
KBS TEKNISK RAPPORT

79

**Transport of oxidants and radionuclides
through a clay barrier**

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Kungl Tekniska Högskolan Stockholm 1978-02-20

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THROUGH A CLAY BARRIER

Stockholm 780220

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Summary

The mass transfer rate for oxidants to, and radionuclides from a capsule in a repository has been computed. The capsule which is 0.75 m in diameter is surrounded by Montmorillonite clay. The hole is 1.5 m in diameter. For one capsule about 1220g copper will corrode due to oxygen corrosion in 10 000 years. If the fissures in the rock nearest the hole are filled with clay, the corrosion will decrease significantly. This is valid for a case where the groundwater is in equilibrium with oxygen of 0.2 bar pressure (normal air pressure). Measurements of the oxygen content in groundwater at large depths show a more than 1 000 times smaller values. The transport rate will then be correspondingly smaller. Corrosion due to sulphate/sulphide corrosion may reach some 590 g in the same time if there is 10 mg/l of the least abundant component.

The radionuclides Sr^{90} , Cs^{137} , Am^{241} and Am^{243} will decay totally in the clay barriers. Pu^{240} will be seriously hindered.

The total dissolution of the uranium oxide in a capsule takes at least 1.8 million years.

Nuclides with high solubilities decrease in about 2 000 years to half their original concentration.

The sodium in the Montmorillonite clay in the fissures is exchanged for calcium in about 20 000 years. The exchange of the sodium in the clay in the hole takes millions of years.

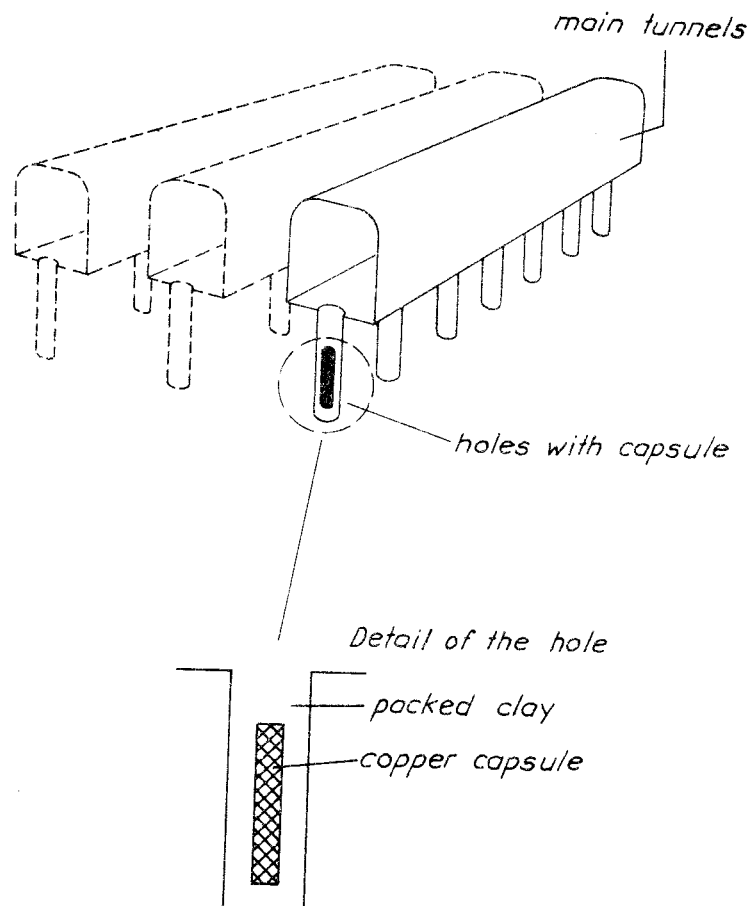
Introduction

In a repository for radioactive waste, copper capsules are surrounded by compacted clay. The clay protects the capsule from components in the groundwater by having a low permeability and diffusivity. The clay is so dense that practically no water will flow through it. Furthermore the clay has good ion exchange properties and will retard many of the radionuclides. The clay has good swelling properties and will probably fill minor fissures in the surrounding rock. In this way the fissures in the rock may be closed to water flow at some distance from the hole.

The configuration of the repository is shown schematically in figure 1. The hole is 1.5 m in diameter and the capsule is 0.75 m in diameter. The capsule is 5 m long. The fissures in the rock are filled to a certain depth with clay.

Figure 1

Schematic view of the repository



One object of this study is to determine how much oxygen and other oxidizing agents will be transported from the flowing groundwater into and through the clay to the capsule wall. Another object is to determine how escaping nuclides will wander through the clay barrier. A third object is to determine how the sodium in the Montmorillonite clay is exchanged for calcium from the groundwater.

Water flow around the buried capsule

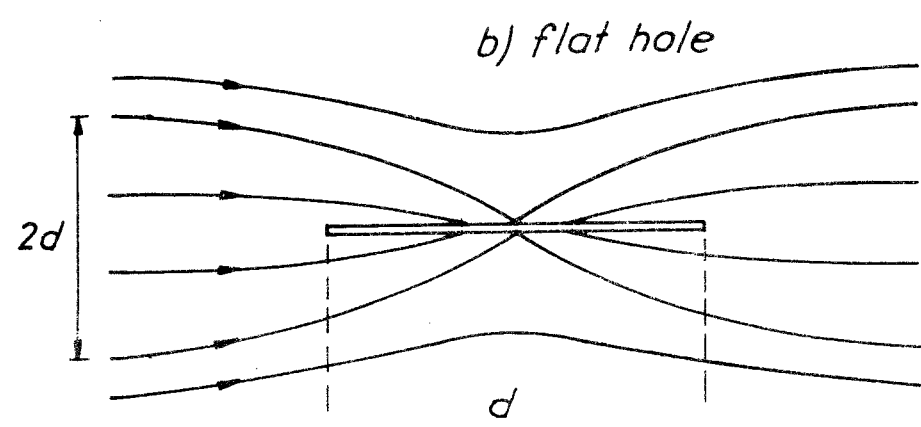
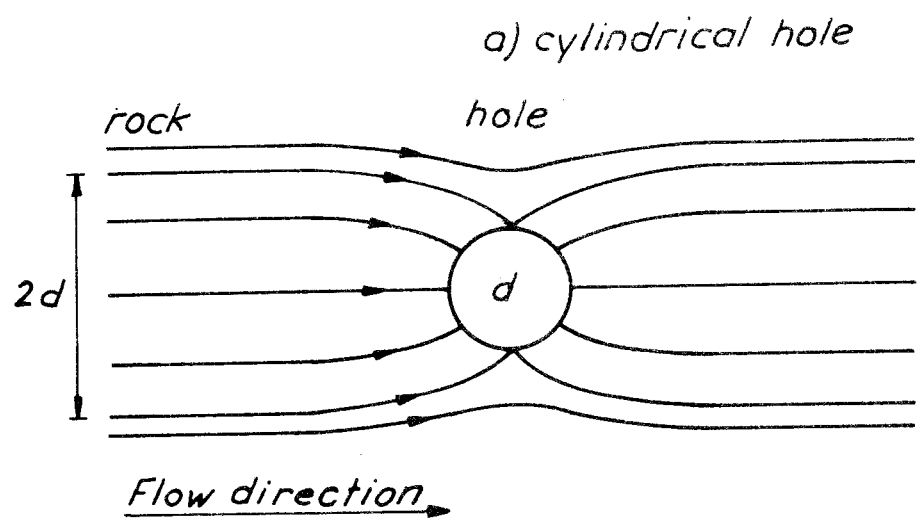
The water flow in the rock will be somewhat larger in the vicinity of the holes, as the rock probably will be more cracked there due to the disturbance from the excavating activities. The increase in water flow will not be very large however. It will not be larger than about two times that in the undisturbed rock. This may be deduced from potential theory (Tietjens). It may be visualised in the following way. If the rock has an infinitely long cylindrical hole perpendicular to the flow as shown in figure 2 a, the water from an area 2 times the diameter of the hole will be drawn into the hole. This means that if the water flow rate or bulk velocity in the undisturbed rock is $u_0 \text{ m}^3/\text{m}^2\text{s}$, then the flow rate in the hole will be $2 u_0$.

A similar flow pattern is obtained for an infinitely long flat hole. This is shown in figure 2b. The hole has a length in the flow direction of d . In this case all the water from an area $2d$ will be drawn into the hole.

The first case is applicable when the fissures in the rock are perpendicular to the hole with the capsule and if the rock near the hole is very much more permeable to water flow than the undisturbed rock far away. It is the permeable rock which has flow properties similar to an empty hole. The presence of an impermeable area - the clay - inside the permeable area does not influence the flow pattern in the rock. The second case would approximately describe the case where there is a wide fissure along the length of the hole and when the water flow is parallel to the axis.

There are no detailed data on the size and orientation of the fissures in the rock. In the following computations the flow

Figure 2.
Pontential flow around a hole in the rock



rate in the vicinity of the hole has been taken to be twice that in the undisturbed rock. This is probably a conservative value as the permeability is finite in reality, while the theoretical treatment above assumes an infinite permeability in the hole.

However some computations are also made to determine the influence of a considerably higher flow rate.

As will be shown later, the retention time t of the water near the hole has a major influence on the transport of the solved species. This is determined by the actual velocity u_p of the water in the fissures and by the distance travelled z_o

$$t = z_o / u_p \quad 1$$

The velocity in the fissures u_p is

$$u_p = u_o / \epsilon_p \quad 2$$

in the undisturbed rock. ϵ_p = porosity of the rock. In the vicinity of the hole the flowrate may increase as described above and the porosity may also be larger than that in the undisturbed rock.

A detailed discussion of the bulk velocities in the rock were made in part II of "Kärnbränslecykelns slutsteg" (1977). A series of measurements in deep wells have shown that u_o is probably less than $2 \cdot 10^{-4}$ m/year or 0.2 liter/m², years. The measurements also indicate that the distance between fissures is very large at depths exceeding 300-600 m.

In this study fissure spacings of 1 m and a fissure width of 0.1 mm is used as conservative values for the rock near

the hole. This leads to permeabilities more than 100 times larger than for the undisturbed rock. An extreme case with fissure spacing 0.4 m and fissure widths of 0.2 mm is also used in the computations to determine the influence of an extremely high porosity and thus retention time near the hole.

A further test of the influence of extreme cases is made by using a bulk velocity $u_0 = 1 \text{ l/m}^2, \text{year}$ in the undisturbed rock.

4 Transport of solved species to and from
the capsule

There are three distinct transport resistances. A component first has to diffuse from the flowing water to the clay in the fissure. The time available for this diffusion is the time it takes for the water to flow past the clay in the hole. During this time only those molecules within a certain distance from the clay will have sufficient time to reach the clay.

The second resistance is that due to diffusion in the clay in the thin fissure.

The third resistance is due to diffusion in the compacted clay in the hole.

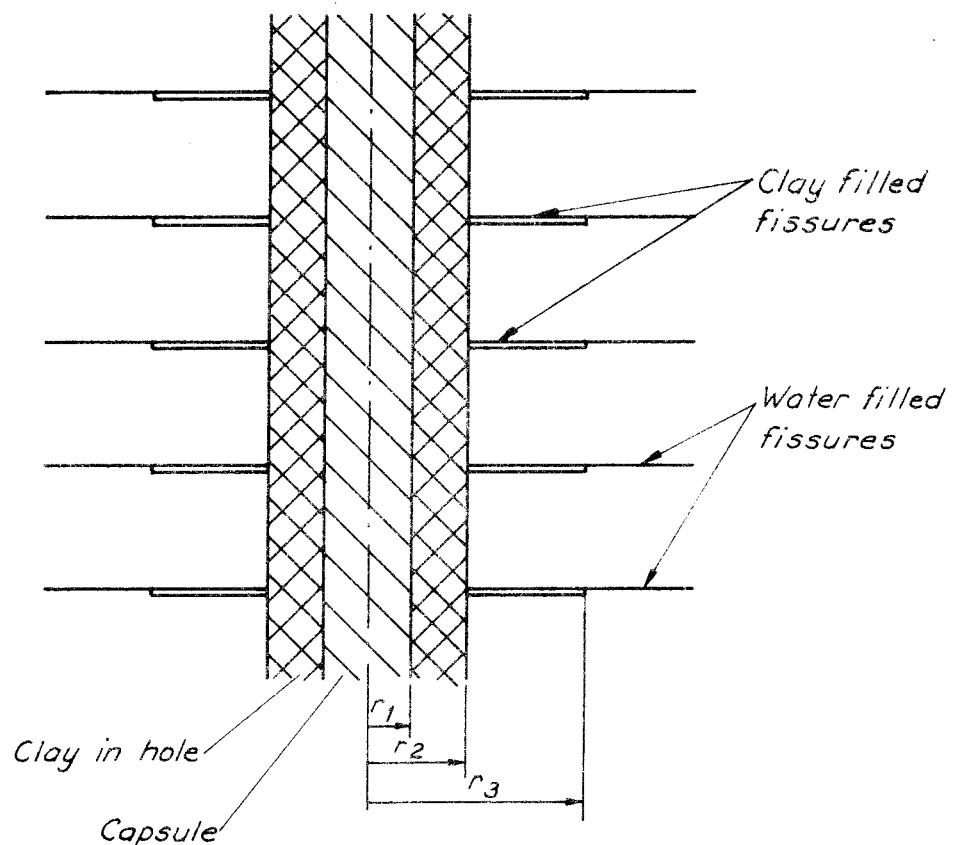
The driving force is the difference in concentration between the bulk of the liquid and at the capsule wall. It is assumed that the oxidizing agents are consumed on arrival at the capsule wall and that the concentration there always is zero of these components.

The oxidizing agents O_2 and the presumed SO_4^{2-}/S^{2-} system do not interact with the clay. All cations (Ca^{2+} and many radionuclides) on the other hand will interact with the Montmorillonite of the clay by ion exchange. The noninteracting species will reach stationary conditions very quickly, whereas the cations may need considerable time to reach stationary conditions due to accumulation in the clay. This has been treated by Neretnieks (1977).

5 Computational methods5.1 Stationary transport

The various transport steps are given by equations 3-5 below (Bird et al 1960). From the cylinder wall out, we have the concentration differences Δc_1 , Δc_2 and Δc_3 over the clay barrier, the clay filled fissures and the "film" surrounding the cylinder with radius r_3 - see figure 3. The cylinder has a length L . The fissures are spaced evenly along the cylinder at a distance S . They have a width $2b$. N is the total mass transferred per unit of time from the cylinder.

Figure 3
Arrangement of capsule and clay in the repository



$$N = \frac{2\delta}{S} 2\pi L \frac{\bar{r}_1}{\Delta r_1} D_1 \Delta c_1 \quad 3$$

$$N = \frac{2b}{S} 2\pi L \frac{\bar{r}_2}{\Delta r_2} D_2 \Delta c_2 \quad 4$$

$$N = \frac{2b}{S} 2\pi L r_3 k_v \Delta c_3 \quad 5$$

\bar{r}_i = the logarithmic mean radius

$$\bar{r}_i = \frac{r_{i+1} - r_i}{\ln \frac{r_{i+1}}{r_i}} = \frac{\Delta r_i}{\ln \frac{r_{i+1}}{r_i}} \quad 6$$

D_1 and D_2 are the diffusivities in the clay in the hole and in the clay in the fissures respectively. δ is a fictitious mean diffusion width in the clay in the hole.

δ is determined by the assumption that the diffusion in the clay barrier has a cylindrical symmetry with the fissure as the central cylinder. δ can be seen as a mean width for the transport from the fissure into the clay. It should be somewhere between $2b$ - the fissure opening and S the spacing between the fissures.

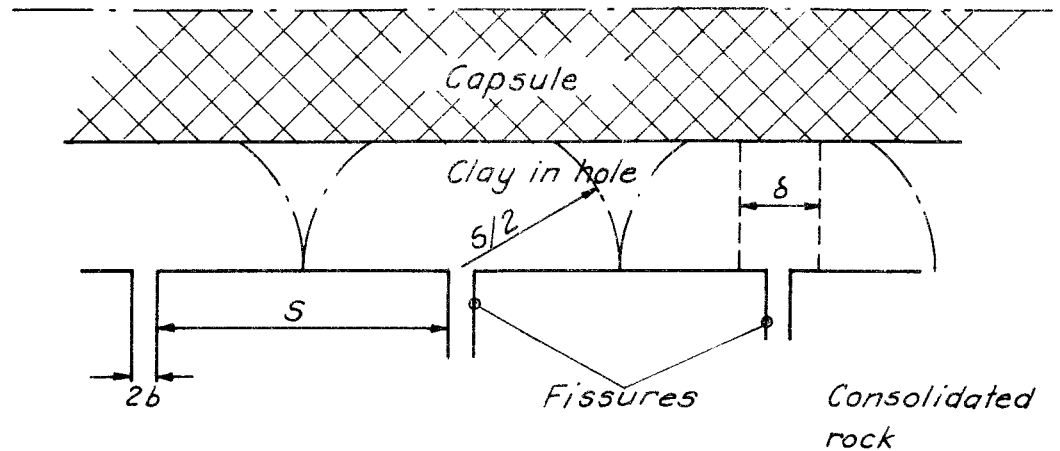
For cylindrical symmetry δ is the logarithmic mean of $2b$ and S .

$$\delta = \frac{S-2b}{\ln S/2b} \quad 7$$

Figure 4 shows the mean transport width.

Numerical computations show that this is a very good approximation.

Figure 4
Two dimensional diffusion from the fissure into the hole



The individual Δc_i 's can be eliminated to give the total concentration difference Δc between the bulk of the water in the rock the capsule wall to give

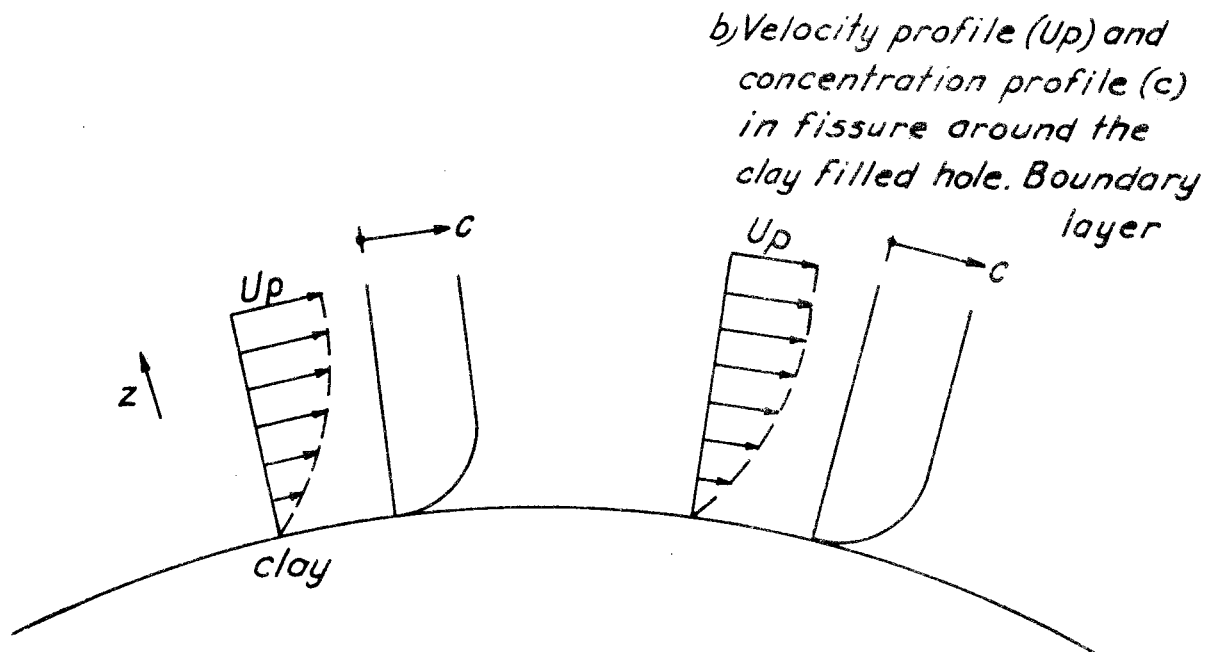
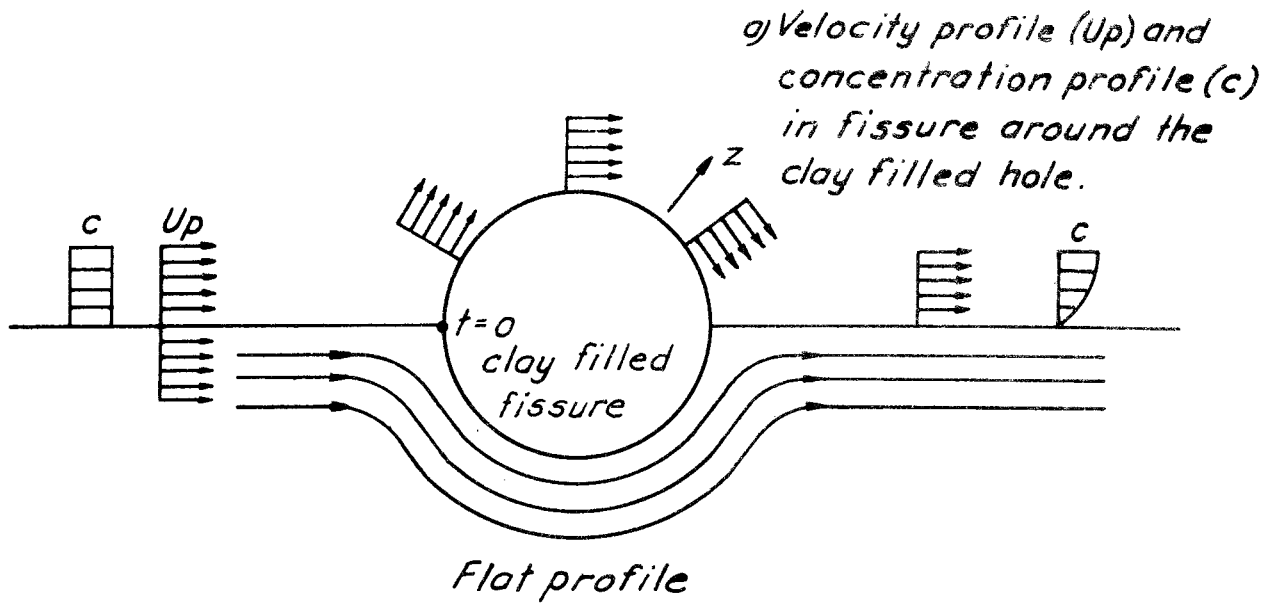
$$N = \frac{2\pi L \cdot 2b \cdot \Delta c}{S \left(\frac{1}{r_3 k_v} + \frac{\Delta r_2}{r_2 D_2} + \frac{\Delta r_1 2b}{r_1 D_1 \delta} \right)} \quad 8$$

The three terms in the denominator are proportional to the mass transfer resistances in the three barriers. From left to right we have the resistance in the liquid surrounding the clay filled fissures, the resistance in the clay filled fissures and the resistance in the clay in the hole.

5.2 "Film transfer coefficient" k_v

The transport from the water flowing in the fissure to the clay may be determined by use of the equation of diffusion for the instationary case. Two flow patterns are considered. In the first case it is assumed that the velocity profile of the water is flat and uninfluenced by the presence of the cylindrical wall. This is shown in figure 5. In this case the velocity is constant and the equation of diffusion may be applied directly.

Figure 5



$$\frac{\partial c}{\partial t} = D_v \frac{\partial^2 c}{\partial z^2}$$

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with initial conditions

$$\begin{aligned} c &= c_0 & \text{all } z & \text{ for } t \leq 0 \\ c &= 0 & z = 0 & \text{ for } t > 0 \\ c &= c_0 & z = \infty & \text{ for } t > 0 \end{aligned}$$

where z = distance from wall

t = contact time for liquid at wall

and D_v = diffusivity in the liquid.

The solution to this classic problem is given in Bird et al (1960) and k_v may be determined directly to be

$$k_v = \sqrt{\frac{4}{\pi}} \frac{D_v}{t}$$

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$$t = \frac{\pi r_3}{u_p} \quad \begin{array}{l} \text{the time it takes for the liquid to pass} \\ \text{the cylinder wall} \end{array}$$

u_p = velocity in the fissure.

The other case considered is where the flow near the clay is retarded by the presence of the surface of the clay. The velocity profile near the clay for this case is given in figure 5b.

If the curvature of the cylinder is small, compared to the distance which the velocity profile is influenced, the boundary layer theory may be used to determine the mass transfer coefficient k_v . Bird et al (1960). A very good approximation to the exact solution is

$$Nu_{AB} = 0.664 \cdot Re^{1/2} \cdot Sc^{1/3}$$

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$$\text{Nu}_{AB} = \frac{k_v 2r_3}{D_v} \quad \text{Nusselt number}$$

$$\text{Re} = \frac{u_p 2r_3}{\nu} \quad \text{Reynolds number}$$

$$\text{Sc} = \frac{\nu}{D_v} \quad \text{Schmidt number}$$

ν = kinematic viscosity

Equation 11 is applicable for $\text{Re} < 1000$. Equation 11 gives somewhat lower mass transfer coefficients than equation 10. In the computations k_v -values are determined by equation 10 as the flow in the fissures is better described by this case. Also this gives a conservative estimate of the mass transfer rate.

5.3 Instationary case

The instationary case is only of interest when the diffusing species is a nuclide which is retarded by the clay barrier due to ion exchange and similar mechanisms. Flow and diffusion with retardation has been treated in detail by Neretnieks (1977) for the case where there is a flat barrier. This is a fair approximation also for cylindrical barriers if the ratio of the radii is not too large. In this case there are composite barriers and this makes it more complex. The first barrier is the clay in the hole. The second is the clay in the fissures and the third is the rock itself, which also interacts with many of the nuclides.

In the following these barriers will be treated separately. This makes the treatment rather conservative. The interaction with the rock has been treated previously by Neretnieks (1977) and is not treated further here.

The following assumption are made: The concentration of the nuclide at the capsule wall is constant. When the concentration at the hole surface (at r_2) has reached 5 % of the concentration at the capsule wall, we have a breakthrough. The transport then starts in the next barrier which is the clay in the fissures. The concentration inside this barrier is then suddenly assumed to be the same as at the capsule wall. Breakthrough in this barrier occurs when the concentration reaches 5 % of the concentration at the capsule wall. This happens approximately when $\frac{Dt}{k(\Delta r)^2} \approx 0.1$.

When $\frac{Dt}{k(\Delta r)^2} \approx 1$ the concentration outside the barrier will have reached 90-95 % of the concentration at the capsule wall. The latter may be used as a criterion for determining when a stationary concentration profile has developed in the barrier.

5.4 Basic data for the computations

The flow rate of water has been treated earlier. In the central case used in the computations a value of $u_0 = 0.2$ $1/m^2$, year is used for the undisturbed rock. The fissure spacing $S = 1$ m and the fissure width $2b = 0.1$ mm.

The diffusivity of methane and hydrogen in compacted wet clay have been measured. Neretnieks and Skagius (1978). Diffusivities of solved gases, small anions and larger complexed cations in water have been computed using standard methods and data in Reid et al (1977) and Landholt, Börnstein.

Based on these data the following diffusivities at 50°C were estimated and are given in Table 1 below.

Table 1

	Water	quartz/clay 90 %/10%	clay 1.9* g/cm^3 (estimated)	clay 2.1* g/cm^3	clay in fissures (estimated)
Relative diffusivity	1	1/10	1/50	1/100	1/5
Diffusivity at 50°C m^2/s for O_2	$3.9 \cdot 10^{-9}$	$3.9 \cdot 10^{-10}$	$7.8 \cdot 10^{-11}$	$3.9 \cdot 10^{-11}$	$7.8 \cdot 10^{-10}$
Anion	$3.9 \cdot 10^{-9}$	$3.9 \cdot 10^{-10}$	$7.8 \cdot 10^{-11}$	$3.9 \cdot 10^{-11}$	$7.8 \cdot 10^{-10}$
Cation	$2 \cdot 10^{-9}$	$2 \cdot 10^{-10}$	$4 \cdot 10^{-11}$	$2 \cdot 10^{-11}$	$4 \cdot 10^{-10}$

*This includes about 10 % water

Diffusivities for the quartz/clay mixture were obtained from measurements of strontium (Neretnieks 1977) and silver diffusion (Allard 1977).

The diffusivity in the clay in the fissures is an assumed value. No data are available on the density there.

6 Results6.1 Transport of oxidants

The solubility of oxygen is $13.9 \text{ g O}_2/\text{m}^3$ at 0°C . At 20°C it is $8.8 \text{ g O}_2/\text{m}^3 \text{ H}_2\text{O}$ when the equilibrium pressure is 0.2 bar (Perry 1973). The groundwater at the depths of interest has a much lower content of oxygen however. Rennerfelt (1977) has found no oxygen in groundwater from depths below 450 m. The oxygen content is less than 0.01 mg/l , which is the limit of accurate measurements. The transport of oxygen to the capsule is directly proportional to the oxygen content in the water if it is assumed that the oxygen is consumed quickly at the capsule wall. The computations are made for the very conservative case with the oxygen content of the water equal to $13.9 \text{ O}_2/\text{m}^3 \text{ H}_2\text{O}$.

The corrosion due to sulphate/sulphide is limited by the species of the smallest concentration. It is assumed that the limiting concentration is 10 g/m^3 of sulphur as either SO_4^{2-} or S^{2-} , the other species being available in unlimited quantities.

Table 2
Transport of O_2 and S to the capsule

Case	u_0 m/year	2b mm	S m	Z_0 m	N/L ΔC g/year, m, g/m ³	Res 1	Res 2	Res 3	t_a years
1	$1 \cdot 10^{-3}$	0.1	1	0	$44 \cdot 10^{-5}$	1	0	4.4	1000
2	"	"	"	2	$1.2 \cdot 10^{-5}$	1	204	2.3	30000
3	"	0.2	0.4	0	$86 \cdot 10^{-5}$	1	0	2.4	500
4	"	"	"	2	$5.7 \cdot 10^{-5}$	1	49	1.3	6000
5	$2 \cdot 10^{-4}$	0.1	1	0	$22 \cdot 10^{-5}$	1	0	10.0	2000
6	"	"	"	2	$1.2 \cdot 10^{-5}$	1	204	5.2	30000
7	"	0.2	0.4	0	$46 \cdot 10^{-5}$	1	0	5.4	1000
8	"	"	"	2	$5.5 \cdot 10^{-5}$	1	49	2.8	6000

Table 2 shows the results for these computations. The different cases are numbered and defined in columns 1-5. Case nr 5 is the central case. In this case there is no clay in the fissures, $z_0 = 0$. Column 6 gives the amount of the oxidant in grams which can be transported to 1 m length of the capsule per year if the concentration of the oxidant in the water in the rock is 1 g/m^3 (or mg/l). Columns 7-9 give the relative resistances in the clay in the hole - Res 1, the clay in fissures - Res 2, and due to the film resistance - Res 3. In the central case the main resistance is due to the film $\sim 90\%$. Column 10 will be discussed later. It should be noted that even a 5-fold increase in water flow and a 5-fold increase in porosity around the hole only gives an increase of oxidant transport by factor ~ 4 - case 3. If the fissures were filled to 15 cm with clay this would bring it back to case 5 again.

Table 3
Corrosion of the copper capsule due to O_2 and S coming from the groundwater

Case	u_0 m/year	2b mm	S m	z_0 m	Corrosion after 10^4 years		Time to corrode away 1 cm of the copper years	
					g Cu due to O_2 ¹⁾	g Cu due to S ²⁾	due to O_2 ⁴⁾	due to S ²⁾
1	$1 \cdot 10^{-3}$	0.1	1	0	2470	1180	$4.6 \cdot 10^6$	$9.6 \cdot 10^6$
2	"	"	"	2	65	32	$173 \cdot 10^6$	$350 \cdot 10^6$
3	"	0.2	0.4	0	4790	2290	$2.4 \cdot 10^6$	$4.9 \cdot 10^6$
4	"	"	"	2	320	152	$36 \cdot 10^6$	$74 \cdot 10^6$
5	$2 \cdot 10^{-4}$	0.1	1	0	1220	590	$9.2 \cdot 10^6$	$19 \cdot 10^6$
6	"	"	"	2	64	32	$176 \cdot 10^6$	$350 \cdot 10^6$
7	"	0.2	0.4	0	2550	1230	$4.4 \cdot 10^6$	$9.2 \cdot 10^6$
8	"	"	"	2	310	144	$37 \cdot 10^6$	$77 \cdot 10^6$

1) For an O_2 concentration in the water 13.9 g/m^3 and $2Cu + 1/2 O_2 = Cu_2O$

2) For an S^{2-}/SO_4^{2-} concentration of 10 g/m^3 and $8 Cu + 3H_2S + SO_4^{2-} + 2H^+ = 4Cu_2S + 4H_2O$

In table 3 the corrosion on the whole 5 m long capsule is given. In column 6 the corrosion due to O_2 is shown and in column 7 that due to SO_4^{2-}/S^{2-} . With this rate about 1.12 kg Cu will corrode away in 10 000 years for the central case 5. It will take 9.2 million years to consume 1 cm copper evenly all around the capsule. Even in the most extreme case in the table - case 3 with very high water flow in the undisturbed rock and a very cracked rock near the capsule, it will take more than 2 million years to consume 1 cm.

Furthermore the oxygen content of the water is probably 1000 times lower and this lowers the corrosion due to O_2 proportionally.

It is thus concluded that the copper capsule will corrode extremely slowly due to the oxygen and sulphate/sulphide content of the groundwater.

Table 4

Nuclide	Half-life years	Retardation factor in clay in hole	Retention time in clay in hole years	Retention time in clay in fissure 2 m deep* years
Sr ⁹⁰	28	600	$1.3 \cdot 10^4$	$1.8 \cdot 10^3$
Tc ⁹⁹	$2 \cdot 10^5$	1	22	31
I ¹²⁹	$2 \cdot 10^7$	1	22	31
Cs ¹³⁷	30	400	$2 \cdot 10^3$	$1.2 \cdot 10^3$
Ra ²²⁶	$1.6 \cdot 10^3$	800	$1.8 \cdot 10^4$	$2.4 \cdot 10^3$
Th ²²⁹	$7.3 \cdot 10^3$	>1000	$> 2 \cdot 10^4$	$> 3 \cdot 10^3$
Np ²³⁷	$2 \cdot 10^6$	200	$4.4 \cdot 10^3$	600
Pu ²³⁹	$2.4 \cdot 10^4$	1200	$2.6 \cdot 10^4$	$3.7 \cdot 10^3$
Pu ²⁴⁰	$6.6 \cdot 10^3$	1200	$2.6 \cdot 10^4$	$3.7 \cdot 10^3$
Am ²⁴¹	458	>4000	$> 9 \cdot 10^4$	$> 1.2 \cdot 10^4$
Am ²⁴³	$7.4 \cdot 10^3$	>4000	$> 9 \cdot 10^4$	$> 1.2 \cdot 10^4$

* The retardation factor is assumed to be 10 times less in the clay in the fissures due to the lower (but unknown) density there.

6.2 Diffusion of nuclides from the capsule

Instationary case. Table 4 gives the retardation factors of various nuclides in the compacted ($\rho_c = 2100 \text{ kg/m}^3$) clay. The data are recomputed from Allards experiments with the clay (10 %)/quartz (90 %) mixture. This retardation factor is based on the bulk velocity.

The following nuclides are retarded more than 30 halflives and thus decay inside the barriers: Sr^{90} , Cs^{137} , Am^{241} . The following are severely hindered: Ra^{226} , Pu^{240} . All the other are delayed but pass the barriers, given time. Sr^{90} , Cs^{137} and Am^{241} are stopped in the clay in the hole already. The clay in the fissures has little additional retardation capacity.

Stationary case. The spent fuel consists of virtually pure uranium oxide - UO_2 from a chemical point of view. After a penetration of the copper capsule, water may come in contact with the spent fuel. The water nearest the fuel will solve the UO_2 . The solved uranium will then diffuse out through the clay in the same way as the oxidizing agents were transported in the other direction. The same treatment can be used to determine the transport rate of uranium and other species out from the capsule as was used for oxidizing agents in.

The driving force is determined by the solubility of uranium in the groundwater. Available data on solubility constants in the literature indicate the carbonate ion is the main component which determines the solubility of uranium (Mesmer and Baes 1977). The maximum probable concentration of carbonate and other complexing agents is about 550 mg/l (as HCO_3^{2-}) according to Rennerfeldt (1977). This would give a maximum solubility of 1070 mg U/l as $\text{UO}_2(\text{CO}_3)_2^{2-}$. This is a very high solubility - about - equal to the total salt content of the northern Baltic. Pačes (1969) has measured the uranium content of groundwater with carbonate contents 143-443 mg/l as HCO_3^- in a mine

in Czechoslovakia. In waters with oxidizing electrochemical properties ($E_h > 0$) the maximum uranium concentration found was 9 mg/l. For reducing conditions the values were less than 0.5 mg/l. The water in the repository has reducing properties.

The use of a solubility value of 1070 mg/l in the following computations is therefore a very conservative value.

Species such as iodine and cesium which are soluble and to some extent may have diffused out of the UO_2 structure will escape quicker and before the uranium oxide is dissolved. Their escape can be described by a first order reaction.

$$-\frac{\partial c}{\partial t} = \text{konst} \cdot c$$

as the concentration difference will decrease as the species escapes. The time to reduce the initial amount to half is given in the last column in table 2 for small ions (anions) and in table 5 for larger ions with lower diffusivities (cations). The time to dissolve all the UO_2 (1.4 tons) is given in column 10 in table 5. In the central case it takes 1.8 million years.

Table 5
Diffusion of uranium from the repository

Case	u_0 m/year	2b mm	S m	Z m	N/LAC g/year, m, g/m ³	Res 1	Res 2	Res 3	Time to dissolve all UO_2^*
1	$1 \cdot 10^{-3}$	0.1	1	0	$30 \cdot 10^{-5}$	1	0	3.2	$0.9 \cdot 10^6$
2	"	"	"	2	$0.60 \cdot 10^{-5}$	1	204	1.7	$4.4 \cdot 10^6$
3	"	0.2	0.4	0	$55 \cdot 10^{-5}$	1	0	1.7	$0.49 \cdot 10^6$
4	"	"	"	2	$2.9 \cdot 10^{-5}$	1	49	0.9	$9.1 \cdot 10^6$
5	$2 \cdot 10^{-4}$	0.1	1	0	$15 \cdot 10^{-5}$	1	0	7.1	$1.8 \cdot 10^6$
6	"	"	"	2	$0.6 \cdot 10^{-5}$	1	204	3.7	$45 \cdot 10^6$
7	"	0.2	0.4	0	$31 \cdot 10^{-5}$	1	0	3.9	$0.86 \cdot 10^6$
8	"	"	"	2	$2.9 \cdot 10^{-5}$	1	49	2.0	$9.3 \cdot 10^6$

* Uranium solubility 1070 g/m³

In the most extreme case of table 5 - case 3 - the time is 500 000 years.

If the clay fills the fissures to 2 m depth the dissolution time increases to 45 million years - case 6. Even a very small penetration of clay into the fissures will reduce the transport considerably.

Species which are accessible to water and are entirely soluble will escape quickly. In the central case half the initial amount will escape in 2250 years.

6.3 Sodium exchange for calcium in the clay

The clay is a sodium Montmorillonite. The sodium may be exchanged for calcium from the groundwater. This changes the swelling properties of the clay and also changes the retardation factors for the nuclides - at present in an unknown way.

The rate of exchange of Na for Ca has been treated in a manner similar to the instationary transport of nuclides in the clay. It is assumed that all the ionexchange capacity -0.6 meq/g - is used up by the calcium. The calcium content of the water is taken to be 50 mg/l (Rennerfeldt 1977).

It takes approximately 12 000 to exchange all sodium for calcium in the clay in the fissures, but several million years to exchange it in the clay in the hole. See table 6.

Table 6
Ion exchange of sodium in the clay for calcium

Case	u_0 m/year	2b mm	S m	Z_0 m	Time to exchange	
					fissures years	Na in fissures and hole years
1	$1 \cdot 10^{-3}$	0.1	1	0	--	$1.6 \cdot 10^6$
2	"-	"-	"-	2	$1.0 \cdot 10^4$	$115 \cdot 10^6$
3	"-	0.2	0.4	0	--	$0.7 \cdot 10^6$
4	"-	"-	"-	2	$1.2 \cdot 10^4$	$26 \cdot 10^6$
5	$2 \cdot 10^{-4}$	0.1	1	0	--	$3.8 \cdot 10^6$
6	"-	"-	"-	2	$1.2 \cdot 10^4$	$116 \cdot 10^6$
7	"-	0.2	0.4	0	--	$1.6 \cdot 10^6$
8	"-	"-	"-	2	$1.5 \cdot 10^4$	$26 \cdot 10^6$

7	<u>Notation</u>	
b	= half width of fissure	m
c	= concentration in liquid	g/m^3
D_1, D_2	= diffusivities in clay layers 1 and 2	m^2/s
D_v	= diffusivity in water	m^2/s
d_1, d_2, d_3	= diameters of layers 1, 2 and 3	m
k	= retardation factor (bulk velocity of water/nuclide velocity)	-
k_v	= mass transfer coefficient	m/s
L	= length of cylinder	m
N	= rate of mass transfer	g/s
Nu_{AB}	= Nusselt number for mass transfer	-
Re	= Reynolds number	-
r_1, r_2, r_3	= radius of the layers, 1, 2 and 3	m
Sc	= Schmidt number	-
S	= distance between fissures	m
u_o	= bulk velocity	m/s
u_p	= water velocity in fissure	m/s
z_o	= penetration depth of clay in fissures	m
δ	= equivalent transport width	m
ϵ_p	= porosity of the rock	-
ν	= kinematic viscosity of water	m^2/s

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SYNPUNKTER PÅ BERÄKNINGAR OCH BEDÖMNINGAR GJORDA AV I NERETNIEKS I UTKASTET "DIFFUSSION OF OXYGEN AND RADIONUCLIDES THROUGH A CLAY BARRIER" AV 771205 SAMT KOMMENTAREN TILL UTKASTET AV 780110.

1. Beräkning av syretransport till en metallbehållare med kärnkraftavfall

Beräkningssättet enligt ekvationerna 1, 2, 3, 4 och 6 är oantastligt vid användande av en fiktiv diffusionsbredd δ . Enligt vår härledning av närmevärdet på δ i ekv 5 har vi emellertid erhållit följande resultat.

$$\delta = \frac{\pi}{4} \frac{S-2b}{\ln \frac{S-2b}{2b}}$$

I ekv 6 föreligger en felskrivning. Δr_2 skall vara Δr_1 i sista termen i nämnaren.

För beräkning av massöverföringskoefficienten ur dimensionslösa samband anser vi att ekvation 7 ger för lågt värde vid Reynoldstal omkring 1. Ett bättre samband här torde vara

$$\begin{aligned} \text{Nu} &= 0.99 \text{Re}^{1/3} \cdot \text{Sc}^{1/3} & \text{Re} &= 0.4-4 \\ \text{Nu} &= 0.91 \cdot \text{Re}^{0.39} \cdot \text{Sc}^{0.31} & \text{Re} &= 4-40 \end{aligned}$$

Vi vill dock instämma med Neretnieks att vid de aktuella mycket låga strömingshastigheterna och stora cylindrarna är det bättre att betrakta filmmotståndet ur ren diffusionssynpunkt. Man beräknar därvid masstransporten i ett volymselement vatten under den tid, som elementet ligger i kontakt med cylindern. Värdet på massöverföringskoefficienten blir då

$$k_v = \sqrt{\frac{4 \cdot D}{\pi \cdot \theta}}$$

där θ är kontakttiden. Vi anser, att ett säkert antagande är att ansätta

$$\theta = \frac{D}{u_p} \quad \text{i stället för} \quad \theta = \frac{\pi \cdot D}{2 \cdot u_p}$$

eftersom medelhastigheten runt cylindern ökas genom den undanträngda vätskemängden. Den så framräknade koefficienten är att betrakta som ett absolut säkert värde om vätskans strömingshastighet är rätt bedömd.

Därest man verkligen får en inträngning av lera i sprickor, som är så små som 0.1-0.2 mm, bidrar givetvis detta kraftigt till att öka motståndet mot syretransport in till cylindern. Detta motstånd kommer då att vara helt bestämmande för transportens storlek. Inträngningen av lera i sprickorna bör fastläggas med praktiska prov.

Beräkningen av δ enligt ekvation 5 utgör ingen exakt lösning. Vi har därför genomfört kontrollberäkningar med en numerisk metod, varvid vi använt datorprogrammet FEMTEMP, framtaget för stationära och instationära värmeledningsberäkningar. Det aktuella massöverföringsfallet är nämligen exakt analogt med värmeöverföringsfallet, där diffusion ersättes med värmeledning.

I diagram 1 visas ett fall med olika placeringar av sprickorna med antaget konstant massflöde. I diagram 2 återges inverkan av sprickornas bredd med data enligt diagrammet. Av diagrammen framgår dels att den lokala syretransporten mitt för sprickan är 25-50% högre än medelflödet. Vidare ger i fallet enligt diagram 2 de numeriska räkningarna ett värde på δ som är 25% mindre än vid beräkning enligt ekvation 5.

Vi har slutligen räknat igenom några av fallen enligt den handskrivna tabellen av 780110. I fall 2 synes resultat vara fel med en 10-potens vi fick $N/L \cdot \Delta c = 3.76 \cdot 10^{-6}$ g/m, år, (g/m^3). I övrigt stämmer resultaten i stort med våra räkningar, varvid diffusionskoefficienten antagits enligt text i tabellen (baserade på mätta värden vid KTH) samt Nerethnieks ekvation för δ använts.

Vi har för fall 1 erhållit $N/(L \cdot \Delta c) = 34 \cdot 10^{-5}$ och i fall 3 $N/(L \cdot \Delta c) = 69 \cdot 10^{-5}$. Dock har vi erhållit helt olika värden för förhållandet mellan de olika motstånden enligt nedan.

	<u>Res 1</u>	<u>Res 2</u>	<u>Res 3</u>	<u>kv</u>
Fall 1	1	0	8.4	$2.5 \cdot 10^{-8}$ m/s
Fall 2	1	800	8.5	$6.6 \cdot 10^{-9}$ m/s
Fall 3	1	0	4.5	$1.2 \cdot 10^{-8}$ m/s

Sammanfattningsvis vill vi helt instämma i slutsatserna enligt sid 2 i kommentaren. Lokala värden på syreinträngningen skulle kunna vara en faktor 1.5-2 högre än de angivna i fall 3 men antagandena som leder till en maximal syretransport av ca 200 g O_2 på 10000 år så konservativa att det inte ändrar helhetsbedömningen.

2. Transport av radionuklider från behållaren

Vid ett plötsligt läckage i kapseln som omger det radioaktiva materialet förutsätts koncentrationen av radionuklider konstant längd kapselns vägg. Genom diffusion kommer radionukliderna att transporteras ut genom lerskiktet. Det blir först ett tidsberoende förlopp, vilket så småningom övergår i det stationära fallet. Det senare är analogt med det som behandlats för syretransporten utifrån och in.

Diffusionen från ett cylindriskt hål bestäms i det instationära fallet vid endimensionellt flöde av differentialekvation

$$\frac{\partial c}{\partial t} = D \left(\frac{\partial^2 c}{\partial r^2} + \frac{1}{r} \cdot \frac{\partial c}{\partial r} \right)$$

med randvärdena

$$\begin{aligned} r = r_0 &= 0.375 \text{ m} & c &= c_0 \\ t = 0 & & c &= 0, r \neq r_0 \end{aligned}$$

Enligt Carslaw-Jaeger fås lösningen till differentialekvation i diagramform och tiden för att $\frac{c}{c_0} = 0.05$ vid $r = r_1 = 0.75 \text{ m}$ kan beräknas.

$$\begin{aligned} \text{Med } D &= 1.25 \cdot 10^{-10} \text{ m}^2/\text{s} \text{ fås } t = 9.6 \text{ år och} \\ \text{med } D &= 5 \cdot 10^{-11} \text{ m}^2/\text{s} \text{ fås } t = 24.0 \text{ år} \end{aligned}$$

Om leran tränger 1 m in i sprickorna fås följande tid från koncentrationen $c = c_0$ vid $r = 0.75 \text{ m}$ till $\frac{c}{c_0} = 0.05$ vid $r = 1.75 \text{ m}$

$$\begin{aligned} D &= 1.25 \cdot 10^{-10} \text{ m}^2/\text{s} \text{ fås } t = 49.9 \text{ år} \\ D &= 2.5 \cdot 10^{-10} \text{ m}^2/\text{s} \text{ fås } t = 25.0 \text{ år} \end{aligned}$$


Dessa värden stämmer relativt väl med de värden Neretnieks erhållit vid en beräkning där det cylindriska fallet approximerats med en flat barriär. De på detta sätt summerade tiderna för genombrott av diffusionsfronten till vattenytan synes konservativa.

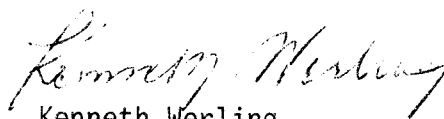
Vid fallet med retardation av radionuklider har Neretnieks definierat en retardationsfaktor k_i som vid vattenströmning genom en porös kropp är förhållandet mellan vattenhastigheten och jonernas medelhastighet. Retardationsfaktorer som synes ha bestämts enligt ovanstående definition, används för beräkning av diffusionsfallet, där diffusiviteten D ersätts med D/k_i .

Vi anser att retardationstiden kan erhållas som $k_1 \times$ diffusionstiden under förutsättning att jämvikt föreligger i varje punkt i leracylindern och att jämvikten kan definieras av ekv 1-5 i "Sorption av långlivade radionuklider i lera och berg del 1" Allard et al. Vid avvikelse från jämvikt sker genombrottet snabbare. Dock torde i det aktuella fallet avvikelsen vara liten. Genombrottet sker också snabbare om de olika nukliderna påverkar varandras jämviktswärden. Detta bör undersökas experimentellt.

Sammanfattningsvis finner vi att de av Neretnieks framräknade retentions-tiderna är de längsta tänkbara men dock sannolika därest de olika nukliderna inte påverkar varandras jämviktswärden.

Lund 1978-02-09


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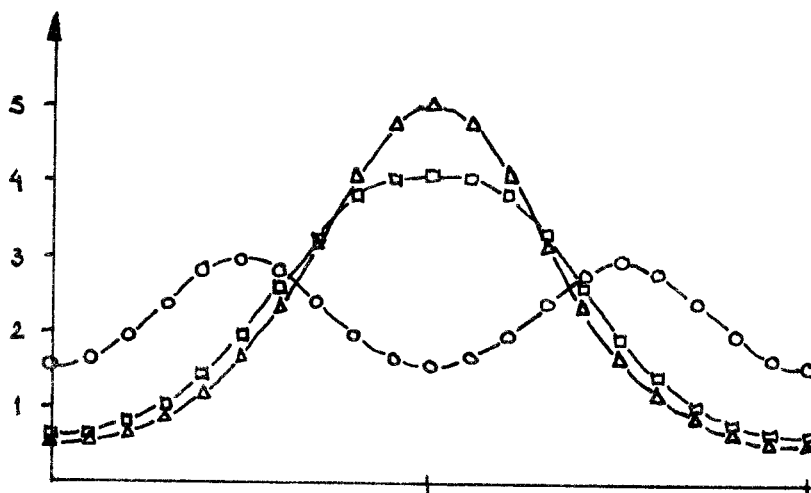
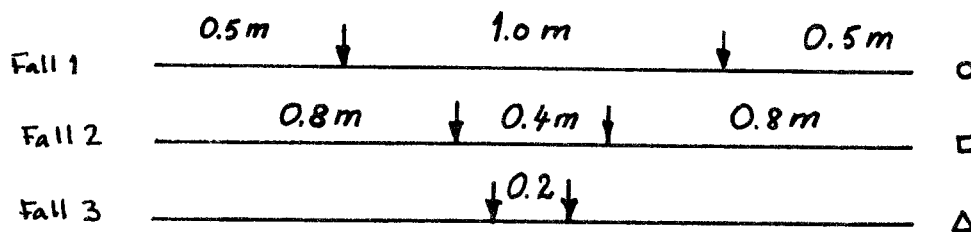
INVERKAN AV SPRICKORNAS PLACERING.

DIAGR. 1

FÖRUTSÄTTNINGAR: DIFFUSIONSMOTSTÅNDEN I SPRICKOR OCH ÖVER FILMEN ANSES SÅ STORA ATT MINDRE KONL. ÄNDRINGAR VID INLOPPE TILL LERCYLINDERN INTE PÅVERKAR DET TOTALA SYREFLÖDET. ETT KONSTANT FLÖDE HAR DÄRFÖR ANSATTS I VARJE SPRICKA.

$$110.36 = 1 \cdot 2\pi r \cdot \frac{dc}{dr}$$

Diagrammet nedan visar c vid $r = r_1 + 0.05m$ vid olika placering av sprickorna. Då c vid $r = r_1$ är 0 kan ett mått på $\frac{dc}{dr} |_{r=r_1}$ direkt fås och därmed fluxets variationer längs leracylinderns inneryta.



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