

**Flow and transport through  
a damaged buffer – exploration  
of the impact of a cemented and  
an eroded buffer**

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December 2006

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*Keywords:* Erosion, Cemented buffer, Flow, Solute transport.

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## **Abstract**

The bentonite buffer around the copper canister protects the canister from mechanical impact, corrosion and, in the case of canister damage, exerts an important barrier to the release of radionuclides. Over time the properties of the buffer might change. Should the buffer be cemented, become brittle and develop fractures its solute transport resistance would change and probably decrease. Should the buffer be eroded due to dispersion of its fine clay particles into the water seeping past in the fractures in the rock its hydraulic conductivity and solute transport resistance will decrease. This short report explores the consequences to canister corrosion and nuclide transport of a hypothetical cementation and cracking of the buffer and of loss of buffer by erosion. Neither the causes of cementation nor of erosion are addressed in this report.

## Förord

Bentonitbufferten som omger kopparkapseln skyddar den från mekanisk påverkan, korrosion och, skulle kapseln vara skadad utgör bufferten en viktig barriär mot uttransport av radionuklider till omgivningen. Med tiden kan buffertens egenskaper ändras. Skulle den cementeras, bli spröd och få sprickor skulle sannolikt dess förmåga att minska transport av korrosiva ämnen till och radionuklider från kapsel försämrats. Skulle bufferten erodera genom att dess små lerpartiklar förs bort av vattnet som sipprar i bergets sprickor kommer dess hydrauliska konduktivitet och dess motstånd mot transport av lösta ämnen att minska. I denna korta rapport utforskas konsekvenserna av att bufferten skulle spricka och av att den skulle erodera. Skälen till cementering och erosion behandlas inte i denna rapport.

## Summary

The compacted bentonite buffer that surrounds the canister with spent fuel is designed not to allow water to flow through it in any appreciable quantities. Should the buffer be severely damaged water could flow to the canister and bring with it corrosive agent such as sulphide. This could speed up the rate of copper corrosion. Should the canister have a damage or be corroded so that the spent fuel inside the canister comes in contact with the water radionuclides could dissolve in the water and be carried away.

Two modes of canister damage and their consequences for solute transport are explored in this report. In one case the buffer can potentially be eroded by the water seeping in the fractures in the surrounding rock and attain a high hydraulic conductivity allowing water to flow through it, at least locally. In another case it is conjectured that the buffer is cemented and becomes brittle. Then a crack could conceivably form in the cemented buffer allowing water to flow through it directly to the canister surface. Both cases are hypothetical although in the erosion case the erosion rate could possibly be quantified.

The cases studied are to be seen as exploratory calculations for “what if” cases. Some worst case assumptions have been used to explore bounding scenarios. Thus for the erosion case it is assumed that the hydraulic conductivity is so high that the buffer exerts negligible resistance to water flow. This implies that all the water that can flow through the fractures intersecting the deposition hole can reach the canister. Similarly for the cemented buffer case it is assumed that the crack in the buffer directly connects the rock fracture to the canister surface.

For these circumstances it is found that the flowrate that can enter the deposition hole will be approximately two times as large as the water flow that would pass a projected area equal to that of the deposition hole. Thus if the flux in the rock some distance from the deposition hole is 1 litre/m<sup>2</sup> and year the presence of a damaged deposition hole with a projected area of 16 m<sup>2</sup> would have a through flow of 2·16 litres per year.

There is however, a major difference for the solute transport to or from the canister if there is an intact low permeability buffer surrounding the canister. In that case the solutes have to first diffuse from the flowing water to the buffer. The solute has only the “short time” for diffusion for which the water flowing past the buffer is in contact with it. This sets a bound on the rate of solute transport. Furthermore the solute has to diffuse through the compacted buffer before reaching the canister surface. This adds to limit the rate of transport. When, however, the water can flow through the eroded buffer to the canister it has a long residence time in the deposition hole and can deposit all its corrosive agents to the canister. This can considerably increase the rate of transport to the canister surface compared to when the water flows around the deposition hole. For high flowrates in the rock fractures the corrosion rate could become much higher than for an undamaged buffer.

Scoping calculations suggest that even under unfavorable circumstances it would take a very long time (> 100,000 years) to erode the buffer sufficiently to allow water to flow through it.

For the cemented buffer case the same increased water flowrate as in the eroded buffer case will pass the canister. The water will supply the corrosive solute to a very thin region on the canister surface. The corrosion can therefore be localized and not spread out over a large surface as in the former case. This implies that for the same rate of transport of the corrosive agent the rate of corrosive penetration depth of the canister would be considerably larger.

However, the water that flows through the fracture in the cemented buffer has a much shorter contact time with the canister than in the erosion case when a large part of the volume of the deposition hole could accommodate the same flowrate. The short contact time of the water with the canister allows only a small fraction of the solute in the water react with the copper canister.

This strongly limits the rate of corrosion. Furthermore the corrosive attack can be expected to spread out in a semi circle into the copper wall and not be limited only to the width of the fracture in the buffer. These effects together imply that although the rate of localized corrosion is larger than for an intact buffer it still would take hundreds of thousands of years for the highest considered flowrate in this note to penetrate the canister.

The release rate of radionuclides from a damaged canister is strongly limited by their rate of diffusion through a hole in the canister. For holes in the canister of up to ten cm in diameter the radionuclide release is limited by the diffusion resistance in the hole. The damages in the buffer causing high flowrates in the deposition hole will not give an increase in nuclide release as long as the hole in the canister is not exceedingly large and provided there is no flow through the canister.

It is also suggested that if in an earthquake scenario the canister is sheared but the buffer is still intact prohibiting water flow through the deposition hole, the diffusion resistance for nuclides in the remaining buffer strongly limits the rate of release of nuclides.

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# 1 Introduction

The buffer surrounding the canisters with spent fuel swells strongly and has a very low hydraulic conductivity. If damaged its barrier function could suffer. Two kinds of damage are explored, erosion and fracturing. In the erosion case the buffer could be eroded by high shear forces exerted by the flowing water on the clay gel. Erosion can also be caused by chemical erosion. Then the passing groundwater, if of low ionic strength, can take up and carry away cations, primarily calcium, from the clay, which in turn transforms the gel to a sol and releases the clay particles into the passing water. When sufficient clay has been lost, the remaining clay, at least locally, can have a high hydraulic conductivity allowing water to flow through it. Then the water can come into direct contact with the copper canister, speed up the corrosion of the canister and carry away any leaking nuclides. The rate of transport to and from the canister can increase compared to when solute transport is limited by diffusion in the buffer and in the seeping water in the fracture.

Water that enters the buffer from the upstream side of a fracture will spread out in the eroded buffer both upward and downward before it again leaves at the downstream side. If the hydraulic conductivity of the eroded buffer is higher than the surrounding rock it may allow water to be drawn in from the flowing fracture.

The water in the porous buffer will have a longer residence time in contact with the canister and may therefore have time to deposit or take up solutes from the canister. Thus the eroded buffer can increase the supply of corrosive agents and nuclide release due to larger water flowrate to the canister and due to longer residence time and larger contact area with the canister, increasing the flowrate.

In the “fracture” case the buffer is assumed to be cemented due to chemical dissolution/precipitation reactions caused by e.g. heating or because the system is not in equilibrium with the groundwater. Whatever the cause, cracks form in the now brittle buffer. It is assumed that a fracture forms which is connected to the water conducting fracture in the rock. Water can then flow directly to the canister short circuiting the diffusion resistances.

We will first estimate the increase in water flowrate to the canister and then assess the increase of corrosive agent and nuclides transfer rate to and from the water passing through the deposition hole.



## 2 Aims and scope

The aim of this note is to make a first exploratory assessment of the possible consequences for nuclide release from the near field if and when the buffer surrounding the canister is damaged. Two kinds of damage are explored. In the “erosion” case it is assumed that the buffer has been partly eroded allowing water to flow through the buffer. In the “fracture” case it is assumed that the buffer has been cemented and that a fracture has formed through the buffer allowing the water to flow through the fracture in to and past the canister. Some very simple approaches are explored with the hope that the results will point to what needs to be investigated in depth later, should the exploratory scoping calculations suggest that this needs to be done.

### **3 Increase of water flowrate to the canister from the fracture in the rock**

Two kinds of damage are explored, the erosion case and the fracture case. In the erosion case the buffer could be eroded by high shear forces exerted by the flowing water on the clay gel. Erosion can also be caused by chemical erosion. Then the passing groundwater, if of low ionic strength, can take up and carry away cations, primarily calcium, from the clay, which in turn transforms the gel to a sol and releases the clay particles into the passing water. When sufficient clay has been lost, the remaining clay, at least locally, can have a high hydraulic conductivity allowing water to flow through it. Then the water can come into direct contact with the copper canister, speed up the corrosion of the canister and carry away any leaking nuclides. The rate of transport to and from the canister can increase compared to when solute transport is limited by diffusion in the buffer and in the seeping water in the fracture.

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We will first estimate the increase in water flowrate to the canister and then assess the increase of corrosive agent and nuclides transfer rate to and from the water passing through the deposition hole.

#### **3.1 Single fracture through a brittle buffer**

This case is treated first because it can be modeled as a two-dimensional flow through a single fracture. The following very simple model is used. Water flows in a fracture with constant hydraulic properties. Far away the streamlines are parallel but as the flow approaches the deposition hole it is drawn into the highly conductive fracture in the buffer. In the limit the fracture in the buffer is taken to be much more transmissive than the fracture in the rock. The fracture in the buffer will seem to have no flow resistance. Then the maximum flowrate of water that can flow through buffer will increase by about a factor of two due to the damage, compared to the water flowrate through the same width of fracture far away. This can be shown by numerical simulations as well as by an analytical solution to the flow equations. The latter is shown below.

For an idealized 2D case with a circular hole with infinite conductivity an analytical solution can be obtained for the flow distribution in the fracture from potential flow theory. /Bird et al. 2002 p 128 ff/ present a solution for the opposite problem for flow around an impervious cylinder. The same solution is valid for flow through an empty cylinder using the real part of the complex potential. The streamlines around and through an empty circle can be described by

$$\Psi(x, y) = -y\left(1 + \frac{1}{x^2 + y^2}\right) \tag{1}$$

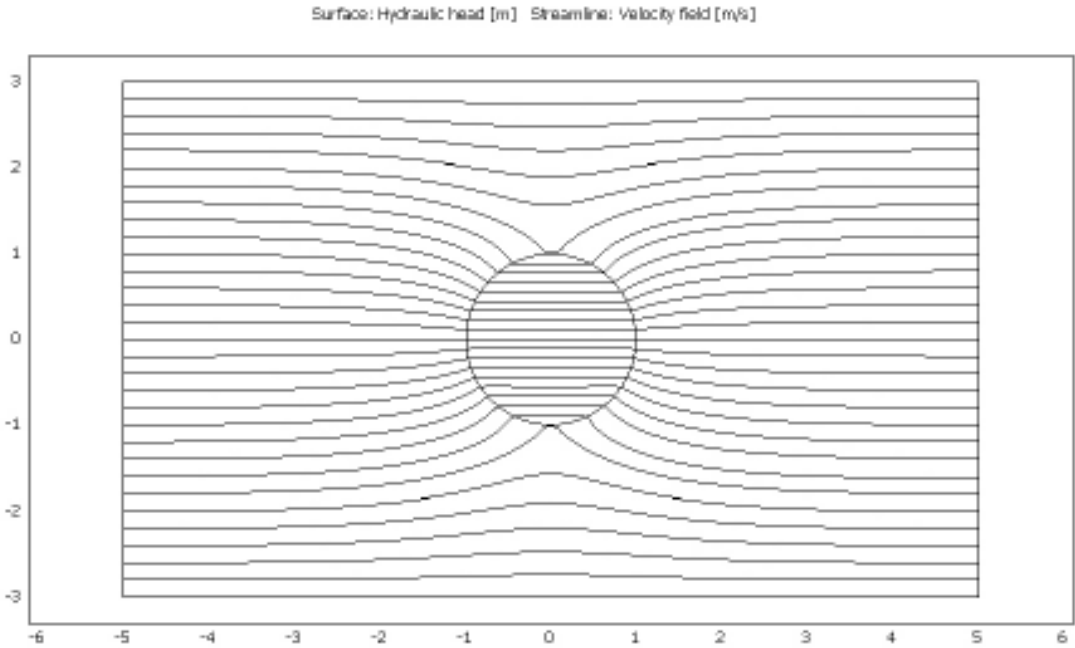
This is shown in Figure 3-1 where the streamlines in the fracture are seen to enter the empty circular hole. The circle has a unit radius and it is seen that streamlines from nearly 2 diameters flow through the circle. Had the figure been made longer in the x direction exactly two diameter streamlines would have entered the circle.

Thus in a fracture that intersects the deposition hole at right angles the capture width is

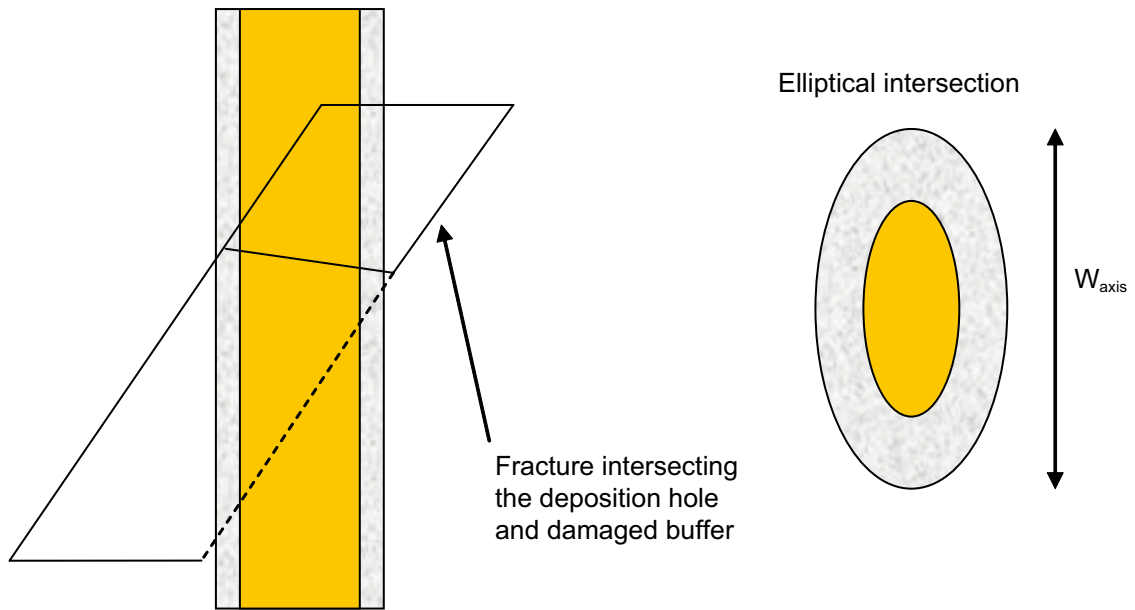
$$W_{capture} = 2d_{hole} \tag{2}$$

For other intersection angles the capture width can be larger. A fair approximation is that the capture width is twice the largest length of intersection. For an inclined fracture it would be twice the long axis  $W_{axis}$  of the ellipse formed by the intersection of the fracture with the deposition hole. In the limit, when the fracture is parallel to the deposition hole the length could be the entire length of the hole.

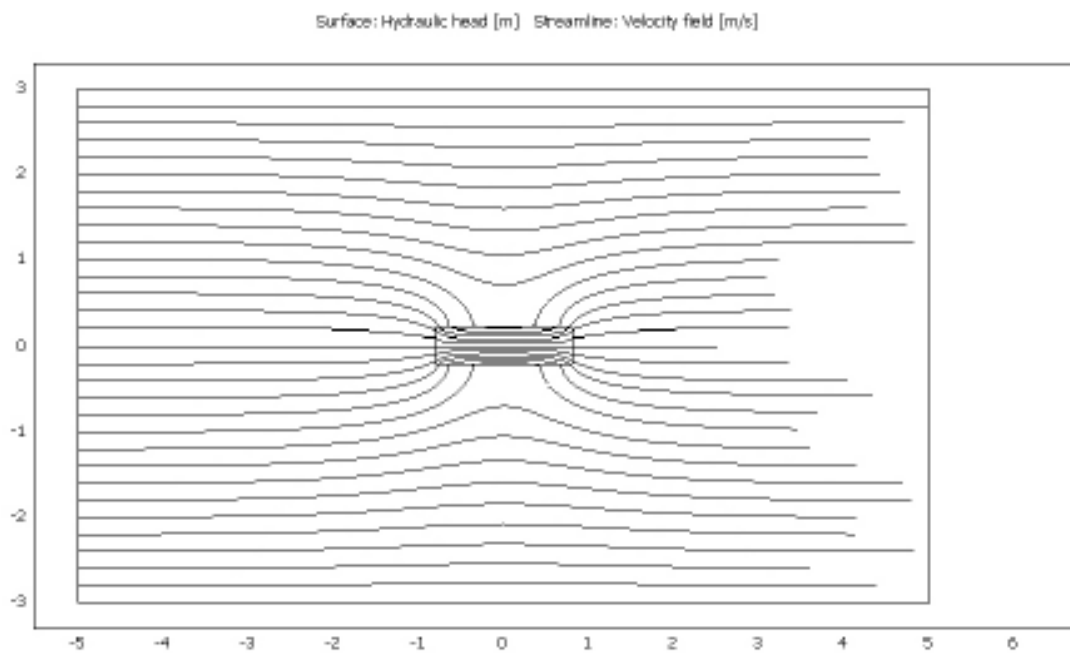
Figure 3-2 shows that fractures can intersect the deposition hole and the damaged zones at an angle. The projected area of the intersection with the damaged zone will vary with the angle. Also the largest length of the projected area will vary with the intersection area.



**Figure 3-1.** Streamlines drawn into a high conductivity circular zone.



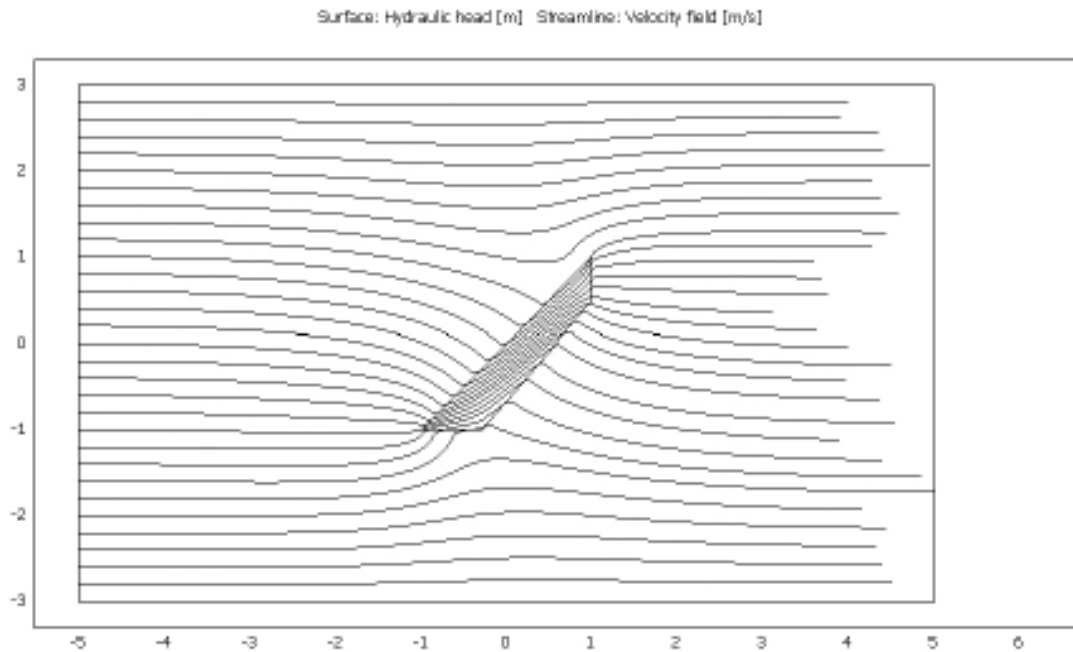
**Figure 3-2.** Fracture intersecting the deposition hole at an angle.



**Figure 3-3.** View in the plane of the fracture. Streamlines drawn into a fracture that intersects the deposition hole along its length.

Figure 3-4 illustrates the streamlines converging into an arbitrarily shaped high conductivity region.

It has not been deemed worthwhile at this stage to account in detail for the variation of flowrate with the intersection angle because of the inherent uncertainty of the shape, extent and properties of the damaged zone. Instead two extremes are studied. In the first case the fracture intersects the hole at right angles. In the second it is parallel to the hole and intersects the deposition hole along its entire length.



**Figure 3-4.** Streamlines drawn into a high conductivity arbitrarily shaped zone.

The case with the vertical fracture and where the hydraulic gradient is near vertical is not expected to be very common but is probably an extreme case with the highest flowrates and the largest exposed area for mass transfer between water and canister.

A vertical fracture that intersects the damaged buffer around the deposition hole will have a capture width equal to twice the length of the hole which is about 8 metres. Another angle of intersection will have a length which depends on the locus of intersection, the angle and the shape of the zone.

The length of intersection can thus vary between  $d_{hole}$  and  $L_{hole}$  i.e. about 1.75 m to 8 m. The capture width  $W_{capture}$  will then vary between  $2d_{hole}$  and  $2L_{hole}$  which is about 3.5 m and 16 m. The flowrate through the zone will be

$$q = T \cdot i \cdot W_{capture} \quad (3)$$

As an example for  $T = 10^{-8} \text{ m}^2/\text{s}$  and a gradient  $i$  of 0.01 the flowrate is between  $3.5 \cdot 10^{-10}$  and  $16 \cdot 10^{-10} \text{ m}^3/\text{s}$  which is about 11 to 50 litres per year. Given enough residence time at the surface of the canister all this water could exchange solutes with the canister. A  $T = 10^{-6} \text{ m}^2/\text{s}$  the flowrate will be hundred times larger, 1.1 to 5  $\text{m}^3/\text{year}$ .

### 3.2 Flow trough deposition hole with an eroded buffer

The same analysis and results apply to the case when the eroded buffer has a sufficiently high hydraulic conductivity. Then water from the fracture in the rock will encounter an “empty” hole. The same flowrates as above will result. The difference is that the water can spread out over a large empty volume in the hole whereas in the fracture case it only has access to the volume of the fracture in the cemented buffer.

It will be shown that this strongly affects the rate of exchange of solutes between canister and flowing water.

## 4 Determination of the equivalent flowrate for radionuclide uptake by the water and reaction with corrosive agents with canister

In this section we explore how much more solute exchange between flowing water and canister could take place due to the potentially large increase in water flowrate.

### 4.1 Fracture case

The details of the number of fractures, their aperture and the resulting porosity and diffusion properties are not known. A worst case assumption would be that all the flowing water would be equilibrated. However, this may be too pessimistic and we estimate the equivalent flowrate for solute transfer to and from the canister. Such a model has been presented earlier for flow in a fracture in contact with the buffer using diffusion theory /Neretnieks 1979/.

From diffusion theory we have that the mean penetration depth of the solute into the water can be determined by integrating the concentration profile from the surface ( $z = 0$ ) to infinity /Bird et al. 2002 p 621/. The concentration profile in the water is obtained from

$$\frac{c}{c_{eq}} = \text{Erfc}\left(\frac{z}{\sqrt{4D_w t_{res}}}\right) \quad (4)$$

The water residence time in the fracture in the backfill is the residence time of the water in contact with the canister

$$t_{res} = \frac{\pi r_{can}}{u} \quad (5)$$

The mean penetration depth of the solute into the water is then obtained from

$$\eta_{mean} = 1.13\sqrt{D_w t_{res}} \quad (6)$$

This is the mean distance into the flowing water that can exchange solute with the canister during the time the water is in contact with the canister.

The expression is valid when it is about 4 times smaller than the distance from the canister surface out to the outer radius of the deposition hole (0.35 m). Otherwise it is approximately equal to this distance and all the flowrate through the fracture in the buffer will be equilibrated. This simplification is deemed acceptable not to complicate the procedure unduly and because there are many uncertainties regarding the geometry and the properties of the zone.

**Table 4-1. Data for the fracture case.**

Entity	Meaning	value
$r_{hole}$	Radius of deposition hole	0.875 m
$r_{can}$	Radius of canister	0.525 m
$D_w$	Diffusivity in water	$10^{-9}$ m <sup>2</sup> /s
$i$	Hydraulic gradient	0.01
$D_{buffer}$	Diffusivity in buffer	$1.2 \cdot 10^{-10}$ m <sup>2</sup> /s

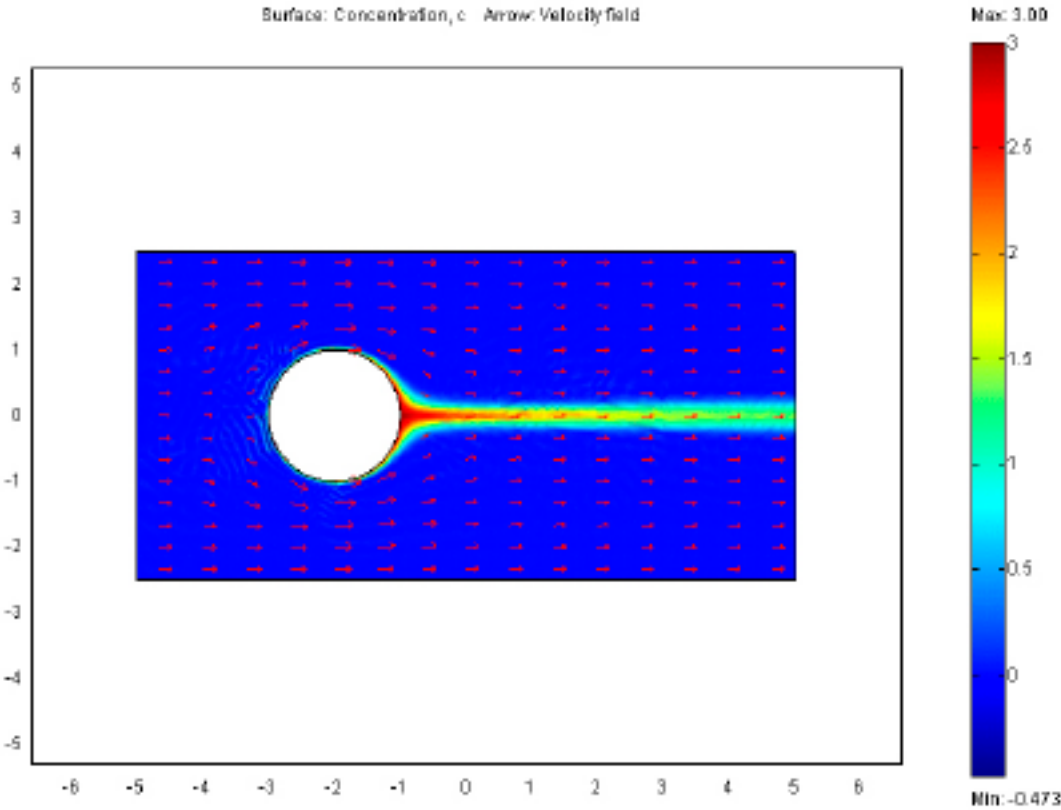
Figure 4-1 illustrates a case where a solute diffuses into the water passing around a leaking canister. The solute has time to diffuse out a short distance into the water while the water is in contact with the canister. The width of lighter colored region downstream (to the right of) the canister illustrates what is meant by penetration depth, one from each side of the canister. The water flowrate in this width has taken up the solute. This is called the equivalent flowrate. The same applies to the case when a solute is transported from the water to the canister.

The equivalent flowrate  $Q_{eq}$  (See Appendix for explanation of the term “equivalent flowrate”) can then be estimated to be

$$Q_{eq} = u \cdot 2b \cdot 2 \cdot \eta_{mean} \tag{7}$$

This expression can be used for transport to and from the canister surface as well as for the case when the buffer is undamaged and water flows around the buffer. The velocities and residence times will be different in the two cases.

If the penetration depth approaches the above mentioned 0.35 m all the water flowrate  $q$  will exchange all its solute with the canister and  $Q_{eqFB} = q$ . It should be noted that in this case one must also consider the diffusion into the water flowing in the *fracture in the rock*. This is done in a similar way but is not elaborated more here as it adds little to the equivalent flowrate. Then  $q$  and  $Q_{eqGB}$  for the rock fracture are to be added. FB stands for Fractured Buffer and GB stands for Good Buffer.



**Figure 4-1.** Groundwater flows in the intersecting fracture around the canister. A solute is released from the canister and diffuses into the groundwater. The concentration is shown by the colour surface plot and the groundwater velocity by the arrow plot.

Due to the higher flowrate in the fracture in the buffer and because some of the volume of the deposition hole is taken up by the canister there will be an increase in water velocity and a decrease in residence time compared to when the buffer is intact. The difference is about a factor of 2 for the velocity and less for the residence time because the canister has a smaller circumference than the deposition hole. This leads to an increase of  $Q_{eqFB}$  by about 1.4 over  $Q_{eqGB}$  in the sample calculations.

In Table 4-2 some examples of equivalent flowrates and penetration depths are shown for different rock fracture transmissivities and apertures.

It can be seen from Table 4-2 that the  $Q_{eqFB}$  is about 40% larger than  $Q_{eqGB}$  when there is a fracture in the cemented buffer and if all the solute is only transported by advection directly to the canister. This is for the case when the fracture intersects the hole perpendicularly. A vertical fracture that intersects the deposition hole would have approximately 2 times higher  $Q_{eq}$ 's.

Note that in this comparison no account has been taken for the diffusion resistance in the buffer itself for the good buffer case. Accounting for this would decrease the transport rate for the good buffer case somewhat. The effects of diffusion resistance in the buffer will be discussed later. The overall difference between the fractured buffer and the good buffer will therefore be slightly larger than what Table 4-2 shows. However, it is evident that by far not all the flowrate  $q$  which flows past the canister will contribute to the solute transfer to or from the canister. In the range of  $T$ 's in Table 4-2 a 1,000 fold increase in flowrate will only result in a 30 fold increase in solute transfer.

The supply of corrosive agent to the canister will be concentrated to a narrow region where the fracture intersects the canister. The corrosion can then be much more localized than when the solute diffuses through the buffer and spreads out over a large area on the canister surface. However the corrosive agent flowing in the fracture could spread by diffusion in the cemented buffer so that not all the agent comes into contact with the canister just at the fracture intersection. This is studied below.

## 4.2 2D diffusion in the cemented backfill of a corrosive agent

Below we explore whether diffusion through the cemented buffer could spread a corrosive agent over a larger area than just that of the fracture aperture. The corrosive agent flows with the water toward the canister but also diffuses from the water into the porous cemented matrix. There it can diffuse toward the copper surface. In this way not all the corrosion will be at the fracture intersection with the canister. The spreading has been estimated for a simplified 2D case.

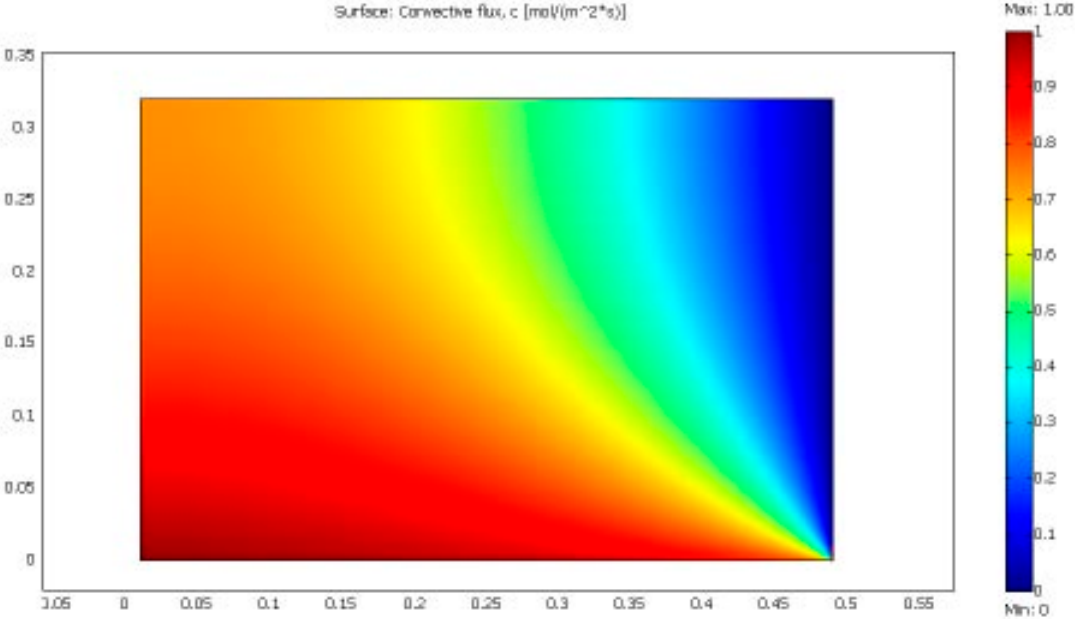
**Table 4-2. Penetration depth, residence time, Equivalent flowrate  $Q_{eqFB}$  and  $Q_{eqGB}$ . Fracture intersects the deposition hole perpendicularly. The hydraulic gradient  $i = 0.01$  m/m. Fracture aperture  $2b$  applies for both rock and buffer.**

$T$ m <sup>2</sup> /s	$q$ l/year	$2b = 1$ mm		$2b = 0.1$ mm			
		$Q_{eqGB}$ l/year	$\eta_{mean}$ m	$Q_{eqFB}$ l/year	$Q_{eqGB}$ l/year	$\eta_{mean}$ m	$Q_{eqFB}$ l/year
$10^{-9}$	1.1	0.13	0.23	0.18	0.042	0.075	0.057
$10^{-8}$	11	0.41	0.074	0.57	0.13	0.024	0.18
$10^{-7}$	110	1.31	0.023	1.8	0.42	0.0075	0.57
$10^{-6}$	1,102	4.16	0.0074	5.7	1.31	0.0024	1.8

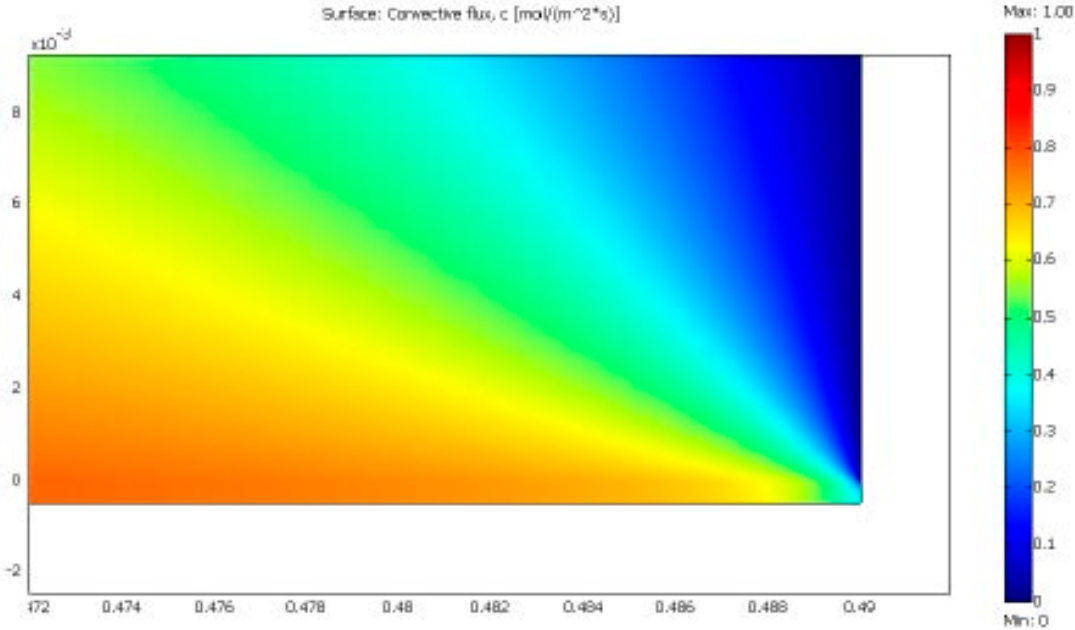


Figure 4-2 shows the concentration profile in the fracture and the buffer extending from 0 to 0.5 in the x-direction (flow direction) and from 0 to 0.3 in the y-direction (up along the copper surface at the right end of the figure). Water flows from left to right in the fracture located at the bottom of the figure. The fracture is too thin to be seen in this magnification. An enlargement of the right hand bottom corner is shown in Figure 4-3.

The inlet concentration of corrosive agent in the fracture is 1 mol/m<sup>3</sup> (arbitrarily chosen). The flowrate in the half fracture is 55 litres per year, valid for a transmissivity  $T = 10^{-7}$  m<sup>2</sup>/s. In the model it is assumed that all the corrosive agent is immediately consumed at the copper surface.



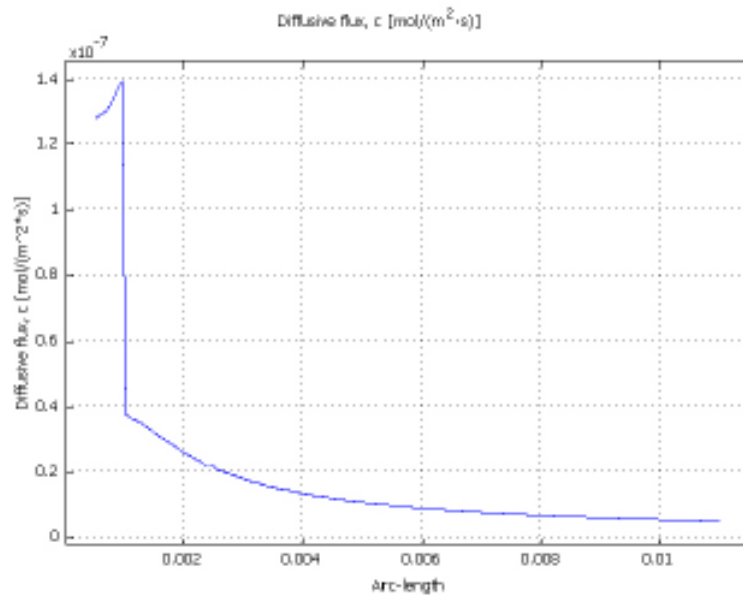
**Figure 4-2.** Concentration distribution along the fracture (x-direction). Copper is at vertical right hand boundary. Diffusion takes place from fracture into the buffer and further to the copper which acts as a sink.



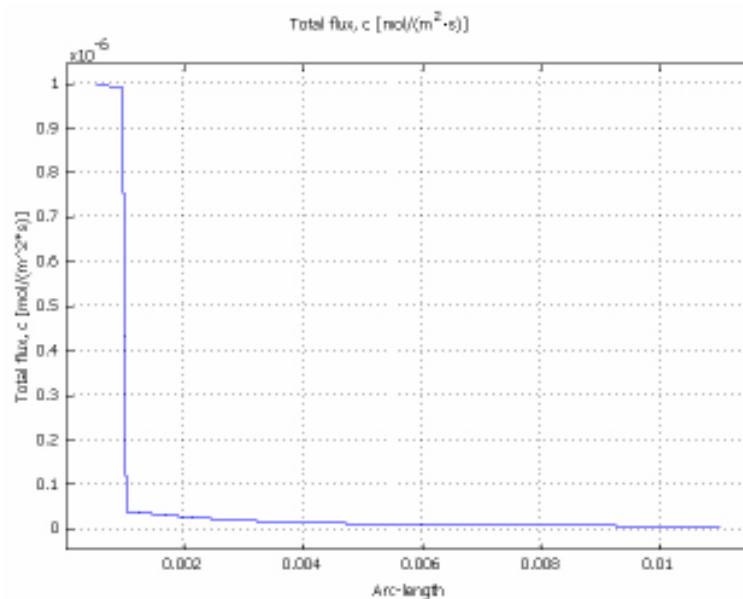
**Figure 4-3.** Enlargement of the bottom-right corner of Figure 4-2.

Figure 4-4 shows the diffusive flux to the copper canister in the 1 mm aperture fracture (only half is shown) from x-coordinate 0.0005 to 0.001 m as well as through the buffer (from coordinate 0.001 m to 0.011 m). The diffusive flux is much smaller than the advective flux in the fracture in the buffer as can be seen from the figure depicting the total flux in the same region shown in Figure 4-5.

It is seen that by far most of the corrosive agent will reach the canister by advection in the fracture. This will deliver the corrosive agent to a very narrow slit on the canister and cause a very local corrosion. For low transmissivities the corrosion will be somewhat more spread out adjacent to the fracture in the buffer due to diffusion in the cemented buffer.



**Figure 4-4.** Diffusive flux to canister surface from flowing water at fracture intersection (leftmost high peak) and through the buffer (to the right of the peak).



**Figure 4-5.** Total (diffusive plus advective) flux to canister surface from flowing water at fracture intersection (leftmost high peak) and through the buffer (to the right of the peak).

## 5 Erosion of buffer

In this section some calculations are presented on the rate of chemical erosion of the buffer for different transmissivities of the fracture intersecting the deposition hole.

A first attempt is made to estimate how much buffer must be eroded before its hydraulic conductivity becomes so low that it will permit water to flow through it.

The erosion of the buffer has been described and modeled by /Liu and Neretnieks 2006/. They find that provided the concentration of dissolved salts in the water is higher than the Critical Coagulation Concentration, CCC (about 1 mmol/l of divalent ions such as  $\text{Ca}^{2+}$ ) it is highly unlikely that the water seeping in the fractures will physically erode the buffer. The cohesive forces between the clay particles are strong enough to overcome the shear force of the seeping water.

However, at lower concentrations than the CCC the cohesive forces binding the clay particles together will become weaker and can even become repulsive in very low ionic strength waters. Then the particles could migrate out into the water and be swept away.

Some clays contain soluble calcium salts that replenish the pore water of the buffer to higher values than the CCC. This will take place as long as the salts have not dissolved and been carried away by the water. The rate of depletion of the salts is readily estimated from the product of the equivalent flowrate and the difference in concentration (solubility) of the dissolved salt in the porewater in the buffer and that in the approaching water.

The rate of depletion of the salt, taken to be a calcium compound, can then be written

$$N_{Ca} = Q_{eq} (c_0 - c_w) \quad (8)$$

where  $N_{Ca}$  ( $\text{mol s}^{-1}$ ) is the rate of loss of the calcium from the outer surface of the bentonite intrusion into the groundwater,  $Q_{eq}$  ( $\text{m}^3 \text{s}^{-1}$ ) is the equivalent flowrate, and  $c_0$  and  $c_w$  ( $\text{mol m}^{-3}$ ) are calcium concentrations at the outer surface of the buffer and in the groundwater, respectively.  $c_0$  can be taken as the CCC<sup>1</sup>. We assume that  $c_w$  is zero.

The rate of buffer erosion due to this cause is then directly obtained from the soluble salt concentration in the buffer

$$N_{clay} = N_{Ca} / X_{salt} \quad (9)$$

where  $X_{salt}$  is the concentration of salt in the buffer, mol salt/kg buffer.  $N_{clay}$  is the loss of clay kg/s. As an example we take the values used by /Liu and Neretnieks 2006/ 1.4% by weight of calcite gives  $X_{Clay} = 0.14$  mol calcite/kg clay<sup>2</sup>. With no calcium in the approaching water and  $c_o = 1$  mol/m<sup>3</sup>,  $N_{Clay} = Q_{eq}/0.14$ . With  $Q_{eq} = 10^{-3}$  m<sup>3</sup>/year (1 litre/year)

$$N_{Clay} = Q_{eq}/0.14 = 7 \text{ g/year}$$

In 1,000 years 7 kg clay will be lost.

Figure 5-2 shows average values for all the buffer volume. Should only a part of the buffer be eroded without the remainder swelling into the depleted regions the density locally could decrease much faster. Some of the consequences of local erosion are explored below.

<sup>1</sup> This is based on the model concept that when the pore water concentration is less than the CCC in the pore water in the buffer the particles solvate and are swept away by the passing water.

<sup>2</sup> Actually the solubility of calcite is less than the CCC. However, there are theoretical and experimental indications that the calcium in ion exchange positions influences (decreases) the CCC. This is presently being investigated.

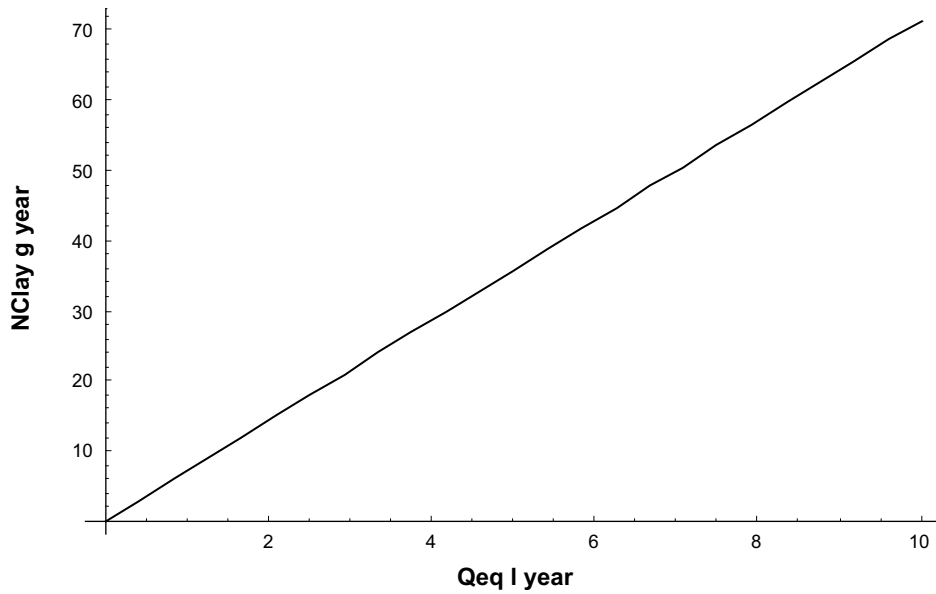


Figure 5-1. Loss of buffer as a function of the equivalent flowrate for conditions in Table 5-1.

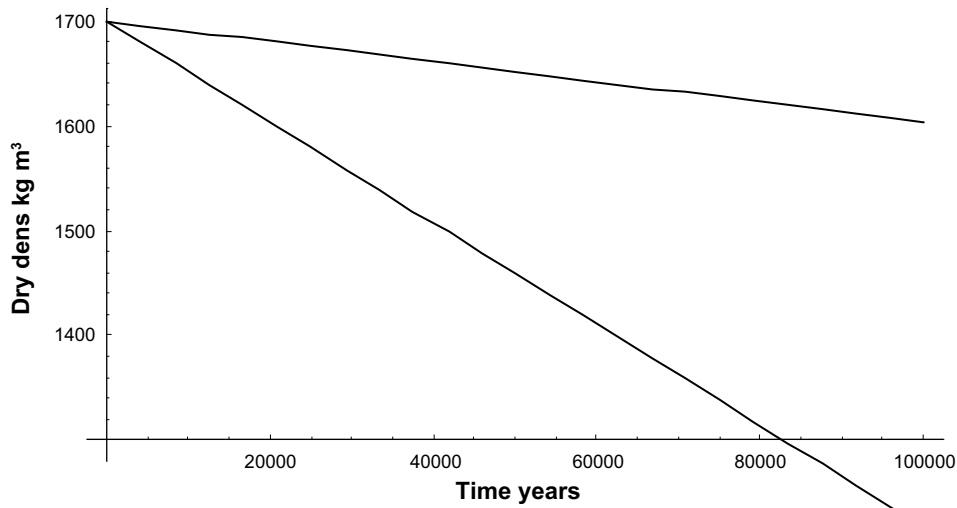


Figure 5-2. Loss of buffer density as a function of time for  $Q_{eq} = 1$  l/year (upper line) and 5 l/year (lower line) for conditions in Table 5-1.

**Table 5-1. Loss of clay from the buffer for different equivalent flowrates. Calcite content in the buffer is 1.4% by weight, CCC = 1 mol/m<sup>3</sup> and approaching groundwater has no calcium. Fracture aperture 1 mm.**

$T$ m <sup>2</sup> /s	$Q_{eqGB}$ l/year	$N_{clay}$ g/year
$10^{-9}$	0.13	0.93
$10^{-8}$	0.41	2.9
$10^{-7}$	1.31	9.3
$10^{-6}$	4.16	29

## 6 Transport through eroded buffer

When the buffer has eroded and the hydraulic conductivity has increased so that flow can take place through the buffer the water will spread out in three dimensions before again converging to flow out through the fracture on the downstream side. The water residence time will be much longer and more solute exchange between canister and water can take place.

The size and geometry of the eroded region are not known. Probably quite different damages could form depending on how re-swelling takes place and how and where the high conductivity channels in the fracture intersect the deposition hole. Therefore a simplifying assumption is used whereby the residence time of the water in the eroded region is assumed to be equal to the contact time with canister surface. This is probably a conservative assumption.

The flowrate of contaminated water  $Q_{eqEB}$  can then be directly determined from the mean penetration depth of the diffusing species. If the mean penetration depth is smaller than the thickness of the buffer only a fraction of the water is affected. This can be expressed as

$$Q_{eqEB} = q \frac{\eta_{mean}}{d_{buffer}} \quad (10)$$

EB stands for eroded buffer.

This is valid approximately when  $\eta_{mean} \leq \frac{d_{buffer}}{2}$ , otherwise  $Q_{eqEB} \cong q$ . In the latter case when all the water in the damaged zone is equilibrated. In addition, the water flowing in the fracture just outside the damaged zone will take up nuclides as they diffuse out to the water as it passes the deposition hole. The mean penetration depth can be obtained from the same equation as before [Equation (5)].

$t_{res}$  is now the residence time of the water drawn into the damaged zone  $V_{zone}/q$ . If the flow resistance due to high hydraulic conductivity in the damaged zone is much lower than the flow resistance in the fracture feeding it, it will act as an empty hole and let much water enter. See the description earlier Section 3.

For illustration purposes assume that 10% of the buffer volume is affected and is devoid of clay. The water volume is 0.77 m<sup>3</sup>. Using the same data as previously, we have:

It is seen in Table 6-1 that all the water will be depleted of a reactive solute in this example. Should the eroded zone be a tenth as large as assumed only the value of  $Q_{eq}$  in the last line would decrease marginally.

**Table 6-1. Equivalent flowrates for the eroded buffer case.**

$T$ m <sup>2</sup> /s	$q$ l/year	$t_{res}$ years	$\eta_{mean}$ m	$Q_{eqEB}$ l/year
10 <sup>-9</sup>	1.1	700	5	1.1
10 <sup>-8</sup>	11	70	1.7	11
10 <sup>-7</sup>	110	7	0.5	110
10 <sup>-6</sup>	1,100	0.7	0.17	1,100

Thus for a seriously eroded buffer all the water that flows into the deposition hole can exchange solutes with the canister and  $Q_{eqEB} = q$ .

Below we explore the flow in the eroded buffer to estimate and visualize how the water flows and how much of the canister surface could be in contact with mobile water. Also we explore to what hydraulic conductivity the eroded buffer must have degraded in order to allow considerable flow through it. Figure 6-1 illustrates how the water flow spreads out in the eroded buffer.

The two dimensional flow in the porous body can be described by a two-dimensional partial differential equation. This has been done and the solution for this case can be found in /Neretnieks 1986/. The solution was obtained for diffusion but it can be directly applied for flow as the governing equations are the same. It was shown that the whole resistance to transport in the porous body can be expressed in terms of the resistance in an (imaginary) short plug with the hydraulic properties of the porous medium. The plug has the thickness of the fracture aperture and the length of 3–8 times the aperture, depending on the geometry of the porous body. This, in the present case, is the geometry of the damaged zone. We chose a factor 5 for this example.

The water flow in the fracture is driven by a gradient  $i$ . The plug with hydraulic conductivity  $K$  has a transmissivity  $T_{plug} = K \cdot 2b$ . The gradient over the plug is larger by a factor  $r_{hole}/d_{plug} = r_{hole}/(5 \cdot 2b)$  over that in the fracture feeding it. This is because the pressure drop that would have taken place over a distance equal to the diameter of the deposition hole now will occur over the length of the two plugs. As the flow through the fracture and the plug is equal the following relation is obtained'

$$q = TiW = K2bWi \frac{r_{hole}}{5 \cdot 2b} = KWi \frac{r_{hole}}{5} \quad (11)$$

From the second and fourth term we obtain

$$K = \frac{5T}{r_{hole}} \quad (12)$$

Thus when  $K \gg 5T/r_{hole}$  the porous medium can be taken to have infinite conductivity for the purpose of the drawing in of water. When  $K = 5T/r_{hole}$  there will be no extra draw in of water and when  $K < 5T/r_{hole}$  the water in the fracture will flow around the buffer as if it is impervious.

For  $T = 10^{-8} \text{ m}^2/\text{s}$ ,  $K = 2.9 \cdot 10^{-8} \text{ m/s}$ . For  $T = 10^{-6} \text{ m}^2/\text{s}$ ,  $K = 2.9 \cdot 10^{-6} \text{ m/s}$ .

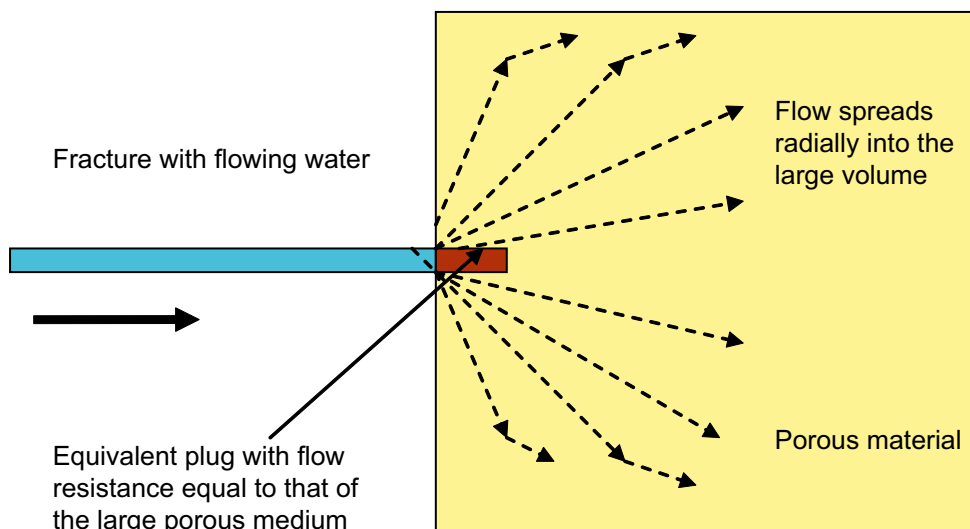


Figure 6-1. Water flowing in a fracture expands out into a large porous body.

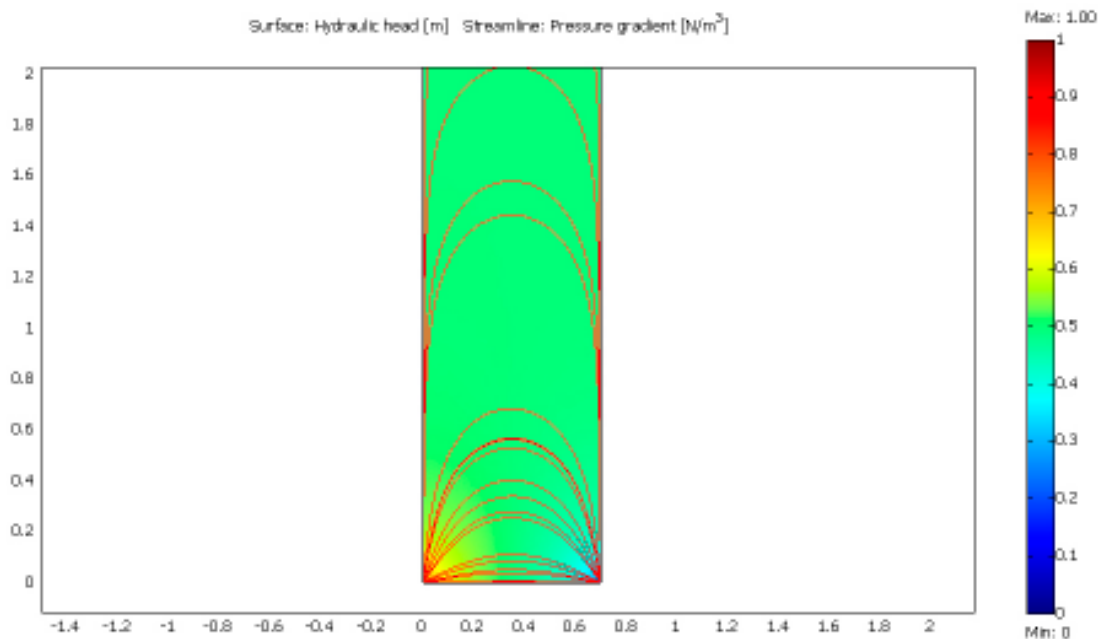
The buffer must be severely eroded to have a conductivity of  $2.9 \cdot 10^{-6}$  m/s. The dry density must decrease to well below  $500 \text{ kg/m}^3$ . Experiments show that for the commercial bentonite MX-80 the hydraulic conductivity at a density of  $500 \text{ kg/m}^3$  still is as low as  $10^{-11}$  m/s and for Deponit CA-N it is  $10^{-9}$  m/s. No measurements are reported for lower densities but extrapolation suggests that the density would have to be below a hundred  $\text{kg/m}^3$  for the conductivity to increase by one order of magnitude. Most of the buffer must have disappeared in the eroded region. It is therefore probable that the hydraulic conductivity of the bentonite will be high enough to prevent water flow through it while it still can be thought of as a swelling bentonite buffer. The swelling pressure at a density of  $500 \text{ kg/m}^3$  is 0.01 to 0.1 MPa for MX-80 and 0.01 for Deponit CA-N. This is about three orders of magnitude down from the design value for the buffer /Karnland et al. 2006/. At low swelling pressures physical erosion by shear forces from the water flowing in the fractures may have to be considered in addition to the chemical erosion that is the main erosion mechanism at higher densities. This has not been studied yet.

If these results can be further substantiated a massive erosion of the buffer would probably allow the canister to sink before the hydraulic conductivity has decreased enough for flow through. Then a different situation would have to be studied. Such a study has not been attempted in this exploratory report.

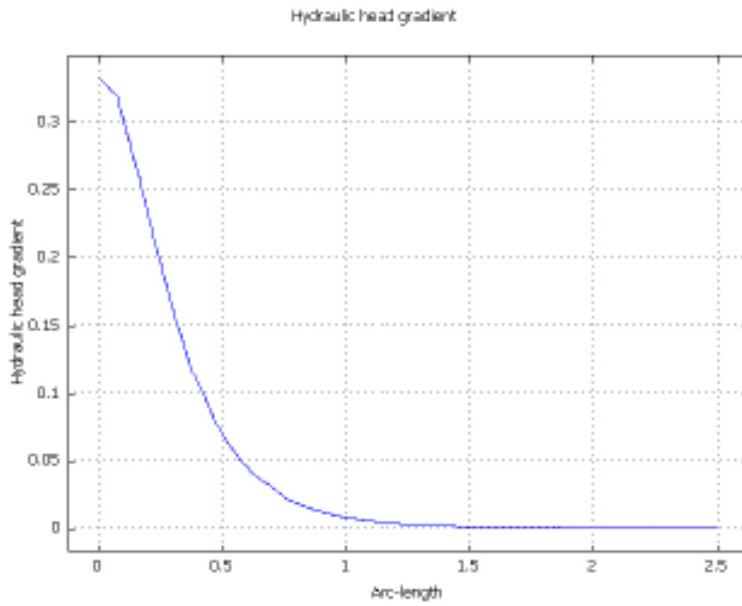
Below we address the second point of how large a cross section of the eroded buffer will be so that most of the water will pass through. The water flowing into the buffer spreads out vertically up and down the canister and converges again to flow out through the fracture at the other side of the hole. A 2D vertical cross section of the hole is modeled but without any canister. The model is set as a 2.5 m high 0.7 m long ( $2 d_{\text{buffer}}$ ) zone. Due to symmetry only the upper half the height of the canister region is modeled as the fracture intersects the hole in the middle. An inlet point (the intersection with the inlet of the fracture) is located at the lower left hand corner and an exit point at the right hand corner.

Darcy's equation is solved for the two dimensional region. The general program Comsol Multiphysics (2005) was used to obtain the numerical solutions. The solution also verified that the plug length discussed above was on the order of 3–5.

Figure 6-2 shows the head distribution and the streamlines in the region above the fracture in the rock.



**Figure 6-2.** Pressure head distribution and streamlines in the eroded buffer.



**Figure 6-3.** Hydraulic head gradient (or flowrate) from location of the fracture and upward the canister.

It is seen from the figures that practically all flow will pass through 5–10% of the height of the hole. For illustrative purpose in this note a fraction “f” of 0.1 is used. However, for long residence times the diffusion in the vertical direction must also be accounted for. For water residence times of tens of years and longer, diffusion will considerably carry the solutes from the streamlines and the above idealized analysis is no longer valid. Then it would be more appropriate to assume that the entire canister surface can be accessed by the solute. This will act to more evenly distribute a corrosive agent over all the surface of the canister.



## 7 Copper corrosion

The water contains sulphide which can corrode the copper. The rate of sulphide transport to the canister can be assessed from the equivalent flowrate  $Q_{eq}$  carrying the sulphide with the concentration  $c_{S^{2-}}$ . One question that must be addressed is how the sulphide spreads out over the copper surface. We again distinguish two cases; the erosion and the fracture case.

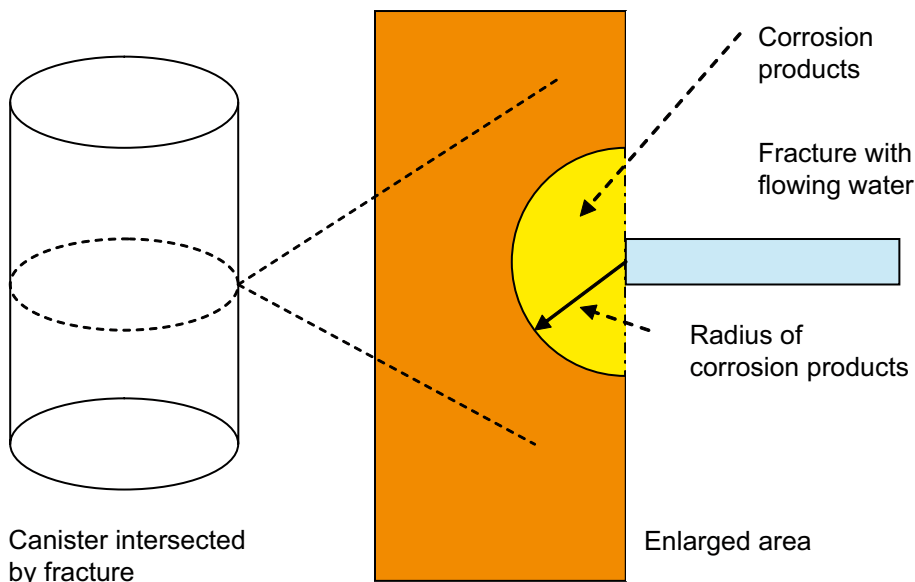
From Figures 6-2 and 6-3 it can be seen that in the erosion case the sulphide spreads out over an area that is the whole circumference of the canister and a length or height on the order of the thickness of the buffer i.e. half a meter to one meter.

In the fracture case the area of corrosion attack is smaller and is on the order of the thickness of the copper shell.

### 7.1 Rate of corrosion for fracture case

Figure 7-1 shows the basic model structure used to quantify the size of the corrosion area on the copper canister.

Water that flows in the fracture carries sulphide or sulphate that could be reduced to sulphide by microbially mediated reactions at the canister surface. The copper is corroded in a thin slit where the fracture intersects the canister. A mass balance can be formulated by assuming that the canister at the intersection corrodes as a half cylinder into the copper. All the sulphide carried by the  $Q_{eqFB}$  in the fractured buffer is transformed to  $Cu_2S$  and remains in the location where the copper corroded. When the half cylinder has attained a radius equal to the thickness of the copper canister wall it is breached. We wish to calculate the time for this to happen. The copper volume  $V_{Cu}$  in the half cylinder which extends all around the canister is that of a half cylinder



**Figure 7-1.** Canister intersected by a fracture in the cemented buffer. Corrosion in a half cylindrical region all around the canister circumference.

which is  $2\pi r_{can}$  long and with a cross section  $\pi \cdot Cu_{Thickness}^2/2$ . In a first approximation the time to breach the canister is then  $V_{Cu}/(Q_{eq} \cdot c_{S^{2-}}) \cdot const$ . The constant “const” translates the volume of copper ( $m^3$ ) that is corroded by one mol of sulphide.

$$t_{breach} = \frac{V_{Cu}}{Q_{eqFB} C_{S^{2-}}} \frac{\rho_{Cu_2S}}{M_{Cu_2S}} \quad (13)$$

The first approximation can be improved by also accounting for the diffusion resistance of the copper corrosion products. The sulphide from the flowing water has to diffuse through the corrosion products in the half cylinder to reach the remaining intact copper. The diffusion resistance in this half cylinder is added to that in the flowing water. The equivalent flowrate accounting also for the resistance in the corrosion products  $Q_{eq}^{tot}$  is obtained by solving the diffusion equation for the half cylinder along all its length  $2\pi r_{can}$ , see /Bird et al. 2002 p 305/. It is

$$Q_{eq}^{tot} = \frac{1}{\frac{1}{Q_{eqDZ}} + \frac{Ln(r/r_i)}{2\pi r_{can} \pi D_{Cu_2S}}} \quad (14)$$

here  $r_i$  is taken to be half the aperture of the fracture in the buffer. The radius of the corroded half cylinder region changes as the corrosion progresses. The mean of the combined equivalent flowrate is therefore obtained by integrating the above Equation (14) from  $r = r_i$  to  $r =$  copper thickness  $= r_y$ .

$$Mean Q_{eq}^{tot} = \frac{1}{r_y - r_i} \int_{r_i}^{r_y} \frac{dr}{\frac{1}{Q_{eqDZ}} + \frac{Ln(r/r_i)}{2\pi r_{can} \pi D_{Cu_2S}}} \quad (15)$$

With the data in Table 7-1 the time to breach the 50 mm thick canister wall is 7 million years for a transmissivity of  $10^{-8} m^3/s$ . For a transmissivity of  $10^{-6} m^3/s$  the time is reduced to 0.7 million years. There is a nearly square root dependence of  $Q_{eq}$  with flowrate. It may also be noted that the influence of the resistance of the corrosion products is very small, on the order of 6% increase in corrosion time. The diffusivity in the corrosion products must be at least one order of magnitude lower to have a serious influence. The value used here would be applicable to a corrosion product with a porosity on the order of 10–20%. As there are no data for this entity it cannot be relied on to have an important effect.

The sulphide concentration in the example was chosen to be 1 mg/litre. Natural waters have lower concentrations which would lead to longer corrosion times. Should there be sulphide formation by microbially mediated sulphate reduction it is conceivable that a higher value for sulphide concentration could be appropriate. Then the time should be proportioned accordingly.

**Table 7-1. Data for the corrosion example.**

Entity	Meaning	value
$R_f (= B)$	Half fracture aperture	0.5 mm
$r_y$	Copper thickness	50 mm
$r_{can}$	Radius of canister	0.525 m
$D_{Cu_2S}$	Diffusivity in corrosion product (Rough estimate)	$10^{-10} m^2/s$
$M_{Cu_2S}$	Molecular weight of corrosion product	159.2 g/mol
$\rho_{Cu_2S}$	Density of corrosion product	5,800 kg/m <sup>3</sup>
$c_{S^{2-}}$	Sulphide concentration in water	1 mg/litre

## 7.2 Rate of corrosion for erosion case

The rate of copper corrosion can also be estimated for the erosion case by a simple mass balance. This is done for different conditions for fracture transmissivities and apertures giving different  $Q_{eq}$ 's.

At the highest flowrate it would take about 200,000 years to penetrate the canister. Note that this is a very unlikely case as it is based on a case where the entire buffer has disappeared in about 10% of the canister length. The time for this to occur would be on the order hundreds of thousands of years even under the most unfavorable circumstances.

**Table 7-2. Corrosion rates for the eroded buffer case when 50 cm height of canister surface (1.65 m<sup>2</sup>) is attacked. Sulphide concentration 1 mg/litre.**

$T$ m <sup>2</sup> /s	$Q_{eqEB}$ l/year	m <sup>3</sup> /year	mm/year
10 <sup>-9</sup>	1.1	3.8 10 <sup>-10</sup>	2.3 10 <sup>-7</sup>
10 <sup>-8</sup>	11	38 10 <sup>-10</sup>	2.3 10 <sup>-6</sup>
10 <sup>-7</sup>	110	380 10 <sup>-10</sup>	2.3 10 <sup>-5</sup>
10 <sup>-6</sup>	1,100	3,800 10 <sup>-10</sup>	2.3 10 <sup>-4</sup>

## 8 Solute transport resistance through hole in canister and in entrance to buffer

In the previous sections we have considered the transport resistances due to diffusion in flowing water. There are, however, additional resistances that play an important role, especially for radionuclide release. The nuclide has to diffuse through a damage in the canister, it then has to diffuse through the backfill and also to diffuse into the narrow mouth of the fracture in the rock. For the copper corrosion the diffusion resistance in the buffer will add to that in the flowing water in the fracture in the rock. All these resistances are additive and the largest resistance will have the largest influence on the rate of transport. These resistances are assessed below in the form of the equivalent flowrate which is the inverse of the resistance.

The diffusion resistance in the corrosion product or clay that intruded the hole in the canister can also be expressed as an equivalent flowrate by the direct application of Fick's first law

$$Q_{eqhole} = D_{fill} A_{hole} \frac{1}{d_{Cu}} = D_{fill} \frac{\pi d_{holeincan}^2}{4d_{Cu}} \quad (16)$$

It may further be noted that the  $Q_{eq}$  obtained for the fractured buffer determines the rate of solute transfer between the flowing water and the surface of the canister. Any transport resistance by diffusion in the buffer is now non-existent in this extreme scenario. The transfer resistance for nuclides from a small hole in the canister, unless directly intersected by the fracture in the buffer, can still be accounted for as before by the use of an equivalent plug now with the diffusion properties of the cemented buffer.

The equivalent flowrate in the buffer at the exit of the hole is obtained from /Romero et al. 1995/.

$$Q_{eqholeExit} = D_{buffer} A_{hole} \frac{4}{d_{holeincan}} = \pi D_{buffer} d_{holeincan} \quad (17)$$

The equivalent flowrate in the buffer at the exit/entrance to the fracture in the rock is obtained from /Neretnieks 1986/ where the number "5" could be between 3 and 8 depending on the geometry of the deposition hole

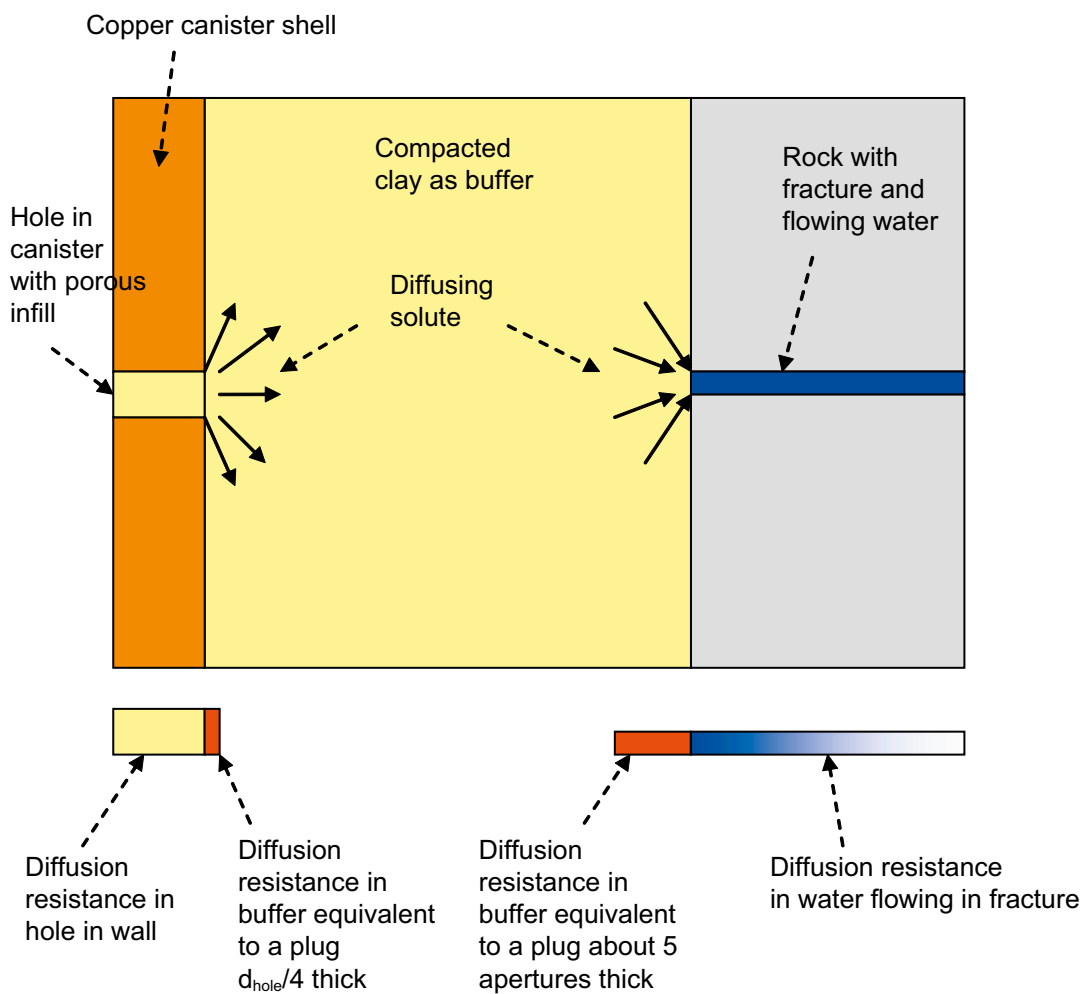
$$Q_{eqFracture} = \frac{D_{buffer}}{5 \cdot 2b} A = D_{buffer} \frac{2\pi r_{hole}}{5} \quad (18)$$

Figure 8-1 illustrates the different resistances and that those at the exit from the hole and entrance to the fracture can be modeled as resistance in plugs. Nuclide release can be strongly limited by the diffusion resistance in the damage in the canister as is shown in Table 8-1. The diffusion coefficient in the infill in the hole in the canister  $D_{fill}$  is taken to be the same as in the buffer.

It is seen from the data in Table 8-1 that the transport resistance through a hole in the canister is very large. It would translate to an equivalent flowrate of 50 micro litres per year. Even if the hole was 10 times larger, i.e. 10 mm diameter, the equivalent flowrate would be only 5 millilitres per year. It is also seen that as long as there is no flow through the buffer the equivalent flowrate would be limited by  $Q_{eqFracture}$  when the flowrate in the fracture in the rock would have allowed more transport.

**Table 8-1. Equivalent flowrates for transport through hole in canister and for entrance/exit into/from buffer.**

Entity	Meaning	value
$d_{hole\ in\ can}$	Diameter of hole in canister	0.001 m
$d_{Cu}$	Copper thickness in canister	50 mm
$D_{buffer}$	Diffusion coefficient in buffer	$1.2 \cdot 10^{-10} \text{ m}^2/\text{s}$
$Q_{eqHole}$	Inverse of resistance to diffusion in hole in canister expressed as equivalent flowrate	$1.8 \cdot 10^{-15} \text{ m}^2/\text{s}$ ( $6.0 \cdot 10^{-5} \text{ l/year}$ )
$Q_{eqHoleExit}$	Inverse of resistance to exit from hole in canister to buffer, expressed as equivalent flowrate	$3.8 \cdot 10^{-13} \text{ m}^2/\text{s}$ ( $0.012 \text{ l/year}$ )
$Q_{eqFracture}$	Inverse of resistance to exit from fracture in rock to buffer, expressed as equivalent flowrate	$7.9 \cdot 10^{-11} \text{ m}^2/\text{s}$ ( $2.55 \text{ l/year}$ )



**Figure 8-1.** Solute transport from a damaged canister through buffer and into flowing water in fracture in rock.

## 9 Earth quake case – sheared canister

Here we estimate the transport resistance from a canister that has been sheared and when the two parts of the canister have been dislocated by 10 cm. The buffer is still there, is some 20 cm thick, it has the original density and diffusion properties. The fracture in the rock can be assumed to have a considerably higher transmissivity due to the shearing and the main transport resistance is due to the diffusion in the buffer.

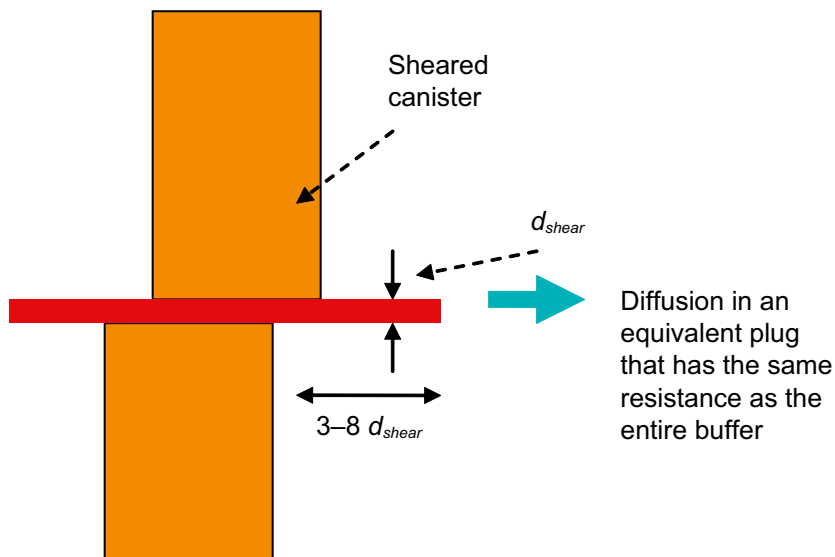
### 9.1 Diffusion from the exposed fuel at the open ends of the sheared canister into the buffer

Diffusion from the “slit” in the sheared canister into the clay is modeled with the same method as transport to and from a fracture in the rock. See Section 8 above and /Neretnieks 1986/. All the resistance to diffusion can be seen as located in a thin band around the canister. The cross section A for diffusion is  $d_{shear} \cdot 2\pi r_{can}$  and the diffusion distance is  $3-8 d_{shear}$ , see Figure 9-1.

Then the equivalent flowrate for a diffusion distance of  $5 d_{shear}$  is

$$Q_{eShear} = \frac{D_{buffer}}{d_{shear}} A = D_{buffer} \frac{2\pi r_{can}}{5} \quad (19)$$

It is about  $7.9 \cdot 10^{-11}$  m<sup>3</sup>/s or 2.5 litres per year and is independent on the shear aperture up to about a cm.



**Figure 9-1.** A canister is sheared and nuclides from the interior diffuse out into the bentonite. All the diffusion resistance can be approximated to exist in a narrow band around the sheared canister.

## 10 Discussion and conclusions

This exploratory report is based on some simplified assumptions regarding the flowrates that can result from damages to the buffer

There are many uncertainties that must be accommodated when trying to assess the consequences of such damages for the corrosion of the canister and for radionuclide release. An incomplete list of issues that should be addressed better if deemed important (or circumvented) is given below.

### 10.1 Cemented buffer

The reasons for cementation and the buffer are not discussed at all. It is just stipulated that it has happened and that subsequently the buffer has become fractured. A fracture is stipulated to connect the flowing fracture in the rock with the canister. Thus the three points below are *stipulated* to have happened without any physical or chemical reasons. Possibly there is an exception for the cementing because it is known that silica dissolution, transport and re-precipitation can take place in a thermal gradient. This might cause some cementing.

- Cementation of buffer.
- Fracturing of buffer.
- Hydraulic properties of cemented buffer.

### 10.2 Eroded buffer

Erosion of the buffer could be caused by the intrusion of low ionic strength water. Such water could come from melting ice during some periods of an ice age. The flowrates could be high and if the duration of the high flowrates with low salinity waters is long the buffer could be noticeably eroded in locations with high flow rates. However, massive amounts of the buffer material must have disappeared before water will flow through the buffer. In order to assess flow and transport properties of a low density buffer the three points below must be further studied. It is questionable if more insights of value could be gained from such studies.

- Relation buffer density and hydraulic conductivity at low densities.
- Erosion shear properties of buffer at low densities.
- Reorganization (swelling) of buffer in hole when local erosion occurs.

### 10.3 Rock properties

The hydraulic properties of the fractures that intersect the deposition holes can influence the flow and solute transport to and from the canister. In the simplified models used here the water flow is assumed to be evenly distributed over the fractures. Channeling or preferential flow paths in the fractures could lead to locally higher flowrates which could influence the erosion. It is at present not deemed to give an important increase above the higher transmissivity values used in the exploratory calculations. However, the points below could be studied more if there are concerns.

- Transmissivity range of fractures that can intersect deposition hole.
- Channeling in fractures.

#### **10.4 Models and modeling**

- The models used are very simple but are deemed to convey the most important mechanisms.
- The ideas presented are based on the  $Q_{eq}$  concept which previously has been a good conveyor of the notion of (inverse of) transport resistance.

This report explores possible consequences in some “what if” cases. Its aim has been to find what could potentially be the consequences of fractures in a cemented buffer and of a severely eroded buffer. The results suggest that in both cases considerable increase of transport rates of corrosive agents and later of radionuclides could result.

The author deems that the main uncertainties at present are due to the causes and extent of fracturing and of the extent of erosion. The simplified analysis method is sufficient at present but could be refined should more information become available on the points noted above.



# 11 Notation

$A_{hole}$	Cross section of hole in canister	$m^2$
$b$	Half of fracture aperture	$m$
$c$	Concentration	$mol/m^3$
$c_{eq}$	Concentration at equilibrium	$mol/m^3$
$d_{buffer}$	Thickness of buffer	$m$
$d_{hole}$	Diameter of hole in canister	$m$
$d_{hole}$	Diameter of deposition hole	$m$
$d_{plug}$	Length of (equivalent) plug	$m$
$d_{shear}$	Aperture of sheared canister	$m$
$D_{buffer}$	Diffusion coefficient in buffer	$m^2/s$
$D_w$	Diffusion coefficient in water	$m^2/s$
$i$	Hydraulic gradient	$m/m$
$K$	Hydraulic conductivity	$m/s$
$L_{hole}$	Length of deposition hole	$m$
$M$	Molecular weight	$g/mol$
$N$	Flowrate of solute	$mol/s$
$N_{Clay}$	Flowrate of clay	$kg/s$
$q$	Flowrate of water	$m^3/s$
$Q_{eq}$	Equivalent flowrate	$m^3/s$
$r_{can}$	Radius of canister	$m$
$r_{hole}$	Radius of deposition hole	$m$
$t$	Time	$s$
$t_{breach}$	Time to breach canister	$s$
$t_{res}$	Residence time	$s$
$T$	Hydraulic transmissivity	$m^2/s$
$u$	Velocity	$m/s$
$V$	Volume	$m^3$
$W$	Width	$m$
$W_{capture}$	Capture width	$m$
$x$	Distance	$m$
$X_{salt}$	Concentration of salt in the buffer	$mol\ salt/kg\ buffer.$
$z$	Distance into flowing water	$m$
$\rho$	Density	$kg/m^3$
$\eta_{mean}$	Mean penetration depth	$m$

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## The meaning and use of the expression “Equivalent Flowrate” $Q_{eq}$

The term equivalent flowrate was introduced to facilitate understanding of how much solute (corrosive agent or radionuclide) could be transported from or to the canister from the water flowing in the rock /Neretnieks 1979/.

The basic idea is to assess the rate  $N$  of solute transport to or from a canister. For visualization purposes it is expressed as the flowrate of water that would be depleted (filled with) of all its solute when the water passes the canister.

$$N = Q_{eq} (c_w - c_o)$$

Here  $N$  is the rate of uptake of a solute from the flowing water having a concentration  $c_w$  to a body (canister) that maintains its concentration at  $c_o$  (For example zero  $c_o$  if the corrosive agent immediately reacts with the copper canister in the present application). The same relation can be used for release of a nuclide from the canister to the passing water. Then  $c_o$  could be the nuclide concentration at the buffer outer surface and  $c_w$  the concentration in the approaching water. The latter would commonly be zero.

$Q_{eq}$  for the transfer from the water flowing in the fractured rock to the outer surface of the buffer can be determined by assessing how far out into flowing water the solute can diffuse during the time the water is in contact with the buffer. When the water ceases to be in contact with the buffer it has taken up (been depleted of) a given mass of solute. This is obtained by modeling the flow and solute transport by well known relations (Darcy flow and Fick's second law).

The method can be extended to account also for other transport resistances in series e.g. in the buffer. A nuclide has to diffuse from the fuel through a hole in the canister to the clay, then from the exit of the hole in the canister out into and through the buffer to reach the flowing water in the fracture in the rock. As the nuclide approaches the fracture in the rock it will have to find the narrow fracture. This can also be expressed as a resistance. All these resistances can be expressed as the inverse of equivalent flowrates. The combined effect of all these resistances in series can be found by adding all the resistances. The overall effect and combined equivalent flowrate is then obtained from

$$N = Q_{eq}^{overall} (c_w - c_o) = \frac{1}{\sum \frac{1}{Q_{eq,i}}} (c_w - c_o)$$

The largest resistance (smallest equivalent flowrate) will have the largest influence of the rate of transport of the nuclide.  $c_o$  will then mean the concentration in the innermost point of the system and  $c_w$ , as before, the concentration in the far away approaching water.

Expressing the transport in this way can be used to visualize which part of the system has the largest impact on the transport. The smallest  $Q_{eq}$  (largest of the resistances in series) will have most impact on limiting the rate of transport to and from the canister.

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