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83-66

The movement of radionuclides past a redox front

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Stockholm, Sweden 1983-04-22

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ROYAL INSTITUTE OF TECHNOLOGY
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1983-04-22

Ivars Neretnieks
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SUMMARY

It is assumed that radiolysis of water in a penetrated canister containing spent fuel has occured. Radionuclides and oxidizing agents are diffusing from the corroded canister and out through the clay barrier. A concentration front of radionuclides as well as of oxidizing agents is developed in the water that is flowing past the repository. The fronts propagates in a plane fissure with infinite extention. In the undisturbed bedrock reducing conditions normally prevail. The oxidizing agents are consumed by oxidation of the reducing components of the bedrock (primarily Fe(II)), and an oxidized region is developed. A redox front develops between the oxidizing and reducing regions. Some of the nuclides are much more soluble in an oxidizing environment than in a reducing. These nuclides will precipitate when they reach the redox front. The redox front moves much slower than the nuclides and can be assumed to be stationary at every moment.

The movement of the radionuclides in the flowing water up to and past the redox front have been computed. In the computations the transverse diffusion and precipitation at the redox front are accounted for. That part of the nuclides which is not precipitated at the passage of the redox front has been computed for different solubility ratios and at various distances downstream from the canister. The solubility ratio indicates the solubility of a nuclide in a reducing environment divided by the solubility in an oxidizing environment.

1. BACKGROUND

A canister containing spent nuclear fuel is assumed to have corroded. Water has penetrated the broken canister and is in contact with the fuel pellets. Radiation - mainly α - from the radioactive material will radiolyze the water and form reducing species (hydrogen gas) and oxidizing species (mainly hydrogen peroxide). The hydrogen gas is not very reactive and will diffuse away, but the hydrogen peroxide is a strong oxidizing agent and will attack the fuel and canister materials. The chemical environment in the canister is converted to oxidizing conditions. Some of the nuclides (Np, Tc, U) have a higher solubility at oxidizing conditions than at reducing conditions. The oxidizing agents, they may be hydrogen peroxide, oxygen or oxidized uranium (VI), and the dissolved radionuclides will diffuse into and through the surrounding clay barrier and then out to the flowing water. The region of oxidizing conditions expands as more of the oxidizing species move out into the bedrock, where normally reducing conditions prevail. The oxidizing species are consumed by the oxidization of reducing components (mainly Fe(II)) in the bedrock. The oxidizing and reducing species can not coexist, as the reaction is very fast. At a distance from the canister all of the oxidizing species are consumed, and the oxidizing conditions are suddenly changed to reducing conditions. There is the redox front which slowly moves with the addition of the oxidizing species. The nuclides with lower solubility at reducing conditions than at oxidizing conditions precipitate to their saturation concentation. The concentration profile of the propagating nuclides is shown in figure 1.

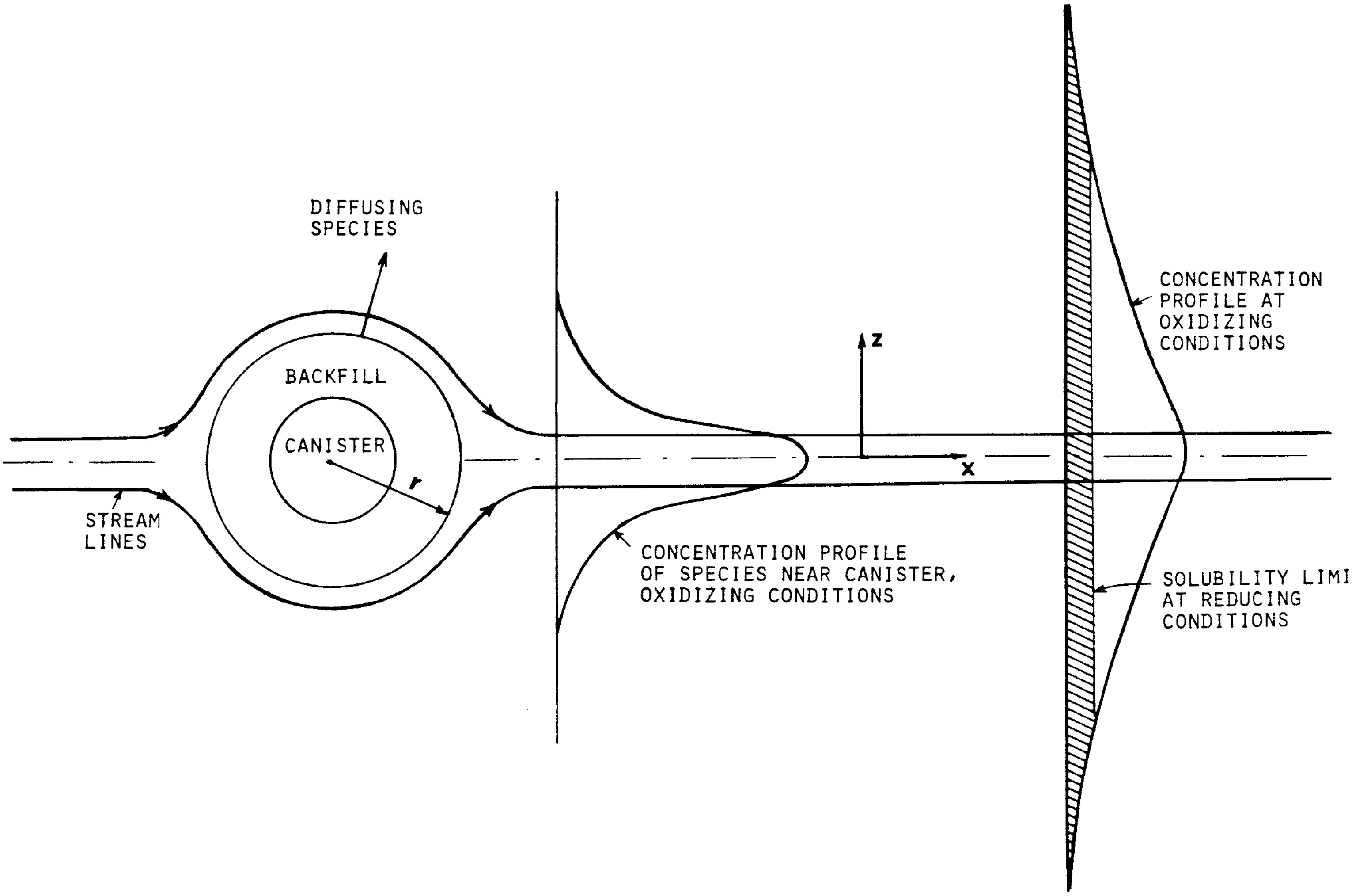


Figure 1. The concentration profile of a species near the canister widens as it moves downstream. Part of the nuclide mass precipitates at the redox front.

2. MODEL

The water flows past the clay barrier that surrounds the canister and further in a fissure with infinite extention in the fissure plane. The velocity of the water in the fissure is assumed to be uniform and can be written

$u_p = u_0/\varepsilon$

u_0 = flow rate of water per unit area of rock

$\varepsilon = \frac{\delta}{s}$

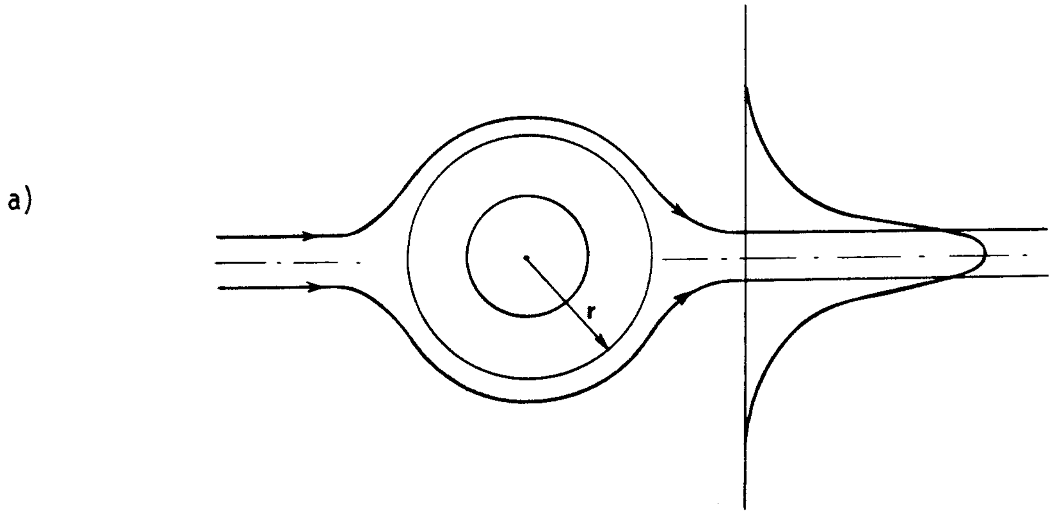
δ = fissure width

s = distance between fissures

It is assumed that there are no nuclides in the water upstream of the canister. The nuclides diffuse out of the clay and into the water, where a concentration profile transverse to the flow direction is developed (see figure 2a). The diffusion in the flow direction is neglected. The concentration of the nuclides is uniform across the fissure at every point in the fissure plane.

The curvature of the clay cylinder is neglected in the computation of the concentration profile. The cylinder is approximated by a plate with the length of $\pi \cdot r$, as is shown in figure 2b. The concentration of the nuclides is constant along the plate wall.

The instationary phase for the build up of a concentration profile in the clay is neglected, only the stationary case is considered.



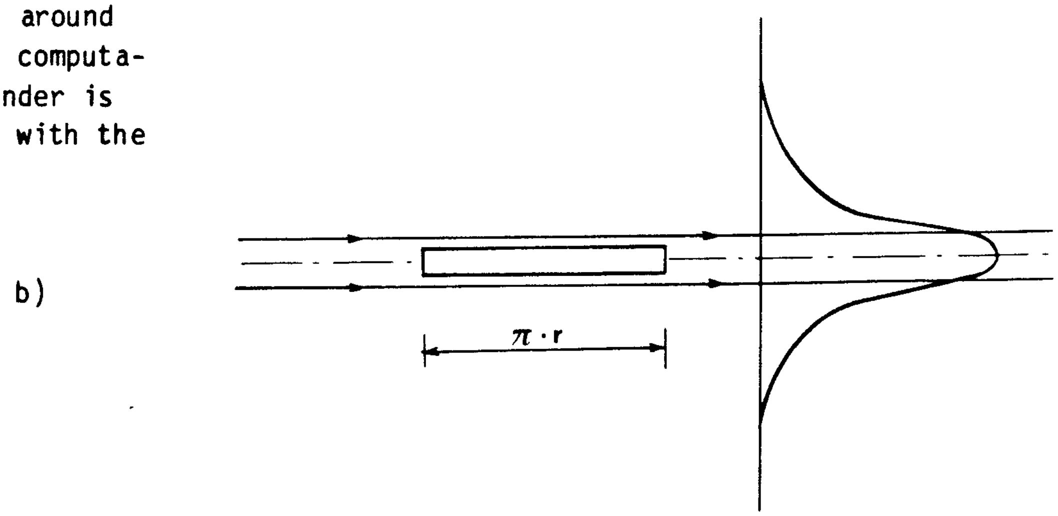


Figure 2. The deveolpment of a concentation front around a leaking canister. For computational reasons the cylinder is approximated by a plate with the same surface area.

The flow of the nuclides from the plate can be written:

$$N_{To} = \delta\, u_p \int_{-\infty}^{\infty} c\Big|_{x=x_o} dz = 2\delta\, u_p \int_{0}^{\infty} c\Big|_{x=x_o} dz \qquad (1)$$

(Symmetry around z=0)

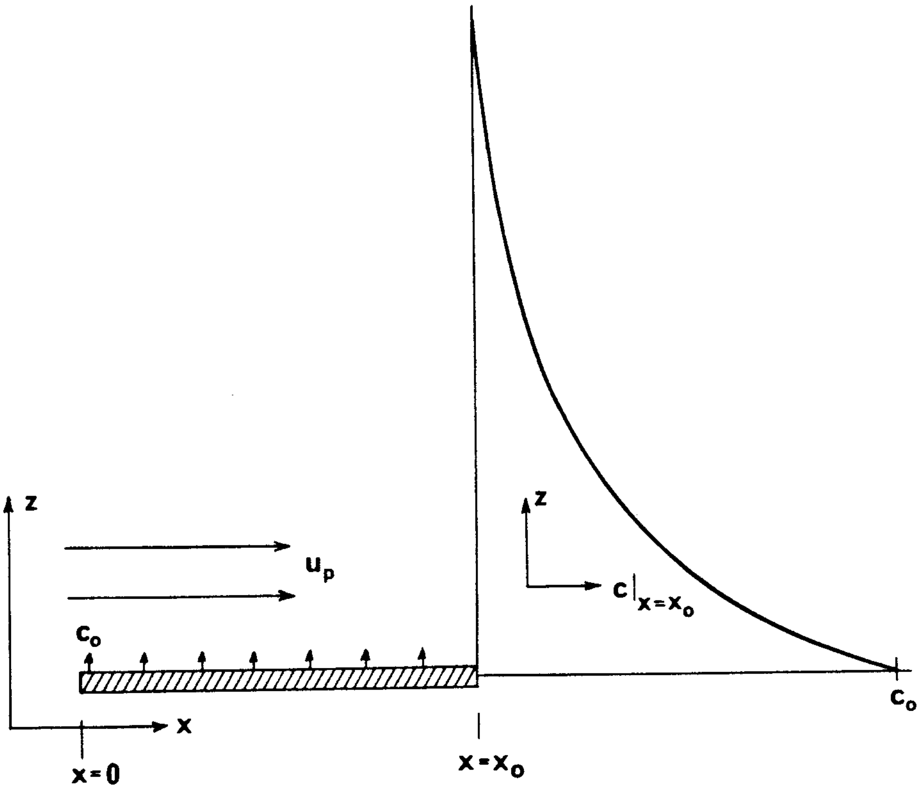


Figure 3. The development of a concentration profile at the surface of the plate.

When the nuclides reach the redox front, there is a precipitation of those nuclides with lower solubility in reducing waters.

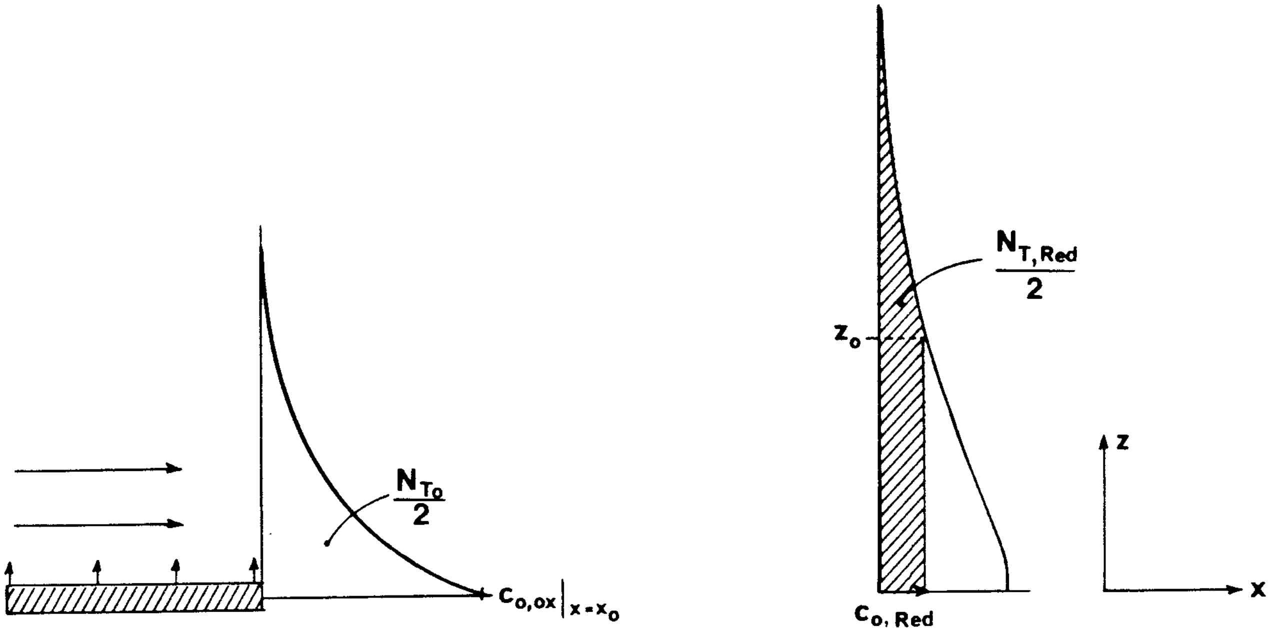


Figure 4. Immediately beyond the redox front the water carries radionuclides in a concentration less than or equal to their solubility (the shaded area).

The concentration of the nuclides in the water decreases to $c_{o,Red}$ at the redox front. It is constant between $-z_o$ and $+z_o$, outside these bounds it decreases, see figure 4. The nuclide flow in the fissure at reducing conditions just past the redox front is given by:

$$N_{T,Red} = 2\delta\ u_p \int_0^\infty c_{Red}\ dz = 2\delta\ u_p\ (z_o\ c_{o,Red} + \int_{z_o}^\infty c_{Red}\ dz) \qquad (2)$$

The ratio between the flow at reducing conditions and at oxidizing conditions is given by

$$\frac{N_{T,Red}}{N_{To}} = \frac{z_o\ c_{o,Red} + \int_{z_o}^\infty c_{Red}\ dz}{\int_0^\infty c|_{x=x_o} dz} \qquad (3)$$

which expresses the part of the diffusing nuclides from the plate that is transported beyond the redox front.

In the computation of the concentration profile along the plate in figure 3, the flow and diffusion equation for stationary conditions (4) is used.

$$u_p \cdot \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial z^2} \tag{4}$$

This equation can be rewritten in a new form with the dimensionless variables

$$X = \frac{x}{x_o} \tag{5}$$

and

$$Z = \frac{z}{\sqrt{\frac{D \cdot x_o}{u_p}}} \tag{6}$$

to give

$$\frac{\partial c}{\partial X} = \frac{\partial^2 c}{\partial Z^2} \tag{7}$$

For $c = c_0$ at $0 < X < 1$ and $Z = 0$

$c = 0 \qquad X < 0$
$c = 0 \qquad Z = \infty$

the solution is (Carslaw and Jaeger)

$$\frac{c}{c_o} = \text{erfc}\,\frac{Z}{2} \tag{8}$$

Downstream of the plate, the conditions for a solution of eq. (7) is given by

$$c = c_o \operatorname{erfc} \frac{Z}{2} \qquad \text{at } X = 1$$

$$\left.\frac{dc}{dZ}\right|_{Z=o} = 0 \qquad \text{at } X > 1$$

$$c \Longrightarrow 0 \quad \text{when} \quad Z \Longrightarrow \infty$$

We have chosen to produce a solution with the integrated finite difference method. TRUMP (Edwards, 1972).

The calculations give the concentration profile of the nuclides, at different distances downstream. This forms the basis that is needed for a calculation of the amount of nuclides which are transported past the redox front (eq. 3). Computations are made for different solubility ratios ($c_{o,Red}/c_{o,ox}$) and at various lengths downstream the plate.

3. RESULTS

The results of the computations are given in diagram 1-3.

Diagram 1 gives the width, Z_o, where the water has the solubility concentration $c_{o,Red}$ and when the redox front is at the distance X downstream the plate. The solubility ratio $c_{o,Red}/c_{o,ox}$ is given as a parameter. In dimensional quantities

$$z_o = Z_o \sqrt{\frac{D x_o}{u_p}} \tag{9}$$

and

$$x = X \cdot x_o \tag{5}$$

These results can be used to calculate the flux of various nuclides past the redox front as this moves slowly downstream.

In diagram 2 $\frac{N_{T.Red}}{N_{To}}$ is given as a function of X. at different $c_{o.Red}/c_{o.ox}$.

The variation of the nuclide concentration with X. at Z = 0. is shown in diagram 3.

3.1 Sample calculations

With $u_p = 3.15 \cdot 10^{-8}$ m/s (1 m/year), $x_o = \pi \cdot r = 2.36$ m (r=0.75 m) and $D = 2 \cdot 10^{-9}$ m^2/s equation (9) simplifies to

$$z_o = 0.387 \cdot Z_o$$

Solubilities of some nuclides at reducing and oxidizing conditions are given in table 1 (Allard, 1982).

Table 1. Solubilities of the actinides and technetium.

Nuclide	Reducing water (mg/l)	Oxidizing water (mg/l)	$\frac{c_{o,Red}}{c_{o,ox}}\Big\|_{\text{max solubility}}$
U	0.001 - 1	950*	10^{-6} - 10^{-3}
Pu	3	$8 \cdot 10^{-3}$	> 1
Np	$8 \cdot 10^{-3}$	630*	$1.3 \cdot 10^{-5}$
Np	-"-	2.1**	$3.8 \cdot 10^{-3}$**
Th	$4 \cdot 10^{-4}$	$4 \cdot 10^{-4}$	1
Am	6	6	1
Tc	$2 \cdot 10^{-4}$	0.64**	$3.1 \cdot 10^{-4}$**

*) Is determined by the carbonate concentration.

**) The concentration at congruent dissolution of the fuel where $c = N_T/Q_{eq}^{ox}$ (Q_{eq}^{ox} = 1.6 l/canister,year; $N_T(Np) = 3.39 \cdot 10^{-3}$ g/year and $N_T(Tc) = 1.03 \cdot 10^{-3}$ g/year). The neptunium and technetium are dissolved in proportion to their concentration in the fuel. In this example the neptunium content = $2.22 \cdot 10^{-3}$ fractions by weight and the technetium content = $6.74 \cdot 10^{-4}$. The fuel consists of 95 % uranium which determines the dissolution by dissolving to its equilibrium concentration in 1.6 l/canister,year.

The values of $c_{o,Red}/c_{o,ox}$ in table 1 for Np, U and Tc are used to get the ratios of $N_{T,Red}/N_{To}$ in diagram 2. These ratios are tabulated in tables 2-4.

Table 2. Flow of Np^{237} in reducing water.

	Maximum solubility*			Congruent dissolution of matrix**		
X	$\frac{N_{T,Red}}{N_{To}}$	$N_{T,Red}$ (g/year)	(µCi/year)	$\frac{N_{T,Red}}{N_{To}}$	$N_{T,Red}$ (g/year)	(µCi/year)
3	$1.4 \cdot 10^{-4}$	$1.4 \cdot 10^{-4}$	0.099	$2.5 \cdot 10^{-2}$	$8.6 \cdot 10^{-5}$	0.061
10	$2.6 \cdot 10^{-4}$	$2.6 \cdot 10^{-4}$	0.18	$4.5 \cdot 10^{-2}$	$1.5 \cdot 10^{-4}$	0.11
30	$4.3 \cdot 10^{-4}$	$4.3 \cdot 10^{-4}$	0.31	$7.3 \cdot 10^{-2}$	$2.5 \cdot 10^{-4}$	0.18

*) N_{To} = 1.0 g/year with Q_{eq}^{ox} = 1.6 l/year and c^{ox} = 0.63 g/l

**) $N_{To} = 3.39 \cdot 10^{-3}$ g/year.

Table 3. Flow of U in reducing water.

X	$\frac{N_{T,Red}}{N_{To}}$	$N_{T,Red}$ (g/year)
3	$1.1 \cdot 10^{-4} - 8.2 \cdot 10^{-3}$	$1.7 \cdot 10^{-4} - 1.3 \cdot 10^{-2}$
10	$2.1 \cdot 10^{-4} - 1.5 \cdot 10^{-2}$	$3.2 \cdot 10^{-4} - 2.3 \cdot 10^{-2}$
30	$3.4 \cdot 10^{-4} - 2.4 \cdot 10^{-2}$	$5.2 \cdot 10^{-4} - 3.7 \cdot 10^{-2}$

$N_{T,Red}/N_{To}$ is calculated for $c_{o,Red}/c_{o,ox}$ between 10^{-5} and 10^{-3}
N_{To} = 1.52 g/year.

Table 4. Flow of Tc^{99} in reducing water.

X	$\frac{N_{T.Red}}{N_{To}}$	$N_{T.Red}$ (g/year)	(µCi/year)
3	$2.7 \cdot 10^{-3}$	$2.8 \cdot 10^{-6}$	0.048
10	$4.8 \cdot 10^{-3}$	$5.0 \cdot 10^{-6}$	0.086
30	$7.9 \cdot 10^{-3}$	$8.2 \cdot 10^{-6}$	0.14

$N_{To} = 1.03 \cdot 10^{-3}$ g/year

4. DISCUSSION AND CONCLUSIONS

The analysis is simplified by not accounting for the diffusion into the matrix of the nuclides. This exaggerates the distance of the transverse diffusion and thus the flux of the nuclides past the redox front.

In the example the data used are within the range of expected values for a repository in "good" crystalline rock. With these data x_0 is 2.4 m and $z_0 = 0.4$ m. Diagram 1 indicates that at X = 2 (X = 1 at the downstream side of the canister) i.e. 2.4 m downstream from the canister, Z = 3.0 - 9.0 and z = 1.2 - 3.5 m. This means that the precipitation width - $2z_0$ - is twice this value i.e. 2.4 - 7.0 m. For a shortest distance between the canisters equal to 6 m, the precipitation fronts from two neighbouring canisters may meet if the redox front is some 10 to 10:s of meters downstream from the canisters.

Under such conditons most water downstream from a repository may attain the solubility concentration under reducing conditions.

5. NOTATION

c	concentration of nuclide	(kg/m^3)
c_0	concentration at the clay boundary	(kg/m^3)
$c_{0,ox}$	concentration at $x = x_0$ and $z = 0$ in oxidizing water	(kg/m^3)
c_{Red}	concentration in reducing water	(kg/m^3)
$c_{0,Red}$	solubility in reducing water	(kg/m^3)
D	diffusivity	(m^2/s)
N_{To}	flow of nuclides in oxidizing water of $x = x_0$	(kg/s)
$N_{T,Red}$	flow of nuclides in reducing water	(kg/s)
r	radius of canister + clay barrier	(m)
s	distance between fissures	(m)
u_0	flow of water per unit area of rock	(m/s)
u_p	velocity of water in the fissure	(m/s)
x	distance in flow direction	(m)
x_0	length of plate = $\pi \cdot r$ and half circumference of clay cylinder	(m)
X	x/x_0	
z	distance transverse to flow direction in the plane	(m)
Z	$z/\sqrt{\frac{D\,x_0}{u_p}}$	
z_0	distance in z from $z = 0$, in reducing water, to where c_{Red} begins to decrease	(m)
ε	porosity of bedrock	
δ	mean fissure width	(m)

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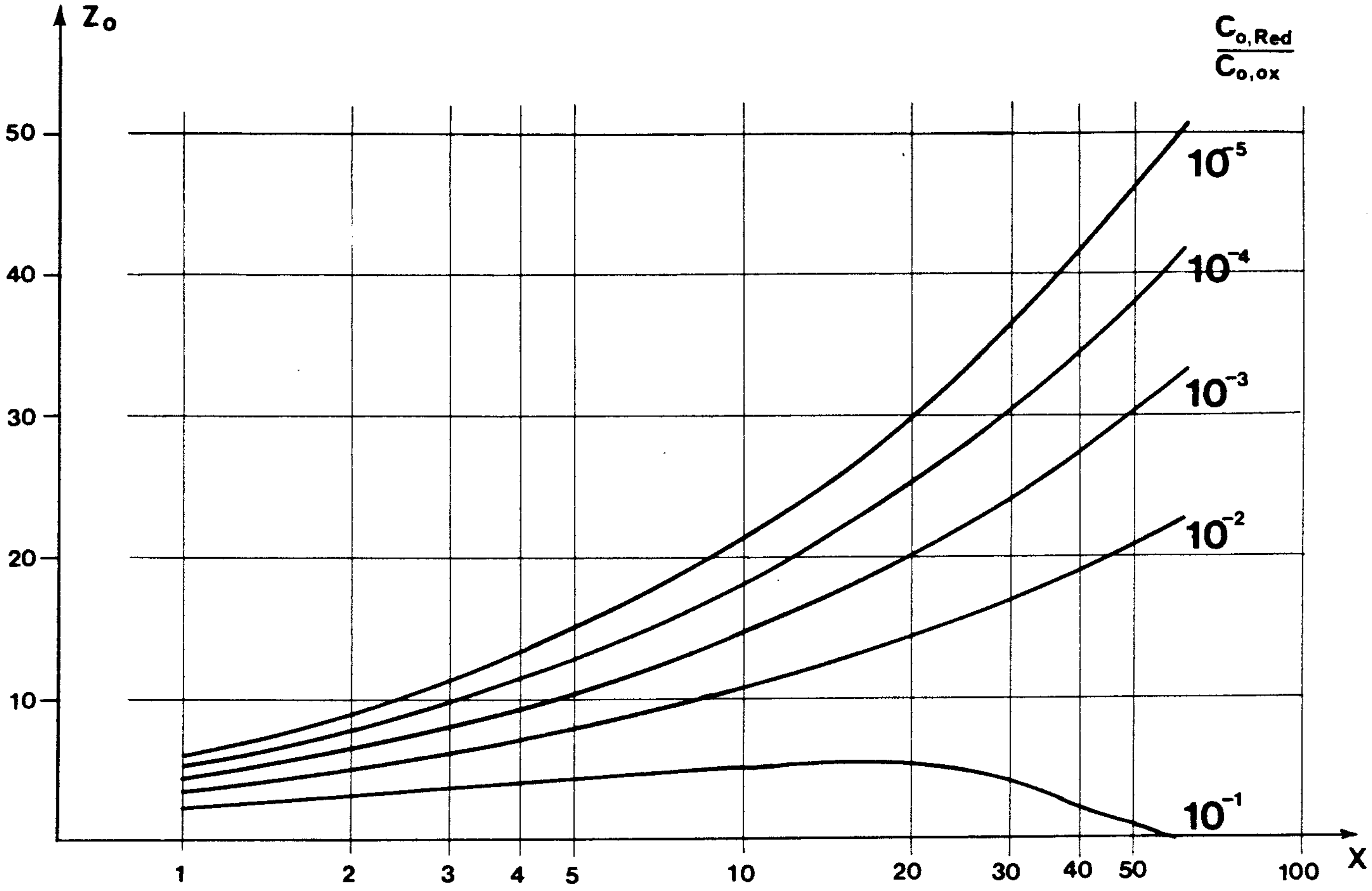

Diagram 1: At the redox-front some nuclides precipitate to give a constant nuclide concentration ($c_{0,Red}$) in the water along a distance (z) transverse to the water flow. In the diagram this distance is given in a dimensionless form (Z_0) and taken from Z = 0. Z_0 is a function of the dimensionless distance downstream from the plate (X) for different solubility ratios.

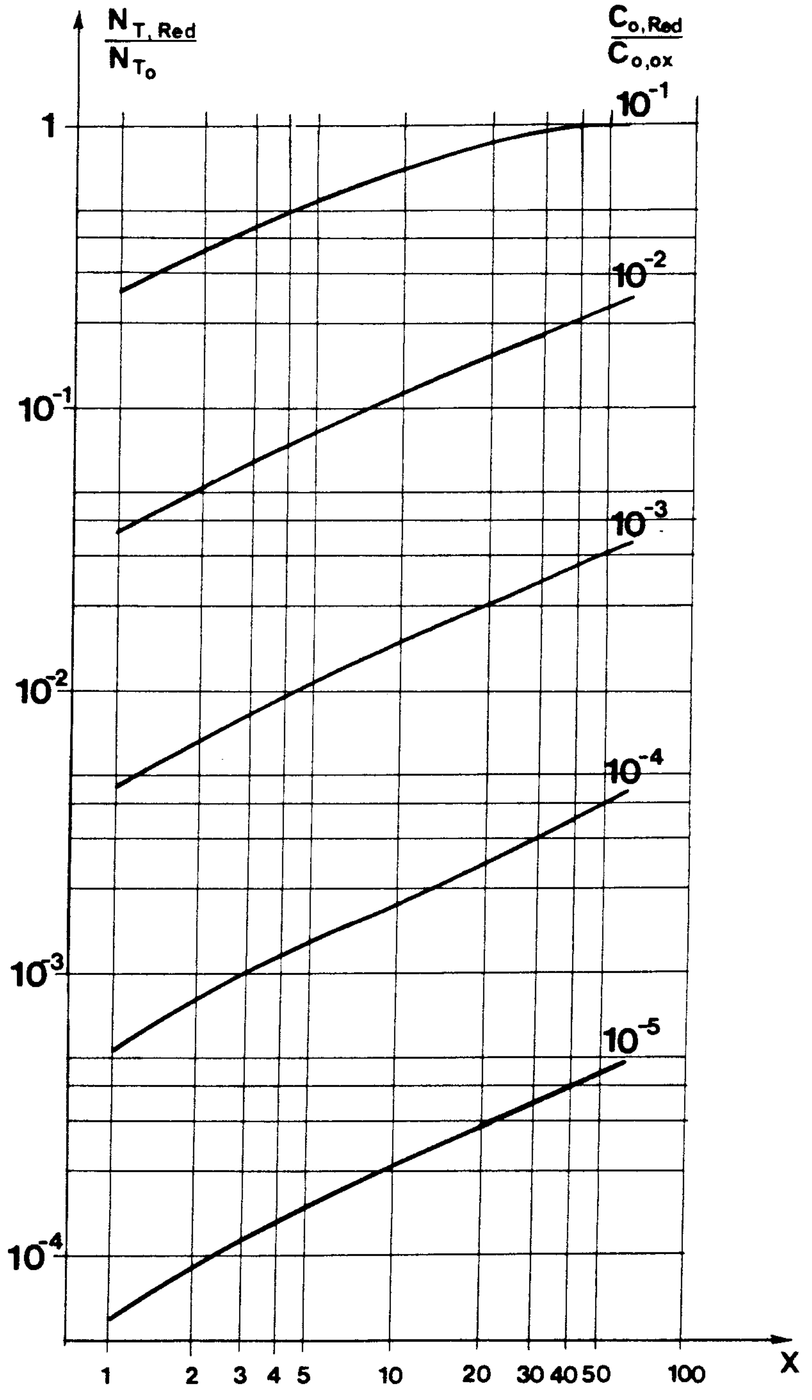

Diagram 2· The part of the nuclides that is transported beyond the redox-front is given as a function of the dimensionless distance downstream from the plate. for different solubility ratios.

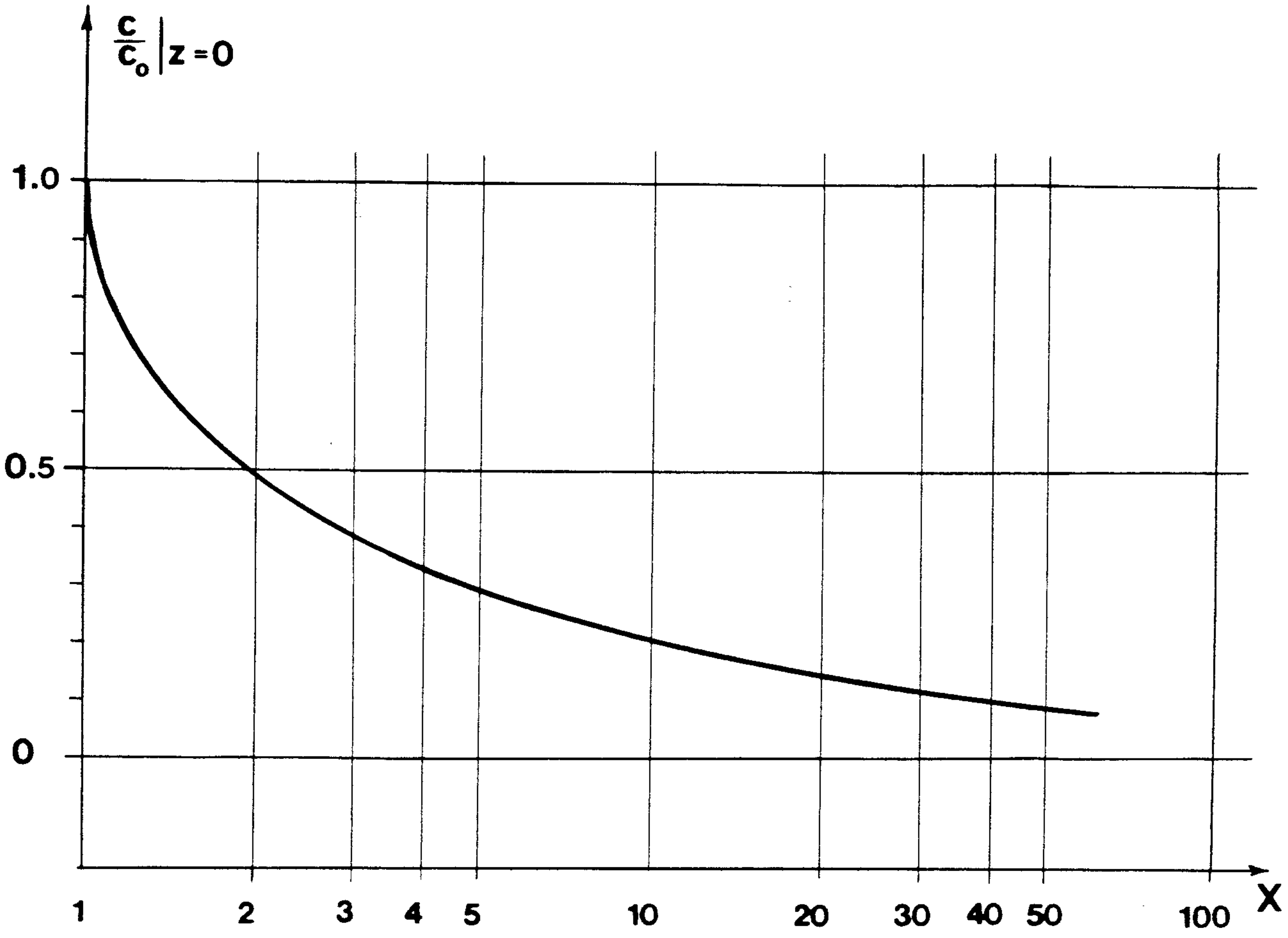

Diagram 3: The nuclide concentration in a dimensionless form, along centerline ($Z = 0$), is given as a function of the dimensionless distance downstream from the plate.

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