracer step input. The tracer concentration will gradually increase and the tracer concentration will build up and approach that of the injected traced water. The breakthrough curve for such a case is shown in Figure 4-4 for a tracer injection concentration equal to 1. The time scale is normalized to time for first arrival.

Figure 4-5 shows the concentration profile along the flowpath. The tip of the velocity profile has travelled a normalized distance equal to 1. The tracer is spread out over the whole distance generating a considerable dispersion.

This type of dispersion would influence the injected tracer solution as well as the recovery curve. This phenomenon could be expected to occur for short injection or withdrawal times and in fractures with large apertures when diffusion between streamlines has not had time to even

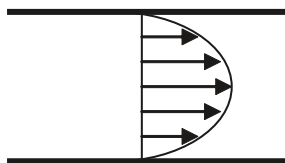


Figure 4-3. Velocity profile for laminar flow in a slit.

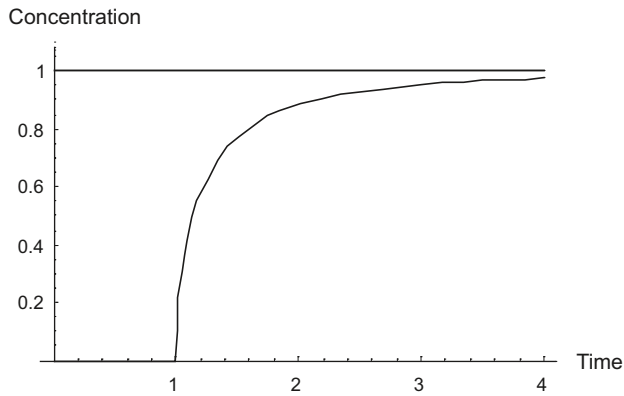


Figure 4-4. Breakthrough curve at a given distance as a function of time.

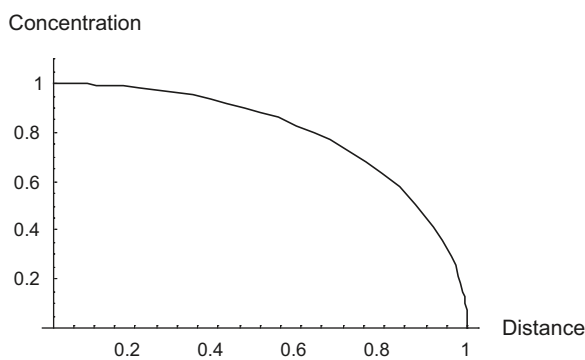


Figure 4-5. Concentration averaged over the aperture as a function of relative distance from entry.

out the concentration across the fracture. Long times or small apertures will allow diffusion to even out the concentration across the aperture. The characteristic time for this can be estimated from Equation (7) as described earlier for particles

$$t_c = 0.2 \frac{1}{D_w} d_p^2 \quad (7)$$

Figure 4-6 shows the characteristic time for a channel and a solute with diffusivity in water of $10^{-9} \text{ m}^2/\text{s}$ as a function of the aperture.

Figure 4-6 suggests that for large apertures a short injection time may not allow the tracer to equilibrate over the aperture and may lead to a tracer distribution along the injection flowpath with considerable dispersion as shown in Figure 4-4 above. In narrow fractures the tracer spreads over the aperture and the front between the displaced water and the traced water becomes narrower. This is called Taylor dispersion and is treated below.

4.4.2 Velocity profile in a tapered slit

In a flowpath in a fracture with variable aperture there may be a velocity distribution in the plane of the fracture in addition to that across the fracture aperture as treated above. Channels in variable aperture fractures will most probably have this kind of velocity distribution. We explore an idealised case below.

Figure 4-7 shows an idealized tapered slit with a maximum aperture of $2b_{max}$ and width W .

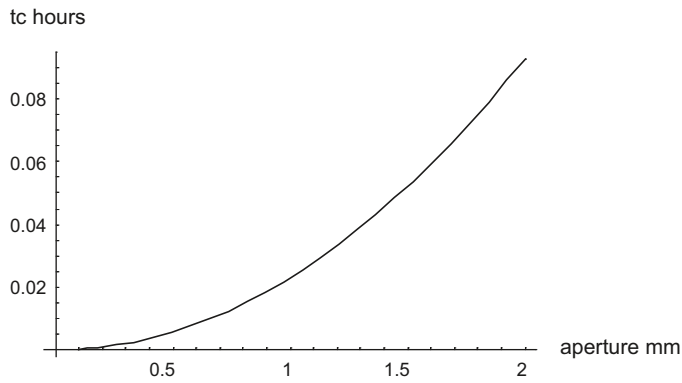


Figure 4-6. Characteristic time to equilibrate the tracer across a fracture aperture by molecular diffusion.

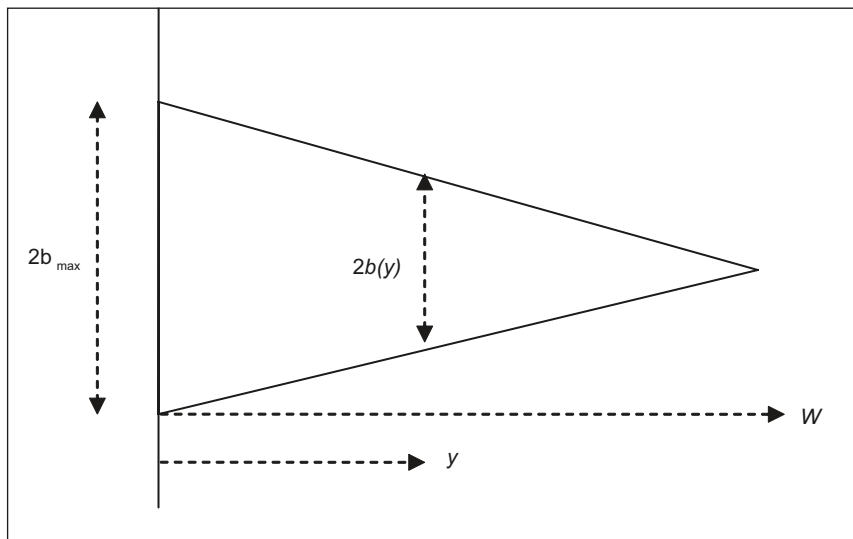


Figure 4-7. Tapered slit formed channel. Water flows in the slit perpendicular to the figure.

The mean velocity $\bar{v}(y)$ at every location y is practically proportional to $b(y)^2$. We neglect the influence of the gradient in the y direction due to the tapering. The flowrate in a vertical slice is proportional to $b(y)^3$ similarly to that discussed previously for the velocity distribution over the aperture. If $W \gg b$ we can neglect the tapering off of the cross sectional area for diffusion and treat the case as for the velocity distribution across the channel.

The characteristic time for diffusion is then approximated by Equation (7) but with d_p exchanged for $2W$.

$$t_c = 0.8 \frac{1}{D_w} W^2 \quad (8)$$

For a case with $W = 0.05$ m $t_c = 23$ days. Thus for short injection times the parabolic velocity profile in the y direction will not be evened out by diffusion. This will also be shown under Taylor dispersion discussed below.

It may be mentioned here already that if an injected non interacting tracer pulse were to be immediately pumped back the pulse that is spread out in the injection step would again “un-spread” because the tracer in the adjacent “streamlines” will not have had much time to diffuse

between them. However, during the waiting period in a SWIW test, of say ten days in this example, diffusion between the streamlines would become noticeable. Furthermore, as will be discussed later the interaction of a tracer by diffusion in the porous rock matrix is very strongly influenced by the fracture aperture as a locally narrow aperture allows much larger depletion of the water in the fracture by uptake into the matrix.

4.4.3 Taylor dispersion

Molecular diffusion will even out the concentration profile across the flowpath /Bird et al. 2002, Logan 1999/. This effect may considerably decrease the spread of the tracer pulse in the flow direction. This will be illustrated by considering the spreading of a narrow pulse travelling with the stream.

The spread of a short (Dirac) pulse due to molecular diffusion in the flow direction can be obtained from Equation (9). This would apply for a flat velocity profile. The pulse is travelling with the velocity v in the x direction.

$$c = \frac{M}{2\sqrt{\pi D_w t}} e^{-\frac{(x-vt)^2}{4D_w t}} \quad (9)$$

This is illustrated in Figure 4-8 for $D_w = 10^{-9}$ m²/s after $t = 1$ hour and velocity 1 m/hour and $M = 1$ (arbitrary unit).

It is seen that for this case the “dispersion” due to molecular diffusion is very small when plug flow (flat velocity profile) is assumed. The pulse has spread to about one cm in the flow direction due to diffusion. This is, however much larger than the fracture apertures we consider. The velocity profile will constantly try to spread the pulse but the diffusion across the aperture will counteract this. The combined effects on dispersion are called Taylor dispersion. It has been quantified by solving the flow and diffusion equations, resulting in Equation (10). It has been confirmed by experiments /Bird et al. 2002/.

$$\frac{D_L}{D_w} = \left(1 + \frac{1}{52.5} \frac{b^2 v^2}{D_w^2}\right) \quad (10)$$

It should be noted that the notion of Taylor dispersion is valid for the case when the traced solution is collected and mixed at the “outlet” of the fracture. If there has not been time to even out the concentration between the streamlines the “dispersion” would not be seen if the fluid was rapidly pulled back as in a SWIW test. Compare the discussion around Figures 4-4 and 4-5.

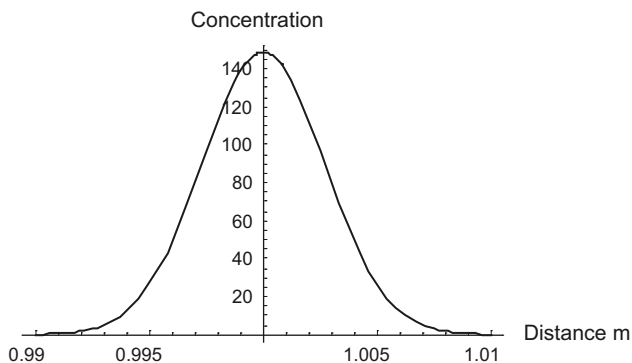


Figure 4-8. Spread of a Dirac pulse in the direction of flow due to molecular diffusion only. Velocity $v = 1$ m/hour, $D_w = 10^{-9}$ m²/s. Observation after 1 hour.

D_L can be used instead of D_w in Equation (9), for a Peclet number $Pe_x > 10$, at least for illustration purposes.

$$Pe_x = \frac{vx}{D_L} \quad (11)$$

Note however, that this is not strictly correct because the form of the pulse by pure diffusion and Taylor dispersion is different. It is seen that for very low velocities or apertures b the first term will dominate and the longitudinal dispersion coefficient approaches the molecular diffusion coefficient. For large apertures or velocities the second term will dominate. This is shown in Figure 4-9 where D_L/D_w is 133 for the narrow curve and 1,470 for the broad curve.

A more rapid injection will increase the dispersion due to Taylor effects (Figure 4-10). Also, if there is a tapered fracture discussed previously, the dispersion that would be observed if the water was collected and mixed at some point downstream would exhibit a considerable dispersion.

Equation (10) with $b = 2W = 0.1$ m and a velocity of 1 m/hour gives $D_L = 1.5 \cdot 10^7 \cdot D_w$. This is well outside the validity of the formula, Equation (9), as it would imply that the pulse has spread much wider than the length of the travel distance (1 m) in the example considered.

Should the same volume be injected over 5 days instead of 1 hour in the tapered fracture, succeeded by an immediate withdrawal there would be a much larger effect of diffusion across the “streamlines”. A very approximate but illustrative assessment of the resulting dispersion

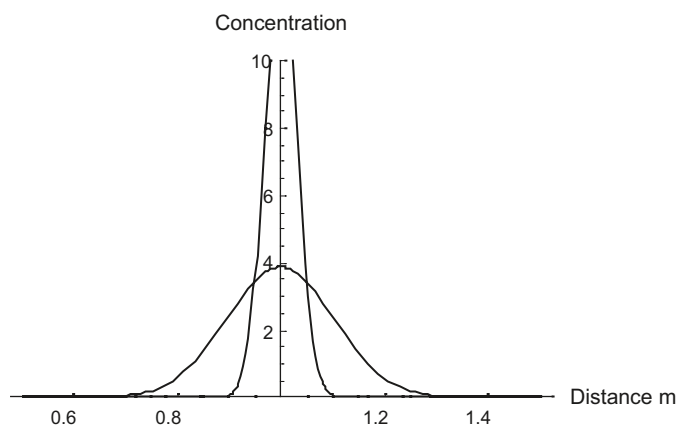


Figure 4-9. Spread of a Dirac pulse in the direction of flow due to Taylor dispersion. Velocity $v = 1$ m/hour; $t = 1$ hour; $D_w = 10^{-9}$ m²/s. High curve for aperture 0.3 mm, low curve for aperture 1 mm. Observation after 1 hour.

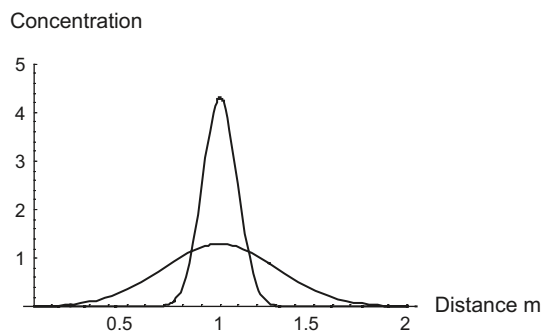


Figure 4-10. Spread of a Dirac pulse in the direction of flow due to Taylor dispersion. Velocity $v = 3$ m/hour; $t = 0.33$ hour; $D_w = 10^{-9}$ m²/s. High curve for aperture 0.3 mm, low curve for aperture 1 mm. Observation after 0.33 hour.

coefficient is $D_L = 1 \cdot 10^{-6} \text{ m}^2/\text{s}$ or 1,000 times larger than the diffusion coefficient in water. The Peclet number, $Pe_x = 2.3$ in this case.

A Pe_x between 1 and 100 is commonly found in tracer tests carried out between two wells. It is mentioned here because it suggests that one reason of pulse spreading in field experiments could be caused by the above effects and can be very much influenced by the way a tracer test is designed. The dispersion coefficient is not only a property of the channel or channel network but is very much influenced by the way the test is performed.

It is seen that the Taylor dispersion can cause considerable pulse spreading and must be considered when designing and evaluating SWIW tests. If extrapolations of results from short term tracer tests are to be made the underlying mechanisms must be well understood and quantified.

Taylor dispersion must also be considered when the water is pumped up from a deep borehole, where the test has been performed, to the ground surface for analysis. A wide tube and fast but still laminar flow can cause a considerable dispersion which can be difficult to distinguish from that caused by the test itself.

5 Flow patterns in fractures

5.1 Flow patterns

The water flowrate varies considerably between different parts of a fracture. Preferential flow paths develop in the fracture and often most of the flow takes place in a small fraction of the fracture. This is called channelling. In practice the actual flow path geometries cannot be a priori determined. The Flow Wetted Surface, FWS, i.e. the surface that a water “package” will contact is not known. Furthermore it is yet not possible to measure the aperture of the flowpath by non-destructive means.

5.1.1 Flow in a fracture with and without infill

In this section we explore what sizes fracture apertures can be expected in a SWIW experiment and what infill particles can do to fracture transmissivity. It will be shown later that this information is crucial for interpretation of SWIW tests.

The larger fractures are typically filled with rock fragments, small particles and clay. Particle sizes and the porosity of the infill will influence the transmissivity of a fracture with infill. A low porosity and a small particle size will exert a high resistance to flow and result in a low local transmissivity. Water will tend to flow in regions with less such infill. The pore space of porous infill with low transmissivity will act similarly to a porous rock or coating where solutes will diffuse in the nearly stagnant water in the infill more rapidly than they are transported by flow.

One can estimate the pressure drop in a fracture with infill by the Blake-Kozeny equation /Bird et al. 2002, p 190/. The transmissivity of an infilled fracture with aperture $2b$ is:

$$T = \frac{2b}{150} \frac{\varepsilon^3}{(1-\varepsilon)^2} \frac{d_p^2 \rho g}{\mu} \quad (12)$$

Figure 5-1 shows the transmissivity of a 1 mm slit filled with small particles with diameter d_p m for different porosities. The horizontal line shows the transmissivity of an open slit according to the cubic law. An open 0.1 mm slit will have 3 orders of magnitude lower transmissivity, i.e. just below 10^{-6} m²/s and the slit aperture will have to be as small as 0.01 mm for the transmissivity to be 10^{-9} m²/s. The transmissivity of the slits with infill on the other hand decrease in proportion to the aperture.

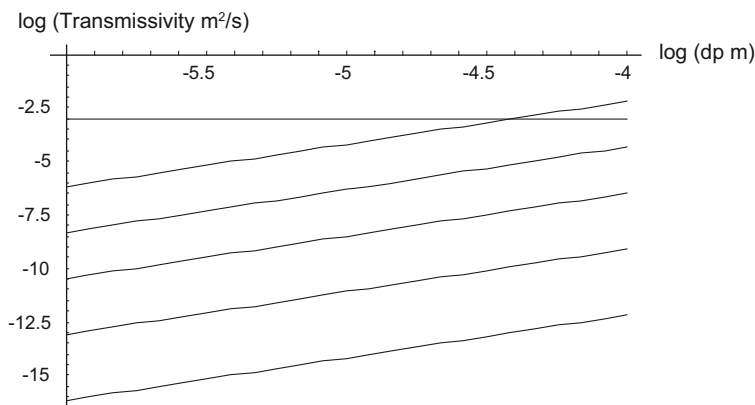


Figure 5-1. Infilled fracture transmissivity as a function of particle size and porosity of the infill. Curves from bottom and up are for porosities of 1%, 10%, 50%, 90% and 99%. The aperture $2b = 1$ mm. The horizontal line shows the transmissivity of an empty fracture according to the cubic law.

The cubic law is shown in Equation (13).

$$T = \frac{1}{12} \frac{\rho g}{\mu} (2b)^3 \quad (13)$$

It has been suggested that the hydraulic transmissivity of the fracture where the SWIW test is made to be used to assess the fracture aperture by using the cubic law. /Guimera and Carrera 2000/ analysed data from 90 tracer experiments and found some support for the use of the cubic law. However, the regression had a large variation and they concluded that it cannot be used for predictions. /Sawada et al. 2001/ used tracer and hydraulic data from fractured rock in granodiorite in the Kamaishi mine to correlate aperture to transmissivity. They found an empiric equation with a square root relation and with a spread of about one order of magnitude.

$$2b = \text{const} \sqrt{T} \quad (14)$$

All data fall within a range where the constant is between 1 and 10 when T is in m²/s and 2b is in mm. Transmissivity ranged from 2·10⁻⁸ to 3·10⁻⁷ m²/s and the aperture 2b between 0.2 and 3 mm. Both relations are plotted in Figure 5-2 using a constant 3.15 for the empiric relation.

Figure 5-2 shows the aperture of a fracture as a function of the transmissivity according the cubic law for water at around room temperature and the empirical relation. The cubic law under predicts the observed apertures by more than one order of magnitude in the range of interest for this report.

Developments are underway to better understand the relations between hydraulic and transport apertures. Recently e.g. /Konzuk and Kueper 2004/ using very detailed measurements, nearly a million points, of the aperture variation over a fracture surface obtained encouraging agreement when using geometric mean aperture and incorporating surface roughness factors. However, the uncertainties in aperture will be the cause of the largest uncertainties in evaluation of the matrix interaction parameters.

5.2 Effects of heterogeneities in the flow paths

/Becker and Shapiro 2000, Wörman and Dverstorp 2001, Xu and Wörman 2003, Lee et al. 2003/ and /Gwo et al. 2005/ studied the impact of various heterogeneities on tracer transport in fractured rocks. /Keller et al. 1999/ in a high resolution imaging of a fracture modelled flow

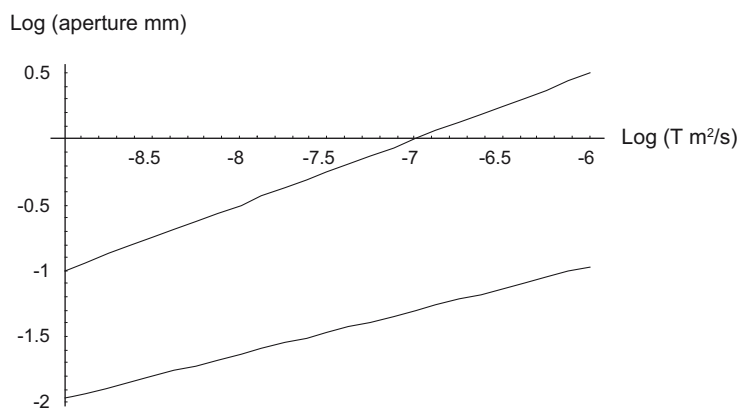


Figure 5-2. Aperture of a fracture as a function of the transmissivity according to the cubic law (lower curve) and the data from the Kamaishi mine (upper curve).

and transport and compared the results with experiments, confirming the strong impact of heterogeneities on the transport properties and that these can be modelled, provided detailed information is at hand.

In flowpaths in fractured zones the block size distribution and channelling effects due to heterogeneities in the hydraulic conductivity can be expected to have a strong influence on the interaction with the tracers. /Rasmuson 1985/ studied the impact on nuclide transport of the presence of particles/blocks of different sizes and shapes in the flowpaths. /Haggerty and Gorelick 1995, Haggerty et al. 2000, 2001, Fleming and Haggerty 2001, McKenna and Meigs 2001/ used the same concept they call multirate model to assess the impact of matrix diffusion effects. The multirate concept implies in essence that different particle sizes are assumed to exist in the flowpaths and that they need different times to be equilibrated. The multirate conceptualization was also found to be sound in an analysis of published tracer experiment /Haggerty et al. 2004/. /Gerke and van Genuchten 1996/ studied the impact of different particle and pore shapes and suggest a method by which complex and mixed types of structures can be modelled.

Quantitative information is never available for the actual flowpaths in in situ experiments and the influence of the heterogeneities on the interpretation of tracer tests must be assessed by other means.

5.3 Hydrodynamic dispersion

The term hydrodynamic dispersion is commonly referred to when discussing spreading of a tracer pulse as it progresses through a porous and/or fractured medium. A dispersion coefficient D_L has long been used in advection-dispersion models to parameterise it. In models it enters in the same way as molecular diffusion does. However the underlying mechanisms are quite different. Some of the reasons for pulse spreading have been discussed above. Taylor dispersion contributes as well as diffusion in and out of stagnant waters along the flowpath. The latter may include the rock matrix, particles in the flowpath, stagnant water in the fracture plane etc. An additional effect comes into play when tracer tests are performed by collecting traced water in a well. It is sometimes called “velocity” dispersion. It is caused by the traced water having moved to the pumping well by different paths with different residence times /Matheron and Marsily 1980, Neretnieks 1981, 1983, 2002a, Chesnut 1994/.

Velocity dispersion can have a strong impact on the breakthrough curves in multi-well and converging pumping tracer test. However, in SWIW tests where the traced water will be pumped back along the same flowpaths used in the injection phase that effect is not present. Dispersion in SWIW tests is then expected to be smaller than in in-between hole tests in the same rock. This could potentially allow a better identification of the other processes.

The lumped effects of all the underlying causes are summarised in the dispersion coefficient or when divided by the average velocity, $\alpha = D_L/v$, a dispersion length α is obtained to characterise the dispersive properties of the medium. It must be recognised that this practice, although commonly used, must be used with utmost caution. It can only be applied to the actual flowpath under the same flowrate conditions, which renders its use for predictive purposes to be very limited as it is a lumped parameter. It includes the effects of several independent mechanisms, which all scale differently with travel distance, water velocity, residence time and even depend on the tracer used. One example of the difficulties using this lumped parameter concept is that the apparent D_L and α are found to increase with observation distance /Gelhar et al. 1992/ and thus are not to be seen as a property of the medium. In the models, however, it is used as such and given a constant value when solving the equations.

It is desirable to separate the different effects causing pulse spreading and to model them separately if the results are to be used for predictive purposes.

6 Processes affecting a SWIW test in a fracture

The following discussion centres on processes in a single fracture. It is at first assumed that only the two fracture surfaces act as contacts between the rock and the traced water. The impact of the presence of splays, infill and larger rock particles in the fracture are discussed separately.

6.1 Injection of a volume of water in a fracture

We start by an idealized case and consider where a known volume of water is injected into a fracture with constant aperture. Then some non-ideal cases are considered where the fracture aperture varies and when channels develop.

6.1.1 Ideal case

Consider a case where a fracture with aperture $2b$ is injected with a traced water volume V_{inj} . The water spreads out radially to a distance r which is given by

$$r = \sqrt{\frac{V_{inj}}{\pi 2b}} \quad (15)$$

An aperture of 1 mm and 10 litres of water will give a radius of 1.78 m. A 0.1 mm aperture will give a radius of 5.65 m.

It is often practiced to inject another volume of water without tracer to force the traced water out from the vicinity of the borehole. If the non-traced water has the same volume the outer edge of the traced water will move a distance 1.41 times further out and the non-traced water will move to the above stated distances. It may be noted that the contact area between the traced water and the fracture surface is inversely proportional to the aperture. This will have a very strong influence on the surface over which solute transport and on which reactions can take place. The uncertainty in aperture will cause a considerable uncertainty when evaluating the reaction and transport mechanisms unless independent means to assess the aperture can be found.

6.1.2 Non-ideal case

In a fracture with variable aperture the water will seek out paths preferentially where the resistance to flow is small. Channels will develop. In practice the size and shape of the traced water volume residing in the fracture will not be known. Below we explore some idealized possible shapes of the traced water and discuss some potential effects this may have on the interaction of the tracer with the solids and other water volumes in the fracture. Figure 6-1 shows some idealized patterns. Non-traced water has been injected after the traced water.

The blob illustrates a case where Taylor dispersion has diluted the tracer in the front and back of injected water. The main interaction mechanisms for the blob are deemed to be by diffusion over the fracture surfaces into coating, alteration and rock matrix as well as into large and small particle infill material.

The narrow channel and the finger like water shape will in addition to the contact with the fracture surface have a large surface area in contact with stagnant water in the fracture and the infill residing there. Solutes may diffuse into these regions. The depletion of the tracer by

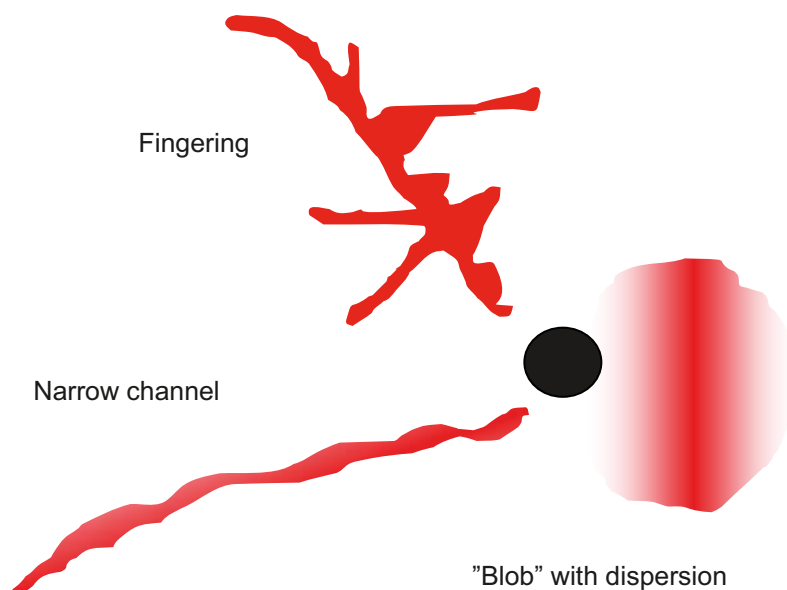


Figure 6-1. Illustration of some conceivable patterns of the injected traced water.

diffusion into the stagnant water could be large if the channel or fingers are narrow. In tube like channels this could dominate the depletion of the tracer in the water in the channel. Furthermore tracer that has entered the stagnant water in the fracture plane can diffuse into the rock via the contact surface in this part of the fracture. This has been found to contribute substantially to the depletion of solute from narrow channels /Neretnieks 2006/.

6.2 Recovery by back pumping

The dispersion observed during back pumping is caused by the mechanisms described previously. If there were no dispersion effects and no interaction between tracer and the material in the fracture, ideally, the injected tracer will be recovered exactly as it was injected. A pulse would be recovered exactly with the same shape as it was injected. A tracer that reversibly and rapidly sorbs and desorbs on the surfaces of the channel and on any small infill will also be recovered as injected. It may be noted that a sorbing tracer in this case will have travelled a much shorter distance than a non-sorbing tracer. It would have much shorter distance to travel back. This also implies that a sorbing tracer might contact a very different part of the fracture surface. Physical and chemical heterogeneities may then influence the difference in interaction between sorbing and non-sorbing tracers. When Taylor dispersion must be considered the recovery will also be influenced by this.

When there is diffusion over the fracture surface, into stagnant waters and in larger particles, the tracer that has entered these regions will not be instantaneously released into the back pumped water. The concentration in the back pumped water will then be less by an amount that is equal to that residing in the stagnant waters in the matrix, in the large particles and in the water in fracture. If withdrawal is rapid only little tracer will diffuse back into the water in the channel. If water withdrawal is slow more tracer will have time to be released into the back pumped water. Some tracer will never be recovered because it continues to diffuse further and further into the rock.

The form of tracer recovery curve will be influenced by at least the following mechanisms:

1. Taylor dispersion
 - a. Across aperture.
 - b. In “tapered” aperture in channel.
2. Diffusion in and out of
 - a. Particles in flowpath.
 - b. Rock adjacent to the flowpath including coating, alteration zone and rock matrix.
 - c. Stagnant water and further migration into the infill in water adjacent to the flowpath.
 - d. Diffusion from stagnant water into rock matrix.
3. Sorption/desorption reaction rate.

These mechanisms depend differently on the time that they are allowed to act. This implies that the rate of injection, the rate of withdrawal as well as the waiting period will influence the form of the recovery curve and the amount of tracer that is recovered.

Below we develop models to account for items 1 and 2. Item 3 is not included because there is no unequivocal data that can be used and because sorption reactions, once the solute is at the sorbing surface, usually are fast compared to diffusional processes. Much of the seemingly slow reaction rates can be caused by diffusional effects, which are modelled.

6.3 Drift of traced volume caused by natural flow

Under in situ conditions there may be flow due to the natural hydraulic gradient. This will tend to move the traced water out from the paths it was injected into. /Altman et al. 2002/ in a study of a single-porosity fracture found that heterogeneity and slow drift during the waiting phase can carry the fluid to areas where the flow is lower during back pumping. This may lead to dispersion and decrease in recovery.

7 Model and simplified versions

Because the main aim of the tests in this report is to validate in situ diffusion and sorption processes it would be advantageous to perform the tests in such a way that these processes exert a dominating effect on the recovery curve. Taylor dispersion during injection and withdrawal phase could be minimised by choosing a suitable injection velocity, see Equation (10). We therefore start by a model where it is assumed that the injection has not perturbed the form of the pulse. The conceptual model is illustrated in Figure 7-1.

The idealised model considers a case where traced water has been injected into a channel. The concentration is c_o at the beginning of the waiting period. We wish to determine the concentration in the water in the channel after time t_{wait} . In the idealised case this water and additional flushing water from the formation could be pumped back and its concentration monitored as a function of the pumped volume. If no dispersion effects are active the concentration in back-pumped water should be constant during the pumping period when the pulse is recovered. It is assumed in this idealised case that the back pumping is slow enough to minimise Taylor dispersion but fast enough to avoid back diffusion from matrix and stagnant water. Although it is obvious that such an ideal experiment cannot be performed in practice it will be shown that there are some interesting features that can be useful in designing and evaluating also real experiments.

The model includes the following solute exchange paths with the surrounding acting in parallel and in series. It is idealised in the sense that the channels in the fracture are given a constant width and aperture, neglecting the known variations. Nevertheless, the models are useful for studies of the interaction of tracers with the fracture material as they convey insights into when different mechanisms can be important. The model contains the solute transport mechanisms shown in Figure 7-1 that act upon the injected traced water.

Some illustrative cases with one or a few mechanisms or processes acting at a time will be discussed to bring out the impact of the individual processes. They are:

- Instantaneous reversible sorption with channel infill and channel surfaces. No diffusion effects.
- “a” plus diffusion from the channel into the rock matrix plus sorption in matrix and diffusion into stagnant water.
- “b” plus diffusion from the stagnant water in the fracture into the rock matrix and sorption there.

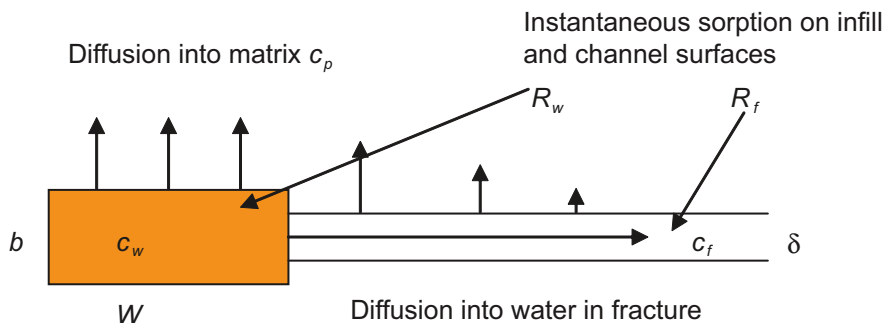


Figure 7-1. Solute from b channel (quarter shown) diffuses into matrix, into δ -fracture with stagnant water and from there into matrix. Channel has surface and infill with retardation coefficient R_w and δ -fracture also has surface and infill with retardation coefficient R_f .

The channel as well as the fracture with stagnant water have surfaces and can contain infill that interacts by rapid reversible linear sorption with the solute. Sorption in the matrix is modelled as linear and reversible as is sorption on fracture surfaces and infill in the flow path as well as in the stagnant water. The model is based on the assumption that the traced volume has been ideally injected in a channel of given width, $2W$, and aperture $2b$. This defines the contact surface per length of channel and the contact area between water in the channel and stagnant water in the fracture. The three cases, with increasing complexity, are explored below

7.1 Case a: Rapidly inject and immediately withdraw traced water.

A volume V_o of traced water is suddenly introduced into a fracture in a rock. The tracer equilibrates rapidly with the small infill particles and fracture surface and is retarded relative to the water by a factor R_w as shown in Equation (16). If the water has travelled a distance x in the channel the tracer will only have travelled x_s because of the retardation.

$$x_s = \frac{\varepsilon \cdot x}{(\varepsilon + (1-\varepsilon)K_d\rho)} = \frac{x}{R_w} \quad (16)$$

The sorbing tracer will have contacted a surface $W \cdot x_s$ which is less than a nonsorbing tracer has. A sorbing and a nonsorbing tracer thus will be in contact with a different part of the channel. The recovered pulse will be identical to the injected pulse.

It may be noted that if a chaser of pure water is injected after the traced water the sorbing and nonsorbing tracer may reside in entirely different parts of the fracture. This also has implications on how the tracers during a waiting period diffuse into the stagnant waters in matrix and fracture as the sorbing tracer has less contact surface over which to diffuse into the stagnant waters.

7.2 Case b: Inject tracer in fracture and monitor concentration in the water in the fracture

We now turn to case “b”. Again the traced water is injected rapidly. The rock matrix is porous and the tracer can diffuse into the matrix of the rock in the z-direction, so depleting the tracer in the water in the fracture. The traced water is present in a narrow channel and the solute may diffuse into stagnant water in the fracture in contact with the channel. Monitoring of the water in the fracture can only be done by withdrawing it so that one would choose a suitable waiting time and then rapidly withdraw the water.

The model is formulated in mathematical terms by making a mass balance of the tracer in the fracture from which tracer is lost by diffusion. It is assumed that the tracer concentration over the aperture b and the width W of the channel is rapidly equilibrated so that there is no concentration gradient across aperture and width of the channel. The validity of this simplification can be assessed by the analysis of the characteristic times involved.

The mathematical treatment is given in the appendix. The concentration in the pore water in the matrix at time t after injection is given by Equation (17). The concentration in the fracture water is obtained by setting $z = 0$. The notation is explained in the appendix and in the “notation” listing.

$$c_p(t, z) = \frac{c_o}{\sqrt{\pi}} \int_0^t \frac{e^{-\frac{\beta^2 z^2}{4(t-\tau)}}}{\sqrt{t-\tau}} \left(\frac{1}{\sqrt{\pi\tau}} - ae^{a^2\tau} \operatorname{Erfc}(a\sqrt{\tau}) \right) d\tau \quad (17)$$

where

$$\alpha = \frac{\delta}{R_w b} \frac{\varepsilon_f}{W} \sqrt{R_f D_f} + \frac{\varepsilon_p}{R_w b} \sqrt{R_p D_p} \quad (18)$$

and

$$\beta = \sqrt{\frac{R_p}{D_p}} \quad (19)$$

It is seen from Equation (18) that matrix diffusion and diffusion into stagnant water in the fracture cannot be distinguished by this experiment as they are additive and influence the results via the same parameter α for a given tracer.

Some calculated cases are presented and discussed below to illustrate the relative impact of matrix and stagnant water diffusion.

7.2.1 Diffusion into rock matrix only

For this case the parameter α only contains the second term of Equation (18).

$$\alpha = \frac{\varepsilon_p}{R_w b} \sqrt{R_p D_p} \quad (20)$$

Figure 7-2 shows the concentration of a non-sorbing tracer in the channel and in the pore water at distance 1 and 2 mm into the matrix for $D_p = 10^{-10} \text{ m}^2/\text{s}$, $\varepsilon_p = 0.01$ and a slit aperture $2b$ of 1 and 0.1 mm as a function of time. Figure 7-3 shows the results for a sorbing tracer in a 1mm aperture fracture. In these examples there is no infill in the fracture.

Figures 7-2 and 7-3 show the impact of the fracture aperture and retardation coefficient. In the 1 mm fracture the concentration has dropped by 20% after ten days whereas in a 0.1 mm fracture the concentration has dropped more than 3 times more. An inspection of the parameter α , Equation (17) shows that the same effect could be caused by an increase of the pore diffusion coefficient by a factor of 100. This points to a difficulty when trying to extract information on the matrix diffusion properties from a SWIW test when the aperture is not known by independent means. Similarly for a sorbing tracer the retardation factor could be different by a factor of 100 for a change in aperture by a factor 10.

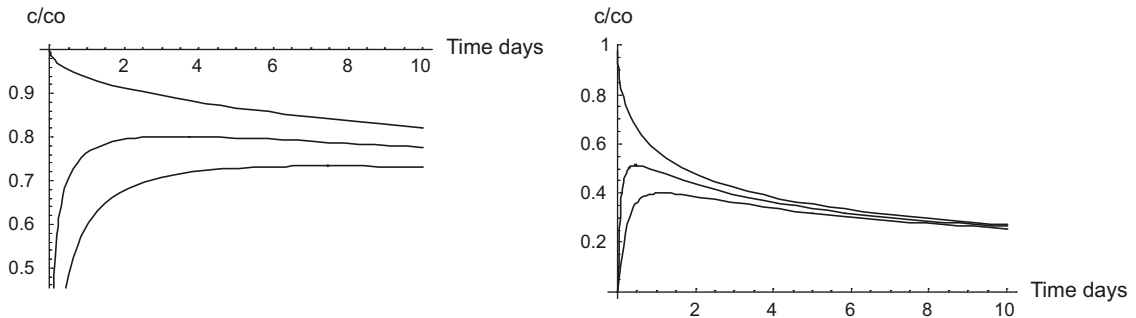


Figure 7-2. Concentration in water in fracture and 1 and 2 mm into the matrix pore water for non-sorbing species for 1 mm aperture (left) and 0.1 mm aperture (right) after waiting time t .

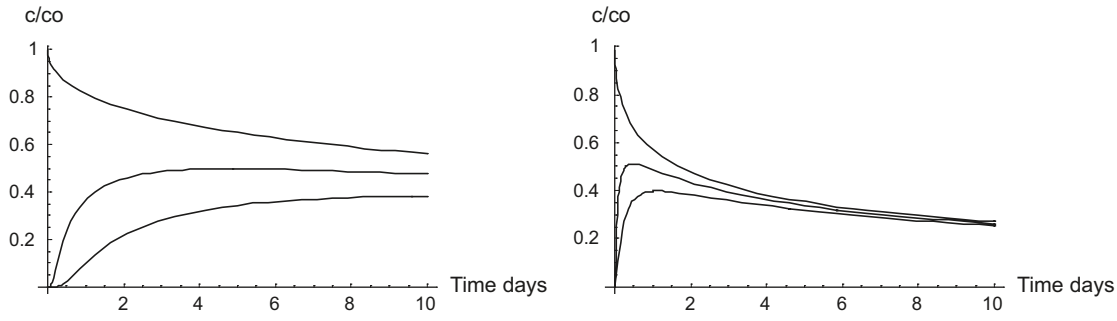


Figure 7-3. Concentration in water in fracture and 1 and 2 mm into the matrix pore water for sorbing species for 1mm aperture and $R_p = 11$ and 101 (left and right respectively) after waiting time t .

If the withdrawal rate is fast enough the concentration in the withdrawn water would be constant for a withdrawal volume V_o as before in case “a”. The length of the waiting period will have a strong influence on the concentration in the water.

7.2.2 Compare effect of matrix and stagnant water uptake

A direct comparison of the impact of the two mechanisms can be made by inspecting the relative magnitude of the two terms. Equation (18) is rewritten

$$\alpha R_w b = \delta \frac{\varepsilon_f}{W} \sqrt{R_f D_f} + \varepsilon_p \sqrt{R_p D_p} \quad (21)$$

It is seen from Equation (21) that the narrower the channel width, W , is the larger influence will the diffusion into the stagnant water be. For a case with a non-sorbing tracer and with $2W = 0.1$ m, $2\delta = 0.2$ mm, $R_f = 1$, $D_f = 10^{-9}$ m²/s, $\varepsilon_f = 1.0$, $R_f = R_p = 1$ and $D_p = 3 \cdot 10^{-11}$ m²/s, the stagnant water uptake is 3.8 times that in the matrix.

When narrow channels develop or when there is fingering the uptake into the stagnant water may dominate over the uptake into the matrix. The two interaction mechanisms cannot be distinguished as they influence the depletion in the water in the channel in exactly the same way. In theory the use of two tracers with different sorption properties could be used to determine the δ/W ratio. It must be remembered that the more sorbing tracer will “see” only the first part of the path and that the δ/W ratio may be different for the two tracers.

7.3 Case c: Diffusion from channel into stagnant water and into matrix from both channel and stagnant water

When there in addition is uptake into the matrix from the stagnant water this further increases the uptake via stagnant water. Then also the uptake develops differently over time. As no analytical solution was found for this case a numeric inversion of the solution obtained in Laplace space is used. Figures 7-4 and 7-5 show the concentration vs. time for a non-sorbing and a sorbing tracer. Data are the same as in the previous example.

It is seen from the figures that for a non-sorbing tracer the impact of diffusion into the stagnant water in the fracture is large whereas the addition of matrix diffusion from the stagnant water is small. The figure also shows a comparison of the analytical solution and that obtained by the numerical inversion for case b. The agreement is excellent.

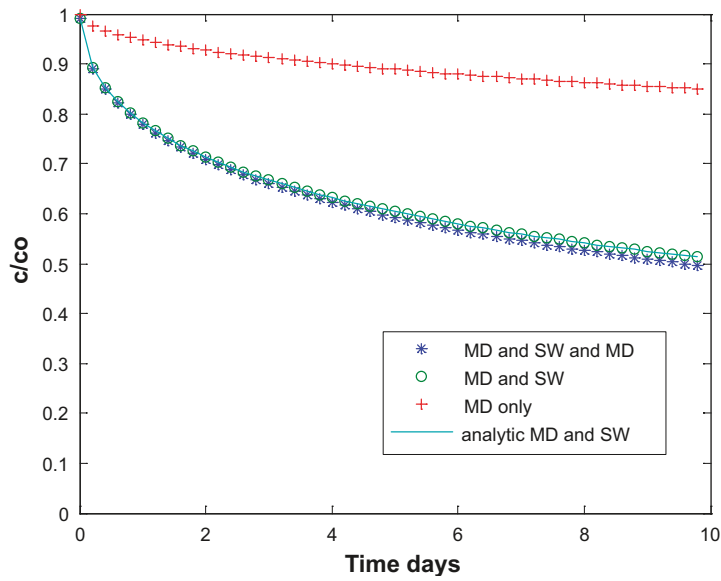


Figure 7-4. Diffusion from a channel into matrix and into stagnant water. Further diffusion into matrix from the stagnant water. Only matrix diffusion (MD) from channel. MD from channel and diffusion into stagnant water (SW) and MD from SW into matrix. Non-sorbing tracer. Analytical solution is also shown.

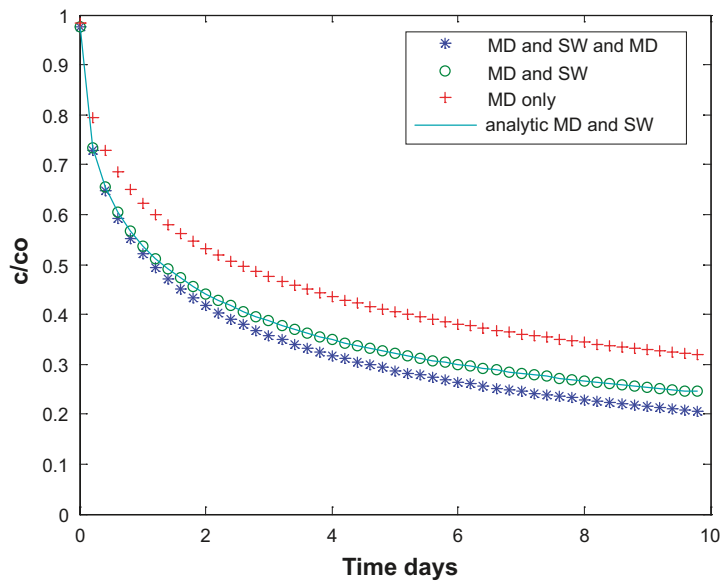


Figure 7-5. Diffusion from a channel into matrix and into stagnant water. Further diffusion into matrix from the stagnant water. Only matrix diffusion (MD) from channel. MD from channel and diffusion into stagnant water (SW) and MD from SW into matrix. Sorbing tracer $R_p = 101$. Analytical solution is also shown.

8 Some difficulties in interpretation of SWIW test

8.1 Summary of some idealised cases

Figure 8-1 summarises the impact of matrix and stagnant water diffusion on the recovery curve for some idealised cases. In figure A, a volume V_o of water with a non-interacting tracer has been injected into the fracture. The gray shaded area shows that the withdrawn water will have the same concentration as that injected for a recovered volume equal to that which was injected. The dashed curve indicates that dispersion effects distort the top hat form of the recovery curve.

In figure B a nonsorbing tracer which can diffuse into matrix and stagnant water has been given time t_{wait} to do so, reducing the concentration in the water in the channel to c_{twait} . All back pumped water up to volume V_o has the attained concentration.

In contrast, in figure C the back pumping is slow and some of the tracer that entered the matrix and other stagnant waters slowly diffuses back to the water in the channel. The form of the recovery curve is influenced by the diffusion processes.

All of these cases have of course in practice been influenced by dispersion as indicated by the dashed curves.

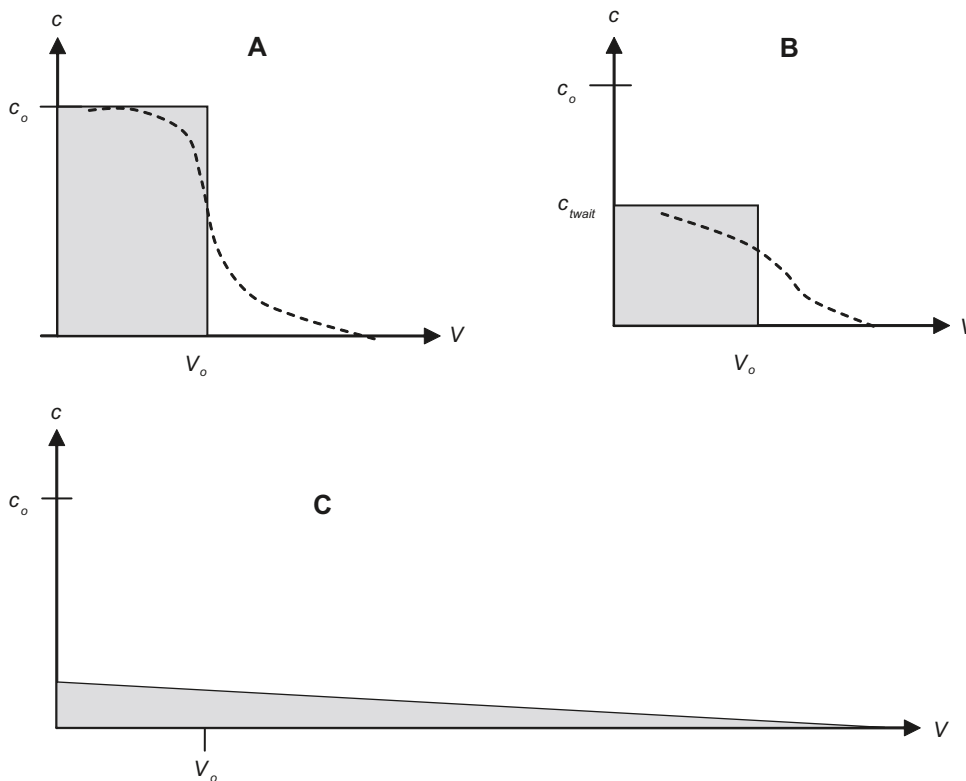


Figure 8-1. Idealized tracer concentration in withdrawn water. A: Non-interacting tracer. B: Tracer diffuses into matrix. C: slow withdrawal and back diffusion from matrix. Dashed curves indicate influence of dispersion.

8.2 Some complicating factors

In the preceding sections a number of different mechanisms that influence the spreading of a tracer pulse in a fracture and uptake of the tracer in the stagnant waters in contact with the traced water were illustrated and explored. The main and often only information available from a SWIW test are the recovery curves of one or more tracers. The parameters of a model that simulates the recovery curve can in principle be obtained by fitting the model to the experimental data by adjusting the parameters to obtain a best fit in some sense. Different parts and characteristics of the recovery curve will be differently influenced by the different mechanisms. Some of the mechanisms will influence the recovery curve in similar or even the same way making it impossible to determine which mechanisms were most active. For example the uptake into stagnant water and into matrix pore water gives the same effect for a given tracer. However, if two or more tracers are used with different diffusion coefficients and sorption coefficients then the two mechanisms may in principle be separated. Theoretically this also could give information on the fracture aperture and the FWS per volume of water plus the width of the channels if many different tracers were used.

Instantaneous reversible linear sorption on infill and channel/fracture surface will make tracers with different sorption properties see different parts of the fracture.

For slow withdrawal the long tail of the recovery can give additional information on diffusion effects and can in principle distinguish diffusion in stagnant waters from other dispersion effects such as Taylor dispersion.

However, there are several complicating factors. Heterogeneities in the flowpaths make the recovery curves deviate from the ideal. The presence of small and moderate size infill particles can strongly influence the rate of tracer uptake and the shape of the late time curve when slow recovery is done. As the matrix interaction parameters are the most important for subsequent use of the SWIW data in safety analysis the following may be noted. Late time slopes have been used to infer deviations from simple matrix diffusion in idealized systems. However, /Haggerty et al. 2000, 2001/ found that the late time slope of concentration vs. time in a log-log plot deviates considerably from that predicted for a system with only very large homogeneous blocks. See also the previous discussion in the section on heterogeneity.

8.3 Interpretation of SWIW tests

/Nordqvist and Gustafsson 2002/, present a literature review and make scoping calculations for homogeneous crystalline bedrock conditions regarding the use of single-well injection-withdrawal tests (SWIW). /Nordqvist and Gustafsson 2004/, also investigated evaluation aspects under heterogeneous crystalline bedrock conditions. Based on this a number of tests were designed, performed and analysed in deep boreholes in two Swedish sites. /Gustafsson et al. 2005, 2006, Wacker and Berg 2004/. In those analyses only one parameter governs the shape of recovery curve of a non-sorbing tracer namely the dispersion parameter. For a sorbing tracer, in addition, the retardation factor influences the shape of curve.

When only advection, Fickian dispersion and linear fast reversible sorption are active a SWIW test in a porous medium or an ideal fracture will give different concentration time curves for sorbing and non-sorbing tracers /Schroth et al. 2001, Cassiani 2005/. This can be utilised to determine the retardation coefficient, R_w , assuming that the dispersion coefficient is the same for both tracers. Heterogeneities and channelling effects can be expected to cause deviations from the idealized behaviour.

These interpretations are made using the advection-dispersion equation. The dispersion parameter(s) D_L or α are then first found by the use of a non-sorbing tracer which is assumed to be the same as for a sorbing tracer or it is determined by a simultaneous fitting of several tracer response curves. The solution derived by /Gelhar and Collins 1971/ and used e.g. by Schroth

assumes that dispersion in this model is due to the same mechanism as molecular diffusion, which it is not. See earlier discussions. The model shows that the recovery curve is very strongly correlated to and sensitive to the dispersion coefficient D_L . The dispersion coefficient used will strongly influence the retardation factor of the sorbing tracer in the fitting. It was found that if the model does not describe the processes correctly then the parameter values obtained by the fit may be considerably in error.

Other complicating factors arise from natural flow superimposed on the flow caused by injection and withdrawal and due to diffusion in stagnant waters that cause dispersion. These also cannot be lumped into the dispersion coefficient used to interpret ideal fracture recovery curves. /Altman et al. 2002/ in a study of a single-porosity fracture found that heterogeneity and slow drift during the waiting phase can carry the fluid to areas where the flow is lower during back pumping. They conclude that this may lead to dispersion and decrease in recovery. A model based on a single-porosity fracture is unlikely to match field data. Matrix diffusion must probably be included in the model.

8.4 Stagnant water diffusion effects included in the idealized single fracture model

When matrix/stagnant water diffusion is active a third parameter group is needed in addition to dispersion and the retardation factor for the sorption on the flowing fracture surfaces. The parameter group is given in Equation (18), repeated here.

$$\alpha = \frac{\delta}{R_w b} \frac{\varepsilon_f}{W} \sqrt{R_f D_f} + \frac{\varepsilon_p}{R_w b} \sqrt{R_p D_p} \quad (18)$$

The parameter group α can be determined from the test by curve fitting of a recovery curve using the model described. It applies to a system with constant material properties and a large extension of both matrix and stagnant water in the fracture. This parameter group will determine the interaction effects due to diffusion. The individual entities in the group cannot be distinguished using only one tracer. Several tracers with different diffusion and sorption properties must be used or independent information must be obtained on e.g. channel widths and apertures.

Note that of special interest for safety assessment verification of models is the last term in Equation (18).

Further complications arise when there are splays and particles in the flowpath which add to the FWS (inverse of the aperture b in the parameter group) and when the particles have a characteristic time for equilibration, which is similar to the duration of the experiment. Then information on the particle size and the diffusion and sorption properties of the particles is needed. As it must be assumed that the particles are not all of the same size and do not have the same properties additional parameters are needed. The addition of more parameters increases the difficulties in deriving values of them from a few SWIW tests. /Haggerty and Gorelick 1995/ suggested that all these properties could be lumped into a few parameters characterised by a distribution of characteristic times called the multirate approach. This has been successfully applied by /Haggerty et al. 2001/ in SWIW tests in a site with fractured dolomite. Nevertheless the authors concluded that the long time end of the characteristic times of the distribution found was far from that of interest to safety assessment.

8.5 Possible use of independent observation to constrain the interpretation

For safety assessment applications the FWS of the channel (inverse of the aperture b) in the parameter group is of the highest importance as it directly influences the velocity of a sorbing tracer. The velocity of the leading edge of a tracer, for a given flowrate in the channel will increase with the square of the aperture /Neretnieks 2006/.

It should be possible to at least constrain the aperture in the low end by using the cubic law. The empirical relation, Equation (14) or other empirical formulas is another alternative to constrain the aperture. Infill porosity and channel width could possibly be assessed by using tracers with different sorption properties. Different tracers may also help to assess the relative impact of matrix diffusion and diffusion into stagnant waters in the fracture. However, this would rely on the assumption that one already has independent information on sorbing tracer behaviour from e.g. laboratory measurements. This is in conflict with the wish to determine or at least verify that field matrix diffusion data agree with laboratory data.

8.6 Use of a hot water instead of a solute as a tracer

Some of the problems with solute tracers can be avoided if heat could be used as a tracer. The heat conductivity and heat capacity of rock and water are well known. These properties do not depend as much on other rock properties as diffusion does. Also the sensitivity to the presence of infill and conduction in stagnant water in the fracture has much less impact because the heat conductivity of water and rock do not differ as much as solute diffusivity does. One problem that comes instead is that the heat diffusivity is several orders of magnitude larger than that of solutes. A SWIW heat tracer test must be performed very rapidly. This is illustrated by some examples below.

The same equations can be used for heat transport as for solute diffusion, changing concentration for temperature and effective diffusivity $D_a = D_p/R_p$ for thermal diffusivity κ . The thermal diffusivity of rock is much larger than that of the water and the conduction into the stagnant water in the fracture can be neglected. Thus only case “b” is considered. However, in narrow channels the radial diffusion must be considered. Models for radial diffusion exist /Neretnieks 2006/. As the discussion below is intended to explore the possible use of heat it suffices to use a simplified treatment. The equations are given below.

For the rock.

$$\frac{\partial \vartheta}{\partial t} = \kappa \frac{\partial^2 \vartheta}{\partial z^2} \quad (22)$$

For the water in the fracture.

$$\rho_w c_{pw} \frac{d\vartheta_w}{dt} = \frac{1}{b} \lambda \frac{\partial \vartheta(t, z=0)}{\partial z} \quad (23)$$

For heat conduction.

$$\alpha = \frac{1}{b\sqrt{\kappa}} \frac{\lambda}{\rho_w c_{pw}} \quad (24)$$

$$\beta = \frac{1}{\sqrt{\kappa}} \quad (25)$$

Table 8-1. Data used in the examples.

Thermal conductivity of granite	λ W/mK	2.83
Thermal conductivity of water	λ_w W/mK	0.609
Thermal capacity of granite	ρc_p MJ/m ³ K	2.10
Thermal capacity of water	$\rho_w c_{pw}$ MJ/m ³ K	4.18
Thermal diffusivity of granite	$\kappa = \lambda / \rho c_p$ m ² /s	$13.5 \cdot 10^{-7}$
Thermal diffusivity of water	$\kappa_w = \lambda_w / \rho_w c_{pw}$ m ² /s	$1.65 \cdot 10^{-7}$

Figure 8-2 shows the temperature in the water in the fracture as a function of time. Injection and withdrawal times will have to be very short. In addition heat losses due to equipment and borehole effects must be mastered. However, if mastered, the aperture of the fractured could be assessed by independent means as the thermal properties are well know in contrast to the diffusive and sorptive properties and the disturbance by infill and conduction in stagnant water in the fracture are much smaller. The analysis of a heat pulse experiment would probably need the use of a finite element or difference model where the presence of water and equipment in the borehole can be handled and in which 3-dimensional conduction can be included.

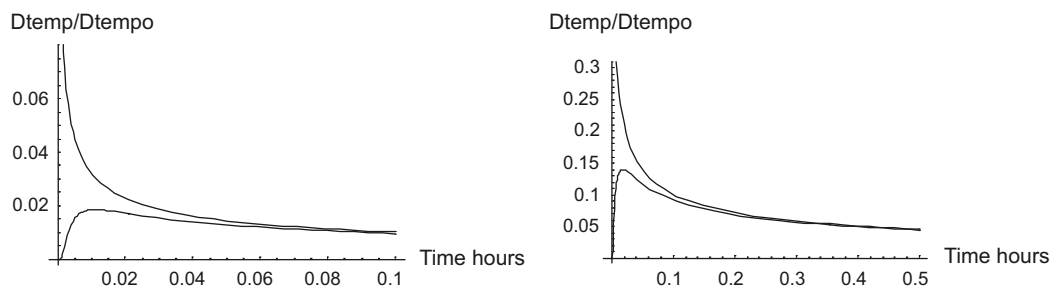


Figure 8-2. Temperature in water in fracture and 1 cm into matrix for 0.4 mm (left) and 4 mm (right) aperture (2b).

9 Discussion and conclusions

Tracer recovery curves from SWIW tests in fractures in crystalline rocks will be influenced by many different mechanisms and processes. When the aim, as in this report, is to obtain quantitative evidence of matrix diffusion effects in fractures in their natural environment, the tests should be designed in such a way that this process gives the strongest impact on the tracer recovery information. The fractures chosen for the tests should be simple in the sense that there should be little secondary coatings and as little infill as possible. Rock particles and infill will cause similar effects on the recovery as matrix diffusion, MD, and it may be difficult or even impossible to distinguish them. It has been shown that the parameter group α strongly influences the uptake into the stagnant waters. It contains information of the material properties of the tracer and the properties of the flowpath. The magnitude of the fracture surface per volume of water has a dominating influence. The material properties and the aperture cannot be separated without having additional independent information or possibly by the use of multiple tracers with different diffusion and sorption properties. Even then fundamental difficulties present themselves because the tracers will be exposed to different parts of the flowpath.

Tracers with different diffusion and sorption properties injected simultaneously will also, in theory, facilitate to distinguish matrix diffusion from diffusion in other stagnant waters. Possibly it would also constrain the range of apertures and channel widths of the channels in the fracture. With every new tracer introduced assumptions must be made on their diffusion and sorption properties relative to the other. One example of an assumption is that they have the same relation of diffusion and sorption coefficients in the fracture as they have had in the independent investigations. This seems reasonable for the diffusion coefficient because it is measured in boreholes under in situ conditions and is a predominantly physical process, /Löfgren and Neretnieks 2003/. The use of laboratory sorption coefficients is probably more doubtful as it is based on crushed rock samples. All assumptions underlying the interpretation of the tests must be clearly stated and reasons given for their reasonableness. Alternative assumptions could be tested to explore the sensitivity to them.

The mechanisms other than matrix diffusion that cause a tracer to be dispersed e.g. various forms of Taylor dispersion can cause significant spreading of a tracer pulse. Taylor dispersion is strongly influenced by the geometry and heterogeneities as well as the velocity distribution in the fracture. It will probably be difficult to separate dispersion due to these effects from dispersion caused by natural flow and from diffusion in stagnant waters, considering the time limitations in practice imposed on a SWIW test. The recovery information of the tailing part of the recovery curve contains valuable information on diffusion effects. For slow recovery the slope in log-log space of concentration vs. time can potentially be used as a strong indication of matrix diffusion effects provided other diffusion effects such as those carrying the tracer into stagnant or slowly moving water can be distinguished /Haggerty et al. 2000/.

A fairly rapid injection, a long waiting phase and short recovery phase has some advantages over a slow injection and withdrawal, and no or short waiting phase, because dispersion effects due to flow would be smaller. The maximum concentration during the recovery phase when the injected volume of traced water is recovered gives direct information of the “loss” of tracer from this volume during the waiting phase. This is of course true only provided other dispersion effects can be distinguished.

Interpretation of SWIW tests by curve fitting should be supplemented by considerations of which effects can be expected to dominate the recovery curves and which effects may shadow the pure matrix diffusion. Judicious weighting of the information from the recovery curve must be made.

Even if the matrix diffusion parameters could be obtained from SWIW tests they would necessarily only apply to the depth of matrix penetrated by the tracers during the test.

For safety assessment time scales the penetration depths for strongly sorbing tracers can be expected to be on the order of centimetres to tens of centimetres. Such penetration depths could nearly be attained using weakly sorbing tracers in a SWIW test. This would lend credence to the question whether sufficient rock depth had been explored. The difficulties encountered for the highly fracture Culebra dolomite /Haggerty et al. 2001/ may not apply to sparsely fractured granitic rock where the rock blocks are very much larger than the tracer penetration depths.

Fracture aperture has the strongest influence on matrix diffusion effects. It is desirable to attempt to find independent information that could be used to at least constrain the value of the aperture. The cubic law and empirical relations could be used to add confidence to the results obtained by curve fitting.

Forward modelling using laboratory and field data on matrix diffusion and laboratory data on sorption and assuming they are valid for in situ conditions could be used to “predict” the outcome. This approach can be used to analyse the effects of the other causes of “dispersion” and recovery such as Taylor dispersion in a heterogeneous fracture.

Possibly other independent tests and observations could be used. The use of a heat pulse is not so sensitive to conduction in stagnant water but would probably need considerable development before it could become practicable. Radon-222 emanating from rock in the extracted water could possibly also be used to constrain the fracture aperture /Neretnieks 2002b/.

The use of SWIW tests has many promising traits but there are a number of uncertainties and difficulties to surmount before they can be confidently used for predictive purposes for safety analysis over long time scales. However, they should be able to build credibility provided they are evaluated with sufficient insights and afterthought.

10 Acknowledgements

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Notation

b	Half fracture aperture
c_f	Concentration of solute in stagnant water
c_o	Initial concentration
c_p	Concentration in matrix pore water
c_{pw}	Heat capacity of water
c_s	Concentration in solid
c_w	Concentration of solute in channel
\tilde{c}	Concentration in Laplace space
dp	Particle diameter or thickness
D_L	Dispersion coefficient
D_f	Diffusion coefficient in stagnant water
D_p	Diffusion coefficient in pore water
D_w	Diffusion coefficient in free water
K_d	Sorption coefficient
L	Length of flowpath
M	Mass of solute instantaneously injected in a stream
Pe	Peclet number, vL/D_L
r	Radial distance
R_f	Retardation factor in stagnant water region
R_p	Retardation factor in matrix
R_w	Retardation factor in flowing channel
s	Laplace variable
t	Time
t_c	Characteristic time
T	Hydraulic transmissivity
v	Water velocity
V	Volume
W	Half width of fracture
x	Distance along flow direction
y	Distance in stagnant water region
z	Distance into matrix
δ	Half aperture of diffusive fracture
ε	Porosity
ε_f	Porosity in stagnant water region
ε_p	Porosity of rock matrix
ε_w	Porosity in flowing channel
$\eta_{0.01}$	Penetration depth
κ	Heat diffusivity
λ	Heat conductivity
μ	Viscosity
ϑ	Temperature
ρ	Density

Model for tracer uptake from a flat channel into matrix, into stagnant water in fracture and from there into matrix

Figure A1 shows three processes. The channel where traced water was injected has an aperture of $2b$ and is called the b -channel. It is $2W$ wide. It has infill which instantaneously is equilibrated with the traced water injected at $t = 0$. The solute can diffuse into the rock matrix adjacent to the b -channel. The solute can also simultaneously diffuse into the stagnant water in the fracture in contact with the b -channel. This is called the δ -channel. It has an aperture of 2δ . This channel also has infill and a retardation coefficient R_f . Solute in the δ -channels can also diffuse into the rock matrix. The retardation coefficient of the rock matrix is R_p . After the injection the flow stops in the model presented below. A model for flow in the b -channel can be found in /Neretnieks 2006/.

The model is formulated in mathematical terms by making a mass balance of the tracer in the channel from which tracer is lost by diffusion into the matrix and into the δ -fracture. Due to symmetry reasons only one quarter of the slit need be modelled. The z -coordinate starts at the wall of the b -channel and δ -fracture. The half aperture b can be expressed as the volume V of the channel per area A . When the channel contains small infill particles the decreased water volume and added sorption capacity of these must be accounted for.

With c denoting concentrations, D molecular diffusion coefficients and R retardation factors, the tracer concentration in the matrix in the pore water is described by

$$R_p \frac{\partial c_p}{\partial t} = D_p \frac{\partial^2 c_p}{\partial z^2} \quad (A1)$$

The mass balance for the water in the δ -fracture is:

$$R_f \frac{\partial c_f}{\partial t} = D_f \frac{\partial^2 c_f}{\partial y^2} + \frac{D_p \varepsilon_p}{\delta} \frac{\partial c_p}{\partial z} (z = 0) \quad (A2)$$

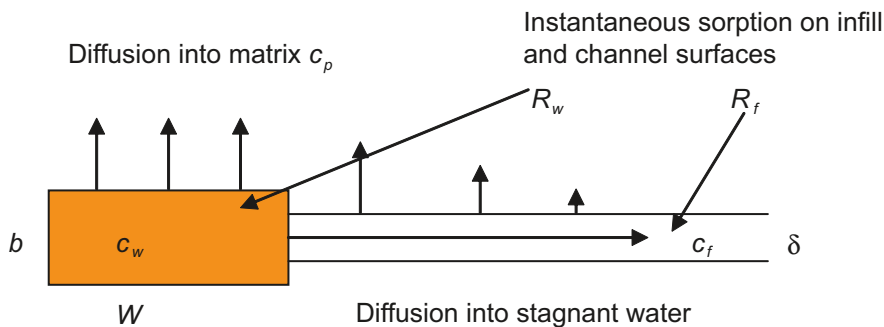


Figure A1. Solute from b channel (quarter shown) diffuses into matrix, into δ -fracture with stagnant water and from there into matrix. Channel has infill with retardation coefficient R_w and δ channel also has infill with retardation coefficient R_f .

The mass balance for the b -fracture with ε denoting porosities is:

$$R_w \frac{\partial c_w}{\partial t} = \frac{\delta}{b} \frac{D_f \varepsilon_f}{W} \frac{\partial c_f (y=0)}{\partial y} + \frac{D_p \varepsilon_p}{b} \frac{\partial c_p (z=0)}{\partial z} \quad (\text{A3})$$

The initial concentrations in the matrix and the δ -fracture are:

$$c_p (t=0, z) = 0 \quad \text{and} \quad c_f (t=0, y) = 0 \quad (\text{A4})$$

The initial condition for the b -fracture is:

$$c = c_o \quad (\text{A5})$$

If there is infill with which the tracer reacts instantaneously and with a reversible sorption equilibrium the concentration in water in the fracture will be less than that of the water injected carrying the tracer by a factor R_w . The sorption coefficients in equations A6, $K_d \rho$, apply to the infill particles in the channel, the infill in the δ -channel and the rock matrix respectively.

$$R_w = \frac{(\varepsilon + (1-\varepsilon)K_d \rho)}{\varepsilon}; \quad R_f = \frac{(\varepsilon_f + (1-\varepsilon)K_d \rho)}{\varepsilon_f}; \quad R_p = \frac{(\varepsilon_p + (1-\varepsilon)K_d \rho)}{\varepsilon_p} \quad (\text{A6})$$

The boundary conditions are; for the water rock interface

$$c_p (t, 0) = c_w (t) \quad \text{in the } b\text{-fracture and} \quad c_p (t, 0) = c_f (t) \quad \text{in the } \delta\text{-fracture} \quad (\text{A7})$$

and for z and y far away

$$c_p (t, z \rightarrow \infty) = 0 \quad \text{in the } b\text{-fracture and} \quad c_p (t, z \rightarrow \infty) = 0, \quad \text{in the } \delta\text{-fracture} \quad (\text{A8})$$

In this paper where short time are considered the infinity conditions are appropriate. Other conditions could be used for long times. For a limited extent of the rock and the δ -fracture, closed boundary conditions at a certain distance could be considered. This makes the solutions somewhat more complex and this case is not further considered here. The reader is referred to the appendix in /Neretnieks 2006/.

The solution to the above equations is obtained using Laplace transformation. We start by finding the concentrations in the δ -fracture and the matrix adjacent to it.

The Laplace transform of (A1) is

$$\tilde{c}_p s - c_p (t=0) = \frac{D_p}{R_p} \frac{d^2 \tilde{c}_p}{dz^2} \quad (\text{A9})$$

and the solution to (A9)

$$\tilde{c}_p = \tilde{c}_f e^{-z \frac{\sqrt{R_p s}}{\sqrt{D_p}}} \quad (\text{A10})$$

and the gradient at $z = 0$ is

$$\frac{d \tilde{c}_p (z = 0)}{dz} = -\tilde{c}_f \sqrt{\frac{R_p s}{D_p}} \quad (\text{A11})$$

Next the concentration in the δ -fracture is derived. The Laplace transform of (A2) is

$$R_f s \tilde{c}_f = D_f \frac{d^2 \tilde{c}_f}{dy^2} + \frac{D_p \varepsilon_p}{\delta} \frac{d \tilde{c}_p (z = 0)}{dz} \quad (\text{A12})$$

Introducing (A11) into (A12) gives:

$$R_f s \tilde{c}_f = D_f \frac{d^2 \tilde{c}_f}{dy^2} - \tilde{c}_f \frac{\varepsilon_p \sqrt{R_p D_p s}}{\delta} \quad (\text{A13})$$

This can be written:

$$B \tilde{c}_f = \frac{d^2 \tilde{c}_f}{dy^2} \quad (\text{A14})$$

where

$$B = \frac{R_f}{D_f} (s + A \sqrt{s}) \quad (\text{A15})$$

and

$$A = \frac{\varepsilon_p \sqrt{R_p D_p}}{\delta R_f} \quad (\text{A16})$$

The solution to (A13) with boundary conditions (A17 and A18)

$$\tilde{c}_f (y = 0) = \tilde{c}_w \quad (\text{A17})$$

$$\tilde{c}_f (y \rightarrow \infty) = 0 \quad (\text{A18})$$

is:

$$\tilde{c}_f = \tilde{c}_w e^{-y\sqrt{B}} \quad (\text{A19})$$

The gradient at $y = 0$ is:

$$\frac{d\tilde{c}_f(y=0)}{dy} = -\tilde{c}_w \sqrt{B} = -\tilde{c}_w \sqrt{\frac{R_f}{D_f}} \sqrt{s + A\sqrt{s}} \quad (\text{A20})$$

This gradient determines the rate of uptake of solute from the b-channel

Equations (A11) and (A20) will be used when solving the equations for the b -channel and δ -fracture. Together they determine the combined and simultaneous uptake of solute from the b -fracture into the matrix and into the δ -fracture.

The Laplace transform of (A3) is:

$$\tilde{c}_w s - c_w(t=0) = \frac{\delta}{R_w b} \frac{D_f \varepsilon_f}{W} \frac{d\tilde{c}_f(y=0)}{dy} + \frac{D_p \varepsilon_p}{R_w b} \frac{d\tilde{c}_p(z=0)}{dz} \quad (\text{A21})$$

Which by introducing (A20) and (A11) becomes:

$$\tilde{c}_w s - c_w(t=0) = -\frac{\delta}{R_w b} \frac{D_f \varepsilon_f}{W} \tilde{c}_w \sqrt{\frac{R_f}{D_f}} \sqrt{s + A\sqrt{s}} - \frac{D_p \varepsilon_p}{R_w b} \tilde{c}_w \sqrt{\frac{R_p s}{D_p}} \quad (\text{A22})$$

From (A22) \tilde{c}_w is obtained

$$\tilde{c}_w = \frac{c_o}{s + \frac{\delta}{R_w b} \frac{\varepsilon_f}{W} \sqrt{R_f D_f} \sqrt{s + A\sqrt{s}} + \frac{\varepsilon_p}{R_w b} \sqrt{R_p D_p s}} \quad (\text{A23})$$

Inversion of (A23) by analytic means has not been attempted but it is readily solved by numerical inversion /Hollenbeck 1998, Neretnieks 2006/. However, when there is no diffusion from the δ -fracture into the rock matrix $A = 0$ and (A23) simplifies to:

$$\tilde{c}_w = \frac{c_o}{s + \alpha \sqrt{s}} \quad (\text{A24})$$

Where

$$\alpha = \frac{\delta}{R_w b} \frac{\varepsilon_f}{W} \sqrt{R_f D_f} + \frac{\varepsilon_p}{R_w b} \sqrt{R_p D_p} \quad (\text{A25})$$

The concentration in the matrix is obtained using (A10) which also is valid for uptake from the b -fracture.

$$\tilde{c}_p = \tilde{c}_w e^{-z\sqrt{\frac{R_p s}{D_p}}} = \frac{c_o}{s + \alpha\sqrt{s}} e^{-z\sqrt{\frac{R_p s}{D_p}}} \quad (\text{A26})$$

Inversion using Duhamel's theorem and from tables of known inverses.

$$L^{-1}\left\{\frac{1}{\sqrt{s}}\right\} = \frac{1}{\sqrt{\pi t}} \quad (\text{A27})$$

$$L^{-1}\left\{\frac{e^{-\beta z\sqrt{s}}}{\sqrt{s}}\right\} = \frac{e^{-\frac{z^2\beta^2}{4t}}}{\sqrt{\pi t}} \quad (\text{A28})$$

$$L^{-1}\left\{\frac{1}{\alpha + \sqrt{s}}\right\} = \frac{1}{\sqrt{\pi t}} - \alpha e^{a^2 t} \text{Erfc}(\alpha\sqrt{t}) \quad (\text{A29})$$

gives:

$$\tilde{c}_w = \frac{c_o}{\sqrt{\pi}} \int_0^t \frac{1}{\sqrt{t-\tau}} \left(\frac{1}{\sqrt{\pi\tau}} - \alpha e^{a^2\tau} \text{Erfc}(\alpha\sqrt{\tau}) \right) d\tau \quad (\text{A30})$$

and for \tilde{c}_p we have:

$$\tilde{c}_p = \frac{c_o}{s + \alpha\sqrt{s}} e^{-z\sqrt{\frac{R_p s}{D_p}}} \quad (\text{A31})$$

which when inverted gives:

$$c_p = \frac{c_o}{\sqrt{\pi}} \int_0^t \frac{e^{-\frac{\beta^2 z^2}{4(t-\tau)}}}{\sqrt{t-\tau}} \left(\frac{1}{\sqrt{\pi\tau}} - \alpha e^{a^2\tau} \text{Erfc}(\alpha\sqrt{\tau}) \right) d\tau \quad (\text{A32})$$

where

$$\beta = \sqrt{\frac{R_p}{D_p}} \quad (\text{A33})$$