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Äspö Hard Rock Laboratory

TRUE Block Scale continuation project

Retention processes discrimination for various assumptions of fracture heterogeneity

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VTT Processes

November 2003

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Abstract

This study examined the effects of the different kinds of heterogeneities on the retention of solutes in tracer experiments. The approach was to include variations in both material and the flow field properties. It was shown that in the longitudinal and lateral types of heterogeneities the retention does not depend on the spatial distribution of the different properties, but only on the effective “quantity” of the different properties. However, a possible correlation between the flow field and a given material property will affect the effective retention properties.

Non-sorbing tracers with different pore diffusivities could be especially useful for determining the heterogeneities. Such kind of tracers could be for example nano-particles. Non-sorbing tracers can give information especially on the diffusion-controlled retention and possible heterogeneity that is associated with a limited capacity of the available immobile pore spaces. Different pumping rates can also help to reveal the heterogeneity that is associated with a limited capacity of the immobile pore space. Diffusion-controlled retention is very sensitive to the flow rate, so decreasing the flow rate will retard the tracer discharge very clearly. If the immobile pore space gets saturated this should manifest itself as a deviation from the power-law tailing of the tracer breakthrough curve as the flow rates are decreased.

Sammanfattning

Denna studie undersöker effekterna av olika sorters heterogenitet på retentionen av lösta ämnen i samband med spårförsök. Tillvägagångssättet var att inkludera variationer både i bergets materialegenskaper och i flödesfältets egenskaper. Det kunde påvisas att för longitudinella och laterala typer av heterogenitet så beror retentionen inte på den rumsliga fördelningen av egenskaper, utan endast på "effektiva medelvärden" på de olika egenskaperna längs flödesvägen. Att notera är att en möjlig korrelation mellan flödesfältet och en given materialegenskap påverkar de effektiva retentionsegenskaperna.

Icke-sorberande spårämnen med olika pordiffusiviteter kan vara speciellt värdefulla för bestämning av effekten av olika typer av heterogenitet. Ett sådant spårämne skulle kunna vara nano-partiklar. Icke-sorberande spårämnen kan också ge information, speciellt om retention kontrollerad av diffusion och heterogenitet kopplad till ändlig kapacitet hos tillgänglig, immobil porvolym. Retention kontrollerad av diffusion är mycket känslig för flödet, där ett minskat flöde kommer att effektivt fördröja genombrottet av ett givet spårämne. Om flödet minskas och om den immobila zonen mättas kan detta påvisas som en avvikelse från det klassiska potensfunktionsavtagandet i genombrottskurvans svans.

Table of Content

Executive Summary

This study examined the effects of the different kinds of heterogeneities on the retention of solutes in tracer experiments. The approach was to identify a set of simple type heterogeneities. The type heterogeneities include variations in both material and flow field properties. It was shown that more complicated heterogeneities can be constructed by using type heterogeneities. It was also shown that in the longitudinal and lateral types of heterogeneities retention does not depend on the spatial distribution of the different properties, but only on the effective “quantity” of the different properties. This holds both for the material and flow field properties if they vary independently. More precisely, the retention depends on the summed local material properties that are weighted by the local flow field properties. This means that a possible correlation between the flow field and a given material property will affect the effective retention properties. Changing material properties as a function of the depth in the immobile zone may cause large differences in the retention if the flow rates are varied. In this case the retention may show characters of limited matrix diffusion for low flow rates, but unlimited matrix diffusion for larger flow rates. Unlimited matrix diffusion show long power-law tailing in the breakthrough curve whereas a limitation in matrix will change the tailing steeper and the tracer discharge pulse gets more compact.

The averaging nature of the retention along the flow path makes it very difficult to study the heterogeneity using tracer tests. Retention depends on a grouped parameter and determination of the individual parameters requires results from several tracers with different properties. Especially useful could be non-sorbing tracers with different pore diffusivities. That kind of tracers could be for example nano-particles. Non-sorbing tracers can give information especially on the diffusion-controlled retention and possible heterogeneity that is connected to the limited capacity of the immobile pore spaces. With the same pumping rates the high diffusivity tracers may show the limitation in capacity of the pore space as the low diffusivity tracers still reflect the diffusion control of the retention.

Different pumping rates can also help to reveal the heterogeneity that is connected to the limited capacity of the immobile pore space. Diffusion-controlled retention is very sensitive to the flow rate, so that decreasing the flow rate should increase retardation clearly. Low enough pumping rates should cause a saturation of the limited immobile pore space that will change the characteristics of the retention. If the dominating immobile pore space gets fully saturated this should manifest itself by changing the tracer breakthrough curve from an asymmetric power-law tailing curve to a retarded Gaussian-type of curve.

1 Introduction

TRUE Block Scale Continuation (BS2) -Project is a continuation of the tracer tests performed in a network fractures at tens of metre scale. The overall objective of BS2 can be summarised as: "Improve understanding of transport pathways at the block scale, including assessment of effects of geometry, macrostructure, and microstructure". This work is a part of the BS2A modelling, which is performed to support the planned BS2B tracer test phase. The issue addressed by this modelling is to improve retention process identification under various assumptions of heterogeneity.

Heterogeneity in both immobile zone and flow field properties may have an important impact on the retention. Identification of the relative contribution of retention processes and effective retention parameters under various situations of heterogeneity helps interpretation of the experimental results and process discrimination in the evaluation phase. In addition, it could be possible to take the heterogeneity into account in the test planning. Basically this offers tools for the process discrimination of the evaluation phase to better explain differences between the flow paths by the differences in the pore space microstructure.

2 Objectives

This activity explores the possibility of improved retention process identification under various assumptions of heterogeneity. Type heterogeneities presented in Appendix C in Poteri et al., (2002) form basis of the analysis. Analysis of the type heterogeneities gives opportunity to examine which experimental parameters (e.g. flow rate, path length, and different tracers) need to be considered to explore the effects of heterogeneities. Expected results of this activity are:

- Understanding of which types of heterogeneity that can be examined by tracer tests and which are perhaps inherently impossible to elucidate by using tracer experiments.
- Connection between tracer test parameters (flow rates, path length) and correspondingly mapped heterogeneities. This can possibly be used to guide the test planning.
- How different tracer "cocktails" can be used to help process discrimination for heterogeneous flow paths. This can possibly be used to guide the test planning.

3 Influence of the heterogeneity on retention

Spatial distribution of the heterogeneity along the transport path cannot be determined from the tracer test data. Several tracer tests that apply different flow conditions and different tracers can give information how the effective retention properties over the whole transport path behave if the transport conditions are perturbed. However, there is no unique correspondence between the effective retention properties and detailed structure of the heterogeneity.

Application of the geological information to assess the retention properties may be feasible because of the integrating nature of the transport. It is a great benefit that in order to assess the retention we do not need to know the detailed structure of the pore space along the flow path. The difficulty in the estimation of the retention properties comes from the characteristics of the geological information that is collected from the boreholes. How do we transform the point measurements of the geological data to the required effective retention properties along the flow paths?

This study examines how the local heterogeneity affects effective retention properties of the transport paths. The approach is to analyse a set of simplified cases for different kinds of heterogeneities. These are called “type heterogeneities” and basically they are taken from the report Poteri at al. (2002) with some additions. The analysed type heterogeneities are introduced in the next chapter.

Tracer mass discharge through a homogeneous path can be expressed by equation (3-1) (pulse input)

$$\frac{\dot{m}(x,t)}{m_0} = H(t - R_a t_w)\frac{u}{\sqrt{\pi}\left(t - R_a t_w\right)^{3/2}}\, e^{-\frac{u^2}{t - R_a t_w}} \qquad (3\text{-}1)$$

where H is the Heaviside’s step function, t_w is the groundwater transit time, R_a is the retardation coefficient of the surface sorption and parameter u is controlling the retention that is caused by the diffusion to the immobile pore space and possible sorption in there. Parameter u is defined by

$$u = \frac{D_e\, x}{v\ 2b}\sqrt{\frac{R_p}{D_p}} = \sqrt{D_e \varepsilon R_p}\,\frac{t_w}{2b} = \sqrt{D_e \varepsilon R_p}\,\frac{W\,x}{Q} \qquad (3\text{-}2)$$

where D_e effective diffusion coefficient in the immobile pore space, x length of the flow path, v groundwater flow velocity, $2b$ aperture of the flow path, R_p retardation coefficient in the immobile pore space, D_p pore diffusivity in the immobile pore space, ε porosity of the immobile pore space, W width of the flow path and Q flow rate through the flow path. Different parameterisations of the parameter u have been applied. Cvetkovic et al. (2002) use $B=2u$ and Neretnieks (e.g. 2002) applies MPG·FWS/q = $2u$, where MPG is the material parameter group and FWS is the flow wetted surface.

Instead of the groundwater transit time it is more correct to use the transit time of the solute particles in the advective flow field in the equation (3-1). For the idealised transport channel, that has only one flow velocity, there is no difference between the t_w of the solute particles and water. However, this may also be extended to well-mixed conditions in case of variable advection. In the well mixed conditions (diffusional mixing of the solute particles in the flow field) the advective transit time for all solute particles through the transport channel is a well defined mean transit time t_w in spite of the variations in the flow velocity. This means that equation (3-1) can be applied to transport channels that are heterogeneous both in material and flow field properties if well-mixed conditions in the flowing part of the channel can be assumed. If well-mixed conditions do not prevail over the whole flowing part of the channel then that channel can be divided into a set of narrower channels that are well mixed.

Characteristic features for the transport in the case of the fracture flow is that the effects of the local heterogeneity in the overall retention are attenuated and the transport path is characterised only by the effective retention along the path. It can be shown that the total retention property u can be summed from the retention properties of the different materials or "legs" of the flow path (see e.g. Appendix B in Poteri, 2002). In the general case the equation (3-2) can be given as an integral along the transport channel

$$u = \int_0^x \sqrt{D_e(s)\varepsilon(s)R_p(s)} \frac{W(s)}{Q} ds \qquad (3\text{-}3)$$

It is emphasised that this study concentrates on the retention properties. The retention needs to be distinguished from the advective delay. A change in the volume of the transport channel will cause the tracer pulse to discharge at a later time i.e. it is delayed but it is not retarded. Typical for the advective delay is that it does not affect the peak discharge rate. Retention also delays the tracer breakthrough but it does it by distributing the tracer discharge over a longer period of time so that also the level of the peak tracer discharge changes. This means that in this study we usually omit the possible differences in the advective delay between different conceptualisations.

4 Type heterogeneities

Physically the retention processes are represented by a large number of parameters, but as it is indicated in the equations (3-2) and (3-3) the overall retention can be represented by two grouped parameters. One of the parameters describes the effects of the flow field and another the effects of the material properties along the transport path.

This is taken into account in the analysis by also dividing the heterogeneities into two groups: i) effects caused by the variations in the flow field along the flow path and ii) spatially variable properties of the immobile pore space. This simplifies the problem and it also makes it easier to perceive the relationships between the retention and different processes.

Heterogeneous transport paths are examined by introducing two different properties along the flow path according to the following assumptions:

- The changed property may be assigned to the flow field or to the material properties.
- The flow path is conceptualised by a channel that has rectangular cross-section.
- Well-mixed conditions prevail at all times in the flow channel.

This means that equation (3-3) is simplified to the sum that is presented in equation (4-1)

$$u = \sum_{i=1}^{2} \left(\sqrt{D_e \varepsilon R_p}\right)_i \left(\frac{WL}{Q}\right)_i \qquad (4\text{-}1)$$

Parentheses in the equation (4-1) indicate that the material properties and flow field properties are varied as separate groups. Analysed cases are simplified examples of the different types of the heterogeneity and they are called type heterogeneities in this report.

The majority of the type heterogeneities has been introduced already in Appendix C of Poteri et al. (2002). In the present analysis those type heterogeneities have been supplemented by a case of varying channel width. The type heterogeneities are presented in Figure 4-1 The effects of the different type heterogeneities on the retention are discussed more in detail in Chapters 5 and 6.

The cases in Figure 4-1 are considered to be the basic “building blocks” of the heterogeneity in a sense that conclusions drawn from the type heterogeneities can be generalised to more complex cases. This can be done by dividing the flow path into a sequence of the type heterogeneities. The additive nature of the retention parameter, that is also demonstrated by the equation (4-1), makes this possible. For example, a square board-like heterogeneity can be constructed using results of the type heterogeneities M2 and M3 (Figure 4-1) and applying the summed up retention property (equation (4-1) along the transport channel.

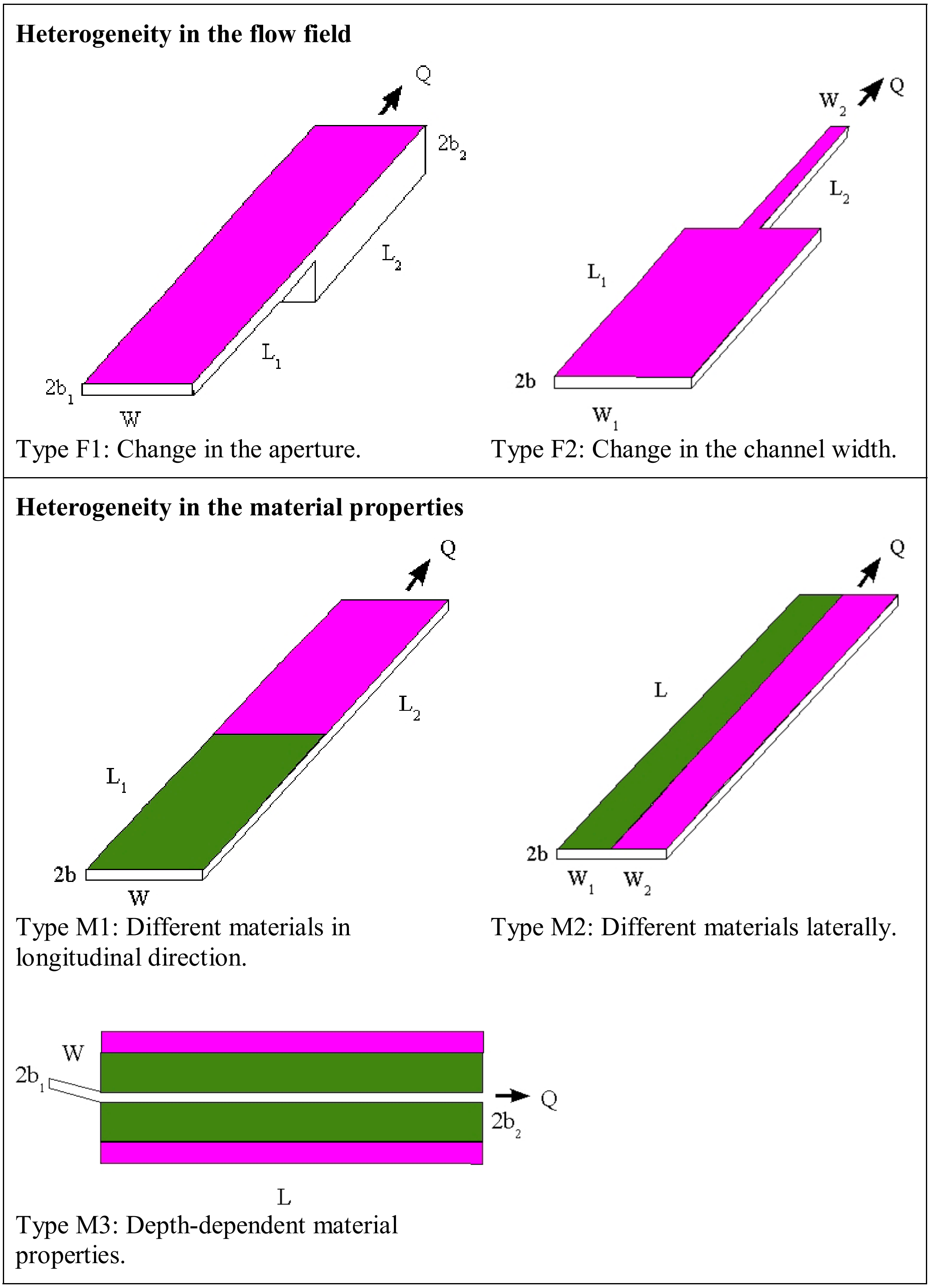
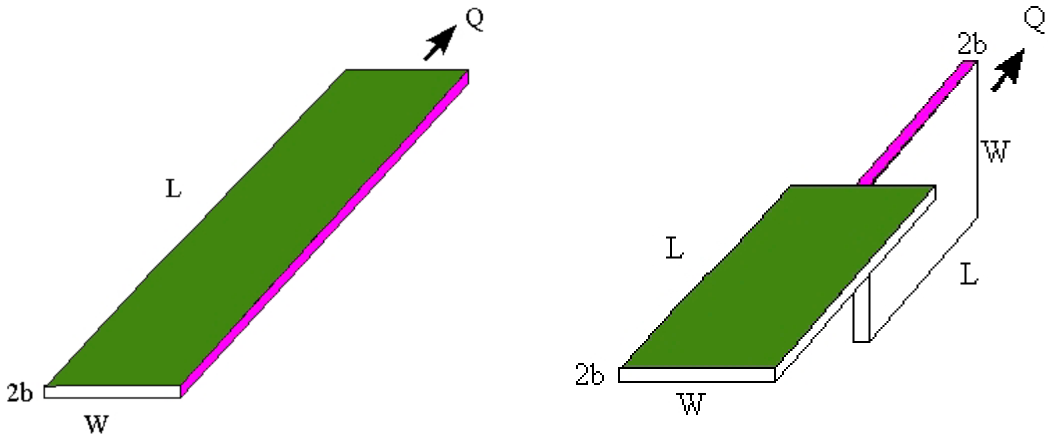


Figure 4-1. *Type heterogeneities that are analysed in this report. Most of the type heterogeneities have been introduced in the Poteri et al. (2002) Appendix C. Different material properties are represent by magenta and green colours.*

An example of a more complex heterogeneity that is composed the type of heterogeneities is illustrated in Figure 4-2. It describes a case where diffusion can take place both "vertically" through the fracture walls, but also laterally through the side walls of the transport channel. This kind of situation may arise, for example, if the transport channel is surrounded by stagnant pools (magenta) and at the same time diffusion can also take place to e.g. altered wall rock (green). To build this example using the type heterogeneities requires a combination that includes almost all different type heterogeneities that are presented in Figure 4-1. Two successive legs that have different material properties (type M1 heterogeneity) and both channel width and aperture change (Types F1 and F2 heterogeneities) have an equivalent retention to this example. It is noted that in this case the application of the type heterogeneities does not preserve the advective groundwater transit time, but the retention properties of these two cases are the same.

The reasoning to end up with the type heterogeneities that are given in the Figure 4-2 is based on the assumption of the well mixed conditions in the flowing part of the transport channel. This means that for the transported solute particles the probabilities to reach the "green" or "magenta" coloured pore space are independent. These probabilities can be thus evaluated separately and then summed up to the combined effect. Similarly, the retention in the "green" coloured material is independent on the retention in the "magenta" coloured material and they can be evaluated separately and then summed up at the end.



Example of heterogeneity where is different material behind the different "side walls" of the transport channel.

The example case divided into type heterogeneities. For the flow field part both F1 and F2 are applied and for the material property part M1 is applied (cf. Figure 4-1)

Figure 4-2. *Division of a heterogeneous transport channel into type heterogeneities.*

5 Heterogeneity in the material properties

5.1 Longitudinal heterogeneity

Heterogeneity of the material properties in the longitudinal direction along the transport path is present as the type M1 heterogeneity in Figure 4-1 (Figure 5-1). Following equation (4-1) the transport path is divided into two parts. The first part is for the material indicated by magenta (material 1) and the second part is for the green material (material 2). Retention parameter u can now be written separately for both of the parts. The retention parameter of the whole transport part is a sum of the two parts. The effective total retention parameter is thus

$$\begin{aligned} u &= \left(\sqrt{D_e \varepsilon R_p}\right)_1 \left(\frac{WL}{Q}\right)_1 + \left(\sqrt{D_e \varepsilon R_p}\right)_2 \left(\frac{WL}{Q}\right)_2 \\ &= \varepsilon_1 \sqrt{D_{p1} R_{p1}} \frac{W L_1}{Q} + \varepsilon_2 \sqrt{D_{p2} R_{p2}} \frac{W L_2}{Q} \end{aligned} . \quad (5\text{-}1)$$

This equation can also be written as

$$u = \left(\frac{L_1}{L}\varepsilon_1\right)\sqrt{D_{p1} R_{p1}} \frac{W L}{Q} + \left(\frac{L_2}{L}\varepsilon_2\right)\sqrt{D_{p2} R_{p2}} \frac{W L}{Q} , \quad (5\text{-}2)$$

where $L = L_1 + L_2$, indicating that this type heterogeneity is equivalent to any case where the effective porosities of the materials 1 and 2 over the whole channel are $(L_1/L)\ \varepsilon_1$ and $(L_2/L)\ \varepsilon_2$, respectively.

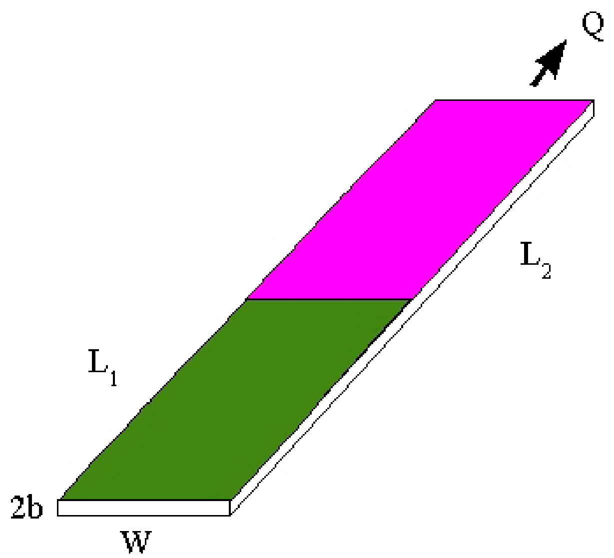


Figure 5-1. *Heterogeneity in the longitudinal direction along the transport channel (denoted as type M1 in Figure-4-1).*

5.2 Transversal or lateral heterogeneity

Transversal or lateral heterogeneity is presented as type M2 heterogeneity in Figure 4-1 (Figure 5-2). Equation (4-1) applies also in this case. It is written here using the same notation as in the equation (5-2), i.e. for the effective porosity of the different materials. This gives the same result as equation (5-2).

$$
\begin{aligned}
u &= \left(\sqrt{D_e \varepsilon R_p}\right)_1 \left(\frac{WL}{Q}\right)_1 + \left(\sqrt{D_e \varepsilon R_p}\right)_2 \left(\frac{WL}{Q}\right)_2 \\
&= \varepsilon_1 \sqrt{D_{p1} R_{p1}} \frac{W_1\ L}{Q} + \varepsilon_2 \sqrt{D_{p2} R_{p2}} \frac{W_2\ L}{Q} \qquad , \quad (5\text{-}3) \\
&= \left(\frac{W_1}{W}\varepsilon_1\right)\sqrt{D_{p1} R_{p1}} \frac{W\ L}{Q} + \left(\frac{W_2}{W}\varepsilon_2\right)\sqrt{D_{p2} R_{p2}} \frac{W\ L}{Q}
\end{aligned}
$$

where $W = W_1 + W_2$. Note, that this result is a consequence of the assumption that well-mixed conditions prevail all the time in the flow channel. Similar equivalence as in the case of the heterogeneity M1 holds also for this case, i.e. it is not possible to distinguish heterogeneity types M1 and M2 from the breakthrough curves if the effective porosities over the transport channel are the same. From the retention point of view it does not matter how the heterogeneity is distributed along the transport channel.

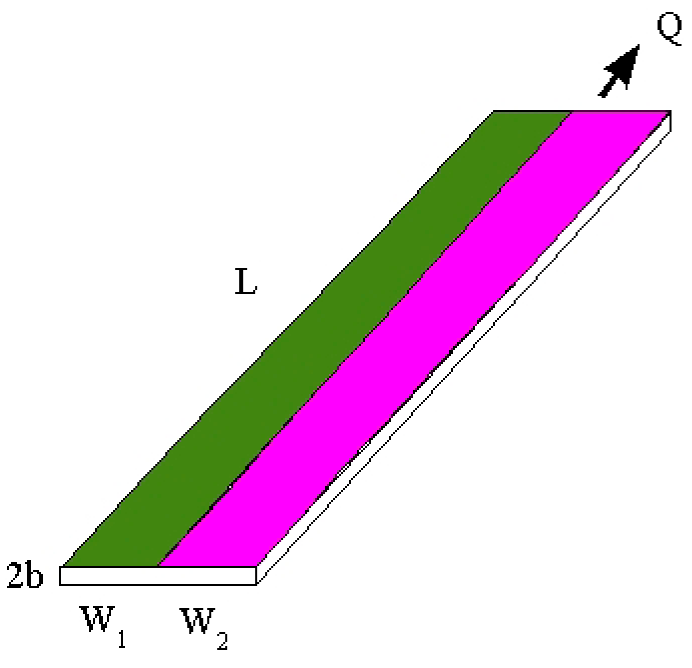


Figure 5-2. *Heterogeneity in the lateral or transverse direction (denoted as type M2 in Figure 4-1).*

5.3 Depth-dependent / immobile zone capacity

Depth-dependent material properties in the immobile pore space cause dynamic behaviour in the effective retention properties. For high flow rates the solute particles have had low probability to penetrate deep into the immobile zone. Thus, in that case the breakthrough curve mainly reflects the retention properties that are connected to the immobile zone closest to the fracture. At lower flow rates solute particles have higher probability to penetrate deeper into the immobile zone. Possible changes in the diffusion properties at some depth inside the immobile zone affects the retention properties as a function of the flow rate. This kind of situation is illustrated in Figure 5-3.

The response of this system under different conditions could be profoundly different because the principal retention process may change. Saturation of the dominating immobile pore space may change the retention from a matrix diffusion like behaviour to a equilibrium sorption type behaviour. Another question is that this may not be clearly observable in the in-situ tests because the time scale of the test and the background flow field limits the dynamic range in the applied flow conditions.

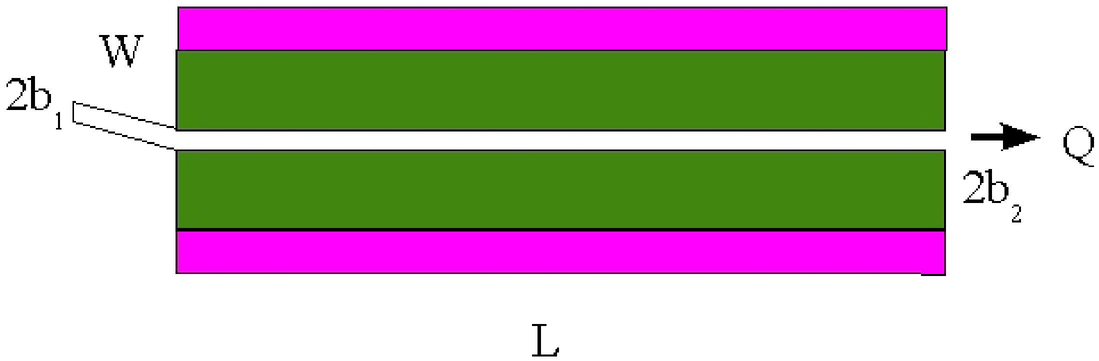


Figure 5-3. *Depth depending material properties. In this case there is a layer of "green" material closest to the fracture flanked by "magenta" material.*

6 Heterogeneity in the flow field

6.1 Aperture variation

The advective transit time is not directly connected to the retention properties. Naturally, the volume of the transport channel affects the breakthrough times of the solute particles because of the advective delay, but the advective delay is not the same as the retention. This becomes evident in the heterogeneity F1 in Figure 4-1 (and Figure 6-1) where the aperture of the flow path changes. In this type heterogeneity part of the channel length has small aperture and the other half has large aperture. All other parameters (*Q*, *W*, *L* and material properties) are kept constant. Applying this to equation (4-1) and writing it for the two parts of the transport path separately gives

$$\begin{aligned} u &= \left(\varepsilon\sqrt{D_p R_p}\right)_1 \left(\frac{WL}{Q}\right)_1 + \left(\varepsilon\sqrt{D_p R_p}\right)_2 \left(\frac{WL}{Q}\right)_2 \\ &= \varepsilon\sqrt{D_p R_p}\,\frac{W\,L_1}{Q} + \varepsilon\sqrt{D_p R_p}\,\frac{W\,L_2}{Q} = \varepsilon\sqrt{D_p R_p}\,\frac{W\,L}{Q} \end{aligned}, \qquad (6\text{-}1)$$

where $\left(\varepsilon\sqrt{D_p R_p}\right)_1 = \left(\varepsilon\sqrt{D_p R_p}\right)_2 = \varepsilon\sqrt{D_p R_p}$ and $L=L_1+L_2$. The resulting retention property is exactly the same as for the constant aperture transport path.

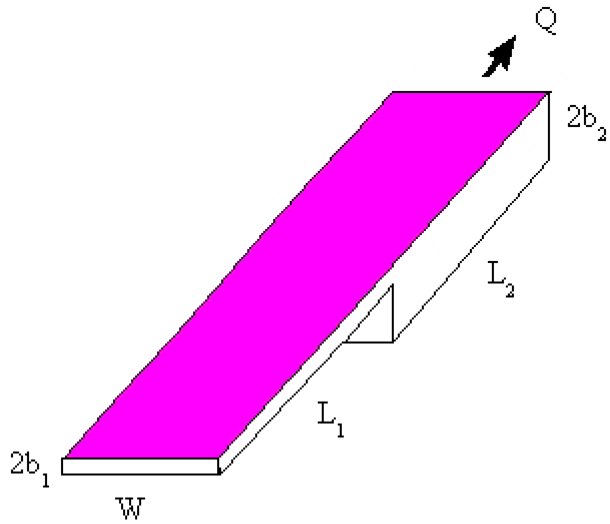


Figure 6-1. *Aperture variation simplified to type heterogeneity F1, which is composed of two different apertures.*

6.2 Changes in the width of the transport path

Change in the channel width also affects the retention. This is examined by the type heterogeneity F2, which contains an abrupt change in the channel width as illustrated in Figure 6-2 (cf. also Figure 4-1). In this case the effective retention along the flow path is calculated by applying equation (4-1).

$$\begin{aligned} u &= \left(\varepsilon\sqrt{D_p R_p}\right)_1 \left(\frac{WL}{Q}\right)_1 + \left(\varepsilon\sqrt{D_p R_p}\right)_2 \left(\frac{WL}{Q}\right)_2 \\ &= \varepsilon\sqrt{D_p R_p}\left(\frac{W_1 L_1}{Q} + \frac{W_2 L_2}{Q}\right) \end{aligned} \quad , \qquad (6\text{-}2)$$

where $\left(\varepsilon\sqrt{D_p R_p}\right)_1 = \left(\varepsilon\sqrt{D_p R_p}\right)_2 = \varepsilon\sqrt{D_p R_p}$. This may also be explained in the following simple way. The transported solute molecules experience two competing processes. Drift along the advective field in the transport channel and diffusion into the immobile zone. Retention along a transport channel depends on the balance between these two processes. We may examine the situation by looking to the advective flow field from the immobile zone. From that view point the solute mass flux changes if the ratio *Q/W* changes.

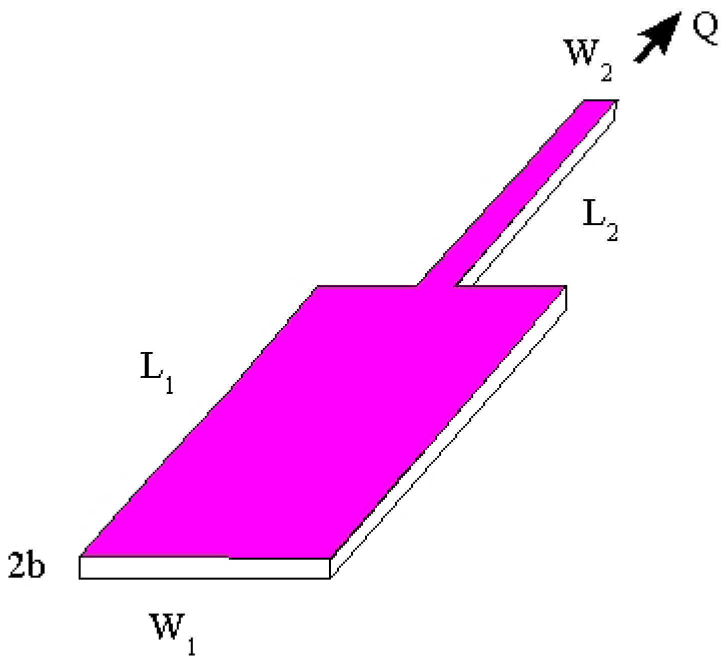


Figure 6-2. *Change in the channel width (type heterogeneity F2).*

7 Tracer test parameters vs. heterogeneity

7.1 Pumping rate

Variations in the pumping rate cannot be used to examine the lateral or longitudinal types of the heterogeneity (M1, M2, F1 and F2 in Figure 4-1). These heterogeneities manifest themselves as an effective retention that scales according to the *WL/Q*. In the evaluation phase these types of heterogeneity shows up as e.g. changed effective porosity as indicated in the equations (5-3) and (5-2).

Different pumping rates can be used to increase confidence in the dominating retention process at the tested scale. With one pumping rate it may be difficult to separate retention from the advective delay, especially if only one type of tracer is applied.

Let us consider three main processes that affect the tracer residence time: advection, equilibrium sorption and matrix diffusion. Each of these scale differently if the flow rate is changed. From scrutiny analysis of the breakthrough curve it may difficult to discriminate between advective delay together with dispersion and retention. In principal, the difference between the delay and retention is that in the case of retention the discharged mass flux is also lowered by the same ratio as the breakthrough is retarded or delayed. This scaling factor for different processes is the following:

- In the case of the advection there is no retention but only delay of the input pulse. This means that there is no change in the discharge rate compared to the input mass flux and thus it does not depend on the *WL/Q*. In the advection-dominated case the lower pumping rate cause only a delay of the breakthrough.

- In the case of the equilibrium sorption we consider surface sorption. In that case the time of the peak discharge rate for pulse input is $t_{max} = t_w\ (1+ 2\ K_a\ /\ 2b)=$ $t_w + 2\ K_a\ t_w/2b = t_w + 2\ K_a\ WL/Q$. Peak discharge rate scales in the same manner. This means that retention depends linearly on the *WL/Q*. In this case a lower pumping rate cause delay that is proportional to the ratio of the pumping rates and the corresponding change in the discharge level.

- In the case of matrix diffusion it can be shown that for the pulse input the time of the peak output is $t_{max} = t_w + 2/3{\cdot}u^2$, where t_w is the advective delay. This means that the retention depends on the u^2 and $(WL/Q)^2$ (cf. equation (3-2)). A lower pumping rate then causes change both in the delay and discharge level that are proportional to the square of the ratio between the pumping rates.

If the transport is purely dominated by one of the processes above, then it may be possible to determine the process from a single test. In practice the flow channel may not be a well mixed volume. This means that there is dispersion (and longitudinal diffusion) in the flow channel that may conceal the ideal behaviour presented in the bullets above. In addition, all retention processes may have a significant effect on the transport simultaneously (superposition). Therefore variations of the pumping rate (i.e. variation of the *WL/Q*) and corresponding changes in the tracer discharge rate and delay can elucidate what is the principal retention process for a given situation.

The heterogeneity type that may affect the retention under a changed pumping rate is the case where the material properties change as a function of the depth in the immobile zone (type M3 in Figure 4-1). The limited capacity of the immobile pore space shows transition in the retention properties from the matrix diffusion type behaviour towards equilibrium type behaviour as the pumping rate is decreased. If the pumping rate is further decreased then the matrix diffusion to the next layer may become dominating. The chart in Figure 7-1 represents roughly the dominant retention process in the layered system. It should be noted that the breakthrough curve is always to some extent mixed from different type responses and that different parts of the breakthrough curve may show different behaviour.

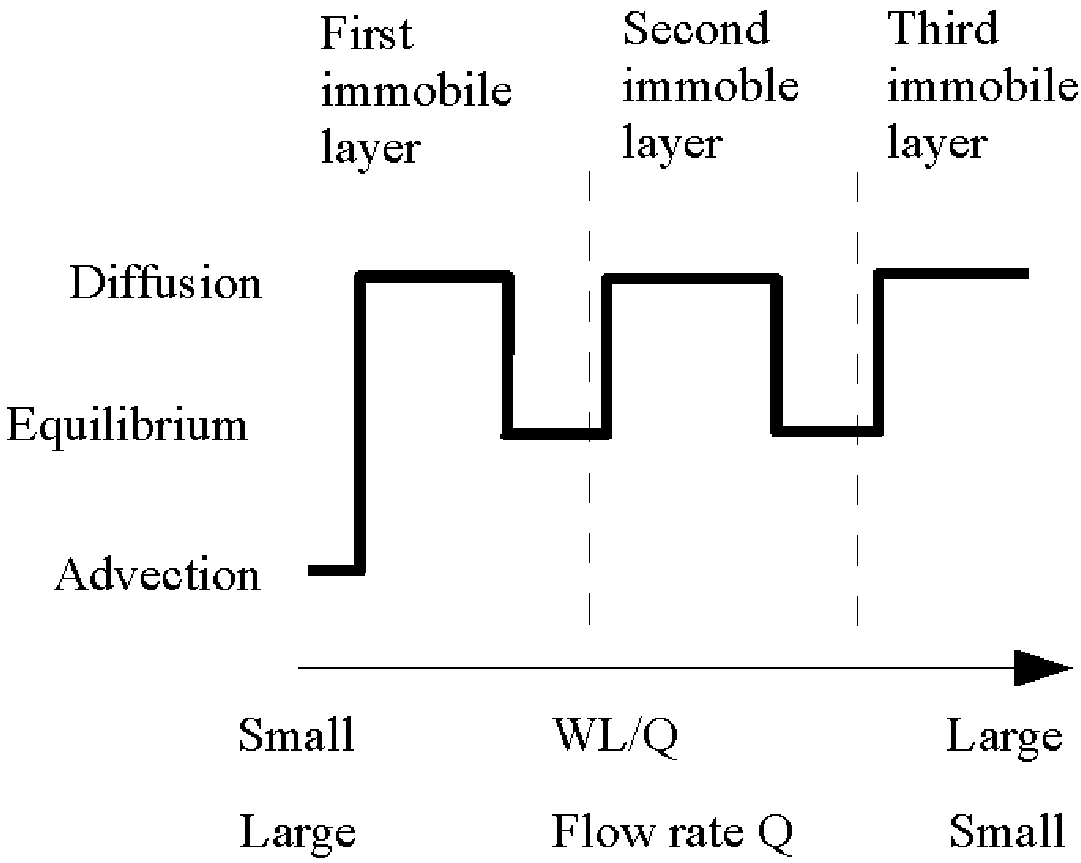


Figure 7-1. *A schematic illustration of the dominant retention or transport process for a non-sorbing tracer in the case of layered material properties in the immobile zone. Word diffusion on y-axis at left means diffusion-controlled retention (matrix diffusion) type behaviour and equilibrium means equilibrium sorption type behaviour.*

7.2 Tracer properties

Influence of the tracer properties on retention can be examined using the parameter u (equation (3-2)). It shows that in the case of diffusion controlled retention (for the infinite immobile zone) when the flow field is the same, the breakthrough curves of the different tracer should scale as $D_p \cdot R_p$ (retention time and level of the peak discharge are proportional to u^2). In the case of equilibrium sorption, like surface sorption, different tracers compare as R_a. This means that it is difficult to discriminate between these two retention processes using different sorbing tracers because K_d and K_a are usually correlated. Therefore, it may be of interest to look for tracers that have variable pore diffusivity because that does not affect the equilibrium sorption. Nano-particles may offer one possibility to have a non-sorbing tracer with a markedly different diffusivity. Below an example is given on how the diffusivity may affect the transport.

In a heterogeneous medium the capacities of the immobile zones may be limited (heterogeneity type M3 in (Figure 4-1). The saturation of the limited immobile zone may manifest itself in the breakthrough curve as a spurious equilibrium sorption even for the non-sorbing tracers. If there is a high porosity immobile zone available then the residence time of non-sorbing tracer does not give the groundwater residence time but a somewhat longer residence time. This is illustrated in Figure 7-2 where breakthrough curves of two different tracers are shown. In the calculated case there is a 0.2 mm layer of porosity 0.05 next to the flow channel. The groundwater transit time is 200 hours and channel aperture is 0.5 mm. Both tracers are non-sorbing and only the pore diffusivity varies. Pore diffusivities are $2 \cdot 10^{-11}$ m^2/s for the blue curve (i.e. geometric factor $G=F/\varepsilon=0.01$, where F is formation factor and ε porosity) of and 10^{-13} m^2/s for the green curve (representing e.g. nano-particles). In this case the tracer represented by the blue curve saturates the immobile zone and the brekthrough curve shows retarded equilibrium sorption type breakthrough. The tracer that is represented by the green curve still shows the groundwater transit time, 200 hours, although signs of diffusion to the immobile pore space are also visible.

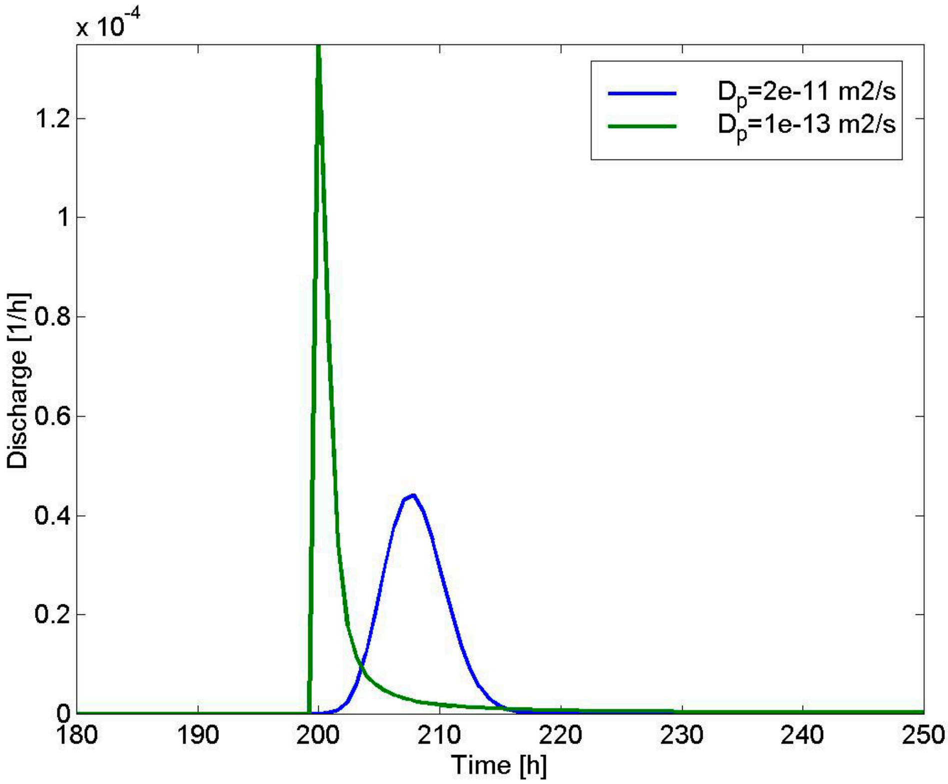


Figure 7-2. *Calculated breakthrough curves for the two non-sorbing tracers in case that the groundwater transit time is 200 h, fracture aperture is 0.5 mm and there is a layer of immobile zone that have porosity of 0.05 and thickness of 0.2 mm. The two tracers have different pore diffusivities in the immobile zone as indicated in the figure.*

The discussions above handled mainly the case of homogeneous immobile zone properties. The difficulties that are connected to the evaluation of heterogeneity from the tracer test results are clarified by the following example. Let us assume that there are two different immobile pore spaces along the path. Both of these immobile pore spaces are infinite and the retention is diffusion-controlled. Different tracers will show breakthrough curves that reflect the average properties along the transport path, i.e.

$$u_{eff} = \frac{WL}{Q}\left(\bar{\varepsilon}_1\sqrt{D_{p1}R_{p1}} + \bar{\varepsilon}_2\sqrt{D_{p2}R_{p2}}\right), \quad (7\text{-}1)$$

where $\bar{\varepsilon}_1$ and $\bar{\varepsilon}_2$ are effective porosities of the different materials as shown in equations (5-2) and (5-3), e.g. if the porosity is 0.01 and that material exist over 10% of the total pore space along the flow path then the effective porosity of this material is 0.001. Equation (7-1) demonstrates that if the tracer-dependent parameters are not known then it is not possible to estimate the effective porosities using tracer test data because in that case every new tracer only increases the number of unknowns. Equation (7-1) also shows that the number of unknown parameters increases markedly when there are different kinds of materials involved, i.e. the more heterogeneous the flow path is.

At the beginning of this section it was stated for the homogeneous case that the time and level of the tracer peak discharge rate for different tracers compare as the product $D_p \cdot R_p$ of the tracers, if the tracers are transported in the same flow field (the same *WL/Q*) and diffusion-controlled retention is the dominant retention process. Equation (7-1) shows that if several different immobile pore spaces are available then the different tracers compare as $\left(\sum_{i=1}^{N} \bar{\varepsilon}_i \sqrt{D_{pi} R_{pi}}\right)^2$, i.e. the square roots of the $D_p \cdot R_p$ weighted by the effective porosities of the different materials. Different tracers may give different "views" of the material properties if the sorption properties of the different materials vary considerably among the tracers.

The limited capacity of the pore space (type M3 heterogeneity) adds a new aspect to the question as to how the tracer properties, heterogeneity and tracer breakthrough curve are connected. What can happen in the case of limited capacity pore space is that some of the pore spaces may get saturated. This means that the corresponding term in the diffusion-controlled retention (terms in the parentheses of the equation (7-1)) dissappears and appears as an enhancement in the equilibrium type retention.

7.3 BS2B Tracer tests

Examination of the tracer properties and heterogeneity suggest that it may be beneficial to apply non-sorbing tracers that have different pore diffusivities. This kind of tracers may for example be nano-particles. The diffusivity of the nano-particles depends on the size and material of the particles. Together with traditional tracers e.g. dyes it may be possible to have a set of non-sorbing tracers that span a wide range of different diffusivities. In principal, using sorbing tracers would yield similar results but non-sorbing tracers have the advantage that they do not have surface sorption. This means that possible differences in the breakthrough times can be directly connected to the diffusion-controlled retention (c.f. Figure 7-2). There may also be difficulties that are connected to the different diffusivities. For example, low diffusivity tracers may have more dispersion that is caused by the variable advection because the diffusional mixing is weaker. Large differences in the dispersion between the different tracers complicate the integrated interpretation of the results.

Different retention processes cause clearly different responses in the breakthrough curve if the flow rate along the flow path is changed (see Section 7.1). Different flow rates along the flow path can be achieved by changing pumping rates, although in the in-situ situation the different pumping rate may not only mean that the flow rate but also the flow path itself is different. However, the discriminating power of the different flow rates on the retention processes is very strong. Especially, it may be used to identify heterogeneity that is connected to the limited capacity of the pore space (type M3 heterogeneity). Saturation of the pore space that has limited capacity will change the retention behaviour from diffusion-controlled retention to equilibrium sorption type behaviour. These two processes behave very differently under changed flow conditions (see Section 7.1).

8 Summary

This study has examined the effects of different kinds of heterogeneities on the retention seen in in-situ tracer experiments. The approach has been to identify a set of simple type heterogeneities. The type heterogeneities include variations in both material and the flow field properties. It is shown that more complicated heterogeneities can be constructed using the type heterogeneities in Figure 4-1. It is also shown that in the longitudinal and lateral type of heterogeneities the retention does not depend on the spatial distribution of the different properties but only on the "effective quantity" of the different properties if well-mixed conditions can be assumed to prevail in the flow channel. This holds both for the material and flow field properties if they vary independently. More precisely, the retention depends on the summed up material properties that are weighted by the flow field properties. This means that possible correlation between the flow field and a given material property will affect the effective retention properties. Changing material properties as a function of the depth into the immobile zone of the wall rock may cause large differences in the retention if the flow rates are varied.

The averaging nature of the retention along the flow path makes it very difficult to study the heterogeneity using in-situ tracer tests. Retention depends on a grouped parameter and determination of the individual parameters requires results from several tracers with different properties. Especially useful could be use of non-sorbing tracers with different pore diffusivities. That kind of tracers could for example be nano-particles if their diffusion properties can be determined reliably enough. Non-sorbing tracers can give information especially on the diffusion-controlled retention and possible heterogeneity that is connected to the limited capacity of the immobile pore spaces.

Different pumping rates can also help to reveal the heterogeneity that is connected to the limited capacity of the immobile pore space. Saturation of the immobile pore space will change the characteristics of the retention. Diffusion-controlled retention is very sensitive to the flow rate, so that decreasing the flow rate will affect the tracer discharge very clearly. If the immobile pore space gets saturated when the flow rates are decreased this should manifest itself clearly in the breakthrough curve.

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