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**Diffusivities of some dissolved
constituents in compacted wet
bentonite clay -MX80 and the
impact on radionuclide migration
in the buffer**

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Stockholm, Sweden 1982-10-29

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DIFFUSIVITIES OF SOME DISSOLVED CONSTITUENTS IN
COMPACTED WET BENTONITE CLAY -MX80 AND THE IMPACT
ON RADIONUCLIDE MIGRATION IN THE BUFFER

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This report concerns a study which was conducted for SKBF/KBS. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

A list of other reports published in this series during 1982 is attached at the end of this report. Information on KBS technical reports from 1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26) and 1981 (TR 81-17) is available through SKBF/KBS.

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Summary

The diffusivities measured by various investigations of several species in compacted bentonite clay have been compiled and analysed. Small anions diffuse slower than an uncharged molecule such as methane. Large anions move orders of magnitude slower still. The actinides Th, U, Pu, Np and Am are considerably retarded by sorption effects. Their movement can be explained by pore diffusion with retardation. Cs, Sr and Pa move considerably faster than can be explained by these effects. The faster mobility is probably due to surface migration.

A simplified model is presented by which the importance of the backfill barrier in retarding the radionuclides can be assessed. It is based on the computation of the evolution of the concentration profile of the diffusing nuclide in the backfill. The flowrate out from the backfill to the flowing water can be compared to the inflow into the backfill due to leaching.

Using the previously obtained diffusivities it is found that a 0.375 m thick backfill will eventually let through all I^{129} , Tc^{99} , Ra^{226} , Pa^{231} , U^{234} , U^{235} , U^{238} and Np^{237} . The maximum release rate for Cs^{137} , Sr^{90} , Pu^{239} , Pu^{240} and Am^{243} will decrease by one to three orders of magnitude compared to the leach rate. Am^{241} will decay to insignificance in the backfill.

Background and purpose

In the Swedish concept of a repository for spent nuclear fuel the canisters are surrounded by a compacted bentonite backfill. The hydraulic conductivity of the backfill is so low that water movement in it is very slow. Dissolved species such as radionuclides move very much faster by molecular diffusion in the backfill than by flow. Most of the nuclides of interest are cationic or neutral and will be retarded by adsorption or ion exchange in the backfill. Nuclides with short halflives and strong retardation may decay considerably during their passage through the backfill.

This study aims at finding a simple method to assess which nuclides will and which will not be considerably hindered by the backfill.

Diffusivities of some dissolved constituents in compacted wet bentonite clay - MX80

Several investigations have been made regarding diffusion in compacted bentonite.

Two techniques are used, one stationary and one instationary. In the first technique a bentonite slab compressed between two porous discs is contacted with a solution containing tracer on one side of the slab. The other side is constantly flushed with tracer free (or practically free) water. At stationary conditions the flux N of tracer through the slab is determined by the pore diffusivity D_p , the concentration gradient $\frac{dc}{dz}$ and the surface area available for diffusion $A \cdot \epsilon_p$. A is the geometric area of the disc and ϵ_p is the fraction of this surface which consists of water - the porosity.

Fick's law can be used to determine the pore diffusivity.

$$N = -D_p \epsilon_p A \cdot \frac{dc}{dz} \quad (1)$$

In the other experiment the surface of a wet compacted bentonite slab is contacted with a solution containing tracer at time zero. The tracer diffuses into the slab which after some time is sliced in thin sections and the concentration profile is determined. There are two variations to this technique. In one, the tracer solution is fully soaked into one thick slab. A second slab is compressed onto this slab. The tracer diffuses from the first and into the other. Another way is to drip a concentrated solution onto the surface of one slab and immediately contact this surface with another fresh slab. The tracer will move from the common interface symmetrically out into both slabs. Profiles are again measured. These techniques utilize the instationary phase of the experiment. The profile techniques are preferably used for substances of low diffusivities or substances exhibiting strong sorption, for which it would take a very long time to make a "diffusion through" experiment.

The governing equation in the instationary experiments is

$$\frac{\partial c}{\partial t} = \frac{D_p}{1+K_d \rho} \cdot \frac{\partial^2 c}{\partial z^2} \quad (2)$$

where K_D is the sorption equilibrium constant and ρ is the density of the compacted clay.

$D_p/(1+K_D\rho)$ is often called D_a the apparent diffusivity.

With the appropriate initial and boundary conditions the solutions for the concentration profiles are (Carslaw and Jaeger 1959).

For the diffusion from one saturated slab to the other

$$c/c_o = 0.5 \left\{ \operatorname{erf} \left\{ \frac{(1-z/a)}{2\sqrt{\frac{D_a t}{a^2}}} \right\} + \operatorname{erf} \left\{ \frac{(1+z/a)}{2\sqrt{\frac{D_a t}{a^2}}} \right\} \right\} \quad (3)$$

where a is the thickness of the impregnated slab.

For the diffusion from the joint surface into both slabs (of large extension) (Carslaw and Jaeger 1959).

$$c = \frac{m}{2A\sqrt{\pi D_a t}} \cdot e^{-\frac{z^2}{4D_a t}} \quad (4)$$

From these equations D_a is determined. ϵ_p and $K_D\rho$ must be determined separately to compare the stationary and instationary techniques.

For many porous structures there is a direct relation between the diffusivity of a substance in bulk water, D_v , and in water contained in pores, D_p

$$D_p = \frac{D_v \delta_D}{\tau^2} \quad (5)$$

where δ_D is the constrictivity
and τ^2 is the tortuosity.

The term δ_D/τ^2 is called the geometric factor and is generally a quantity specific for a porous structure. It is determined from the ratio D_p/D_v .

Table 1 is a compilation of measured diffusivities for a variety of compounds in compacted bentonite. The density of the wet bentonite has been 2000-2100 kg/m³. The porosity is about 0.35 at this compaction.

All the species except the large molecular weight species have diffusivities in bulk water of $1-3 \cdot 10^{-9}$ m²/s. If there were no interaction effects between the species and the clay except those measured as sorption capacities K_p , the values in the column " $D_p \epsilon_p$ " should lie within a factor 1-3 for the small molecules and ions. This is not the case. The gases H₂ and CH₄ have much higher $D_p \epsilon_p$ than Cl⁻ and I⁻ which in turn have a higher diffusivity than HS⁻. In the latter case it must be expected that some reaction has occurred. The sulfide is known to react with iron which is present in the clay. The sorption capacity of HS⁻ in the bentonite is not available.

The results compiled in table 1 show that there are other mechanisms active than those accounted for in the analysis because the geometric factor δ_D/τ^2 is not constant. For Sr and Cs there is some mechanism that considerably speeds up the migration as δ_D/τ^2 is larger than 1. Pore diffusion only, cannot account for this. One speeding up mechanism is surface diffusion (Rasmuson 1982). This is not well understood at present. The diffusivity of hydrogen is lower than that of methane which also is unexpected because hydrogen is a smaller molecule. The independent measurements on I⁻ and Cl⁻ where stationary as well as instationary techniques have been used, differ by nearly an order of magnitude. Eriksen et al. (a) found a lower concentration gradient in the pore liquid of I⁻ and Cl⁻ than expected from the bulk concentration difference over the disc. This may be due to the very small size of the pores and the negative charge of the pore surface which hinders negatively charged species to accumulate. The difference is about a factor 2 according to Eriksen et al. (a). There is, however, an even larger discrepancy between the instationary and the stationary experiments on I⁻ and Cl⁻. For I⁻ the instationary measurement gives a D_p value of $4 \cdot 10^{-12}$ m²/s whereas the stationary experiment gives a $D_p \epsilon_p$ value of $0.2 \cdot 10^{-12}$ m²/s. This would give a porosity of 0.05 which is 6-7 times lower the actual porosity. The same figure emerges for Cl⁻. The effective water volume for transporting negative ions thus seems to be smaller than the actual water volume.

For larger molecules with negative charges AQS⁻ (Sodium anthraquinone-2-sulfonate) with a molecular weight of 354, Eosin (Disodium 2,4,5,7-Tetra-bromofluorescein) with a molecular weight of 646 and the lignosulfonate molecule fractions with molecular weights of 5600, 12000, 24000 and 30000, the diffusivities are 1-2 orders of magnitude lower than for I⁻ and Cl⁻.

Although there is fair agreement between diffusivities for the actinides Th, U, Np, Pu and Am if the geometric factors of the pore diffusivities are compared, the value for Pa differs considerably. The high geometric factor for Pa indicates that there may be considerable migration due to surface diffusion.

Transport of decaying radionuclides from spent fuel through the backfill - Instationary period.

The migration of the radionuclides in the backfill takes place in a fairly complex geometry. The nuclides will migrate from the spot where they are dissolved out into the backfill and then from the backfill into those sections of the rock which are intersected by fissures carrying water. A three dimensional treatment of the transport in the backfill by numerical methods has been done by Andersson et al. (1982) assuming that all the canister surface has the same reactivity and that fissures intersect the repository hole in a regular fashion. The main emphasis of this treatment was on the steady state part of the transport.

Although this work treated the transport of corrosive agents in to the canister, the mathematics for the corrosion case and those for the leaching case are identical. It could be concluded that the main resistance to transport is in the slowly moving fluid in the bedrock when steady state has been reached. During steady state the transport from the fuel surface is proportional to the difference in concentration between the surface of the fuel and for concentration in the water approaching the waste.

Here we want to study the early part of the leaching when the concentration profile is building up in the backfill. The main matrix constituent, be it silica from vitrified waste or uranium from spent fuel, is assumed to have reached a steady state earlier than the sorbing nuclides which are retarded during their transport. This is a fair approximation as neither silica nor uranium under oxidizing conditions will sorb strongly and thus will not be much retarded in the clay.

The sorbing nuclides will be considerably retarded during the instationary phase and if the time for first arrival at the outer boundary of the backfill is very large compared to the halflife of the nuclide, the nuclide may decay to insignificance before it reaches the outer boundary of the backfill.

A simplified treatment to assess the importance of the retardation is done below. The geometry of the backfill is taken to be a very thick slab of backfill. Nuclides are released into one side of the slab by congruent dissolution, and the transport rate of the nuclide past a plane at distance z_0 is calculated. As long as the transport rate past z_0 is very small the thick slab is a fair approximation of a slab with thickness z_0 with any reasonable boundary condition applicable to the subsequent transport in the bedrock.

In the following the treatment will be based on the dissolution of the spent fuel matrix. Vitrified waste can be treated in an analogous way.

When the radionuclides are leached from the spent fuel they will start to migrate through the backfill. Some of them like Sr will, except for a short initial phase, be released by congruent dissolution. This means that they will be released in proportion to their concentration in the fuel at any given moment. For all practical purposes the decay of the main matrix constituent U^{238} may be neglected in comparison to all other nuclides. The mass of the shorter lived nuclides will

decrease in the fuel due to decay according to $m = m_0 \cdot e^{-\lambda t}$ if they have no precursor. m_0 is the mass of nuclide i at the time of burial.

When the matrix dissolves it will attain an uranium equilibrium concentration at the surface of the fuel equal to the solubility $c_{U,so}$ of uranium in the ground water. The matrix dissolves away as components diffuse out through the backfill. The dissolution rate can be determined. This has been discussed in Neretnieks (1982). The rate for the U^{238} of the matrix is N_U . Other nuclides are released in proportion to their abundance in the fuel. Nuclide i is released with rate N_i

$$N_i = N_U m_i/m_U \quad (6)$$

In the following treatment it will be assumed that the nuclides do not have a solubility limitation which would make them precipitate on the fuel surface. All released nuclides are assumed to move into the backfill.

For brevity we omit the index for nuclide "i". The transport through the backfill is described by equation (7) for the case of a flat backfill barrier. (We neglect the curvature in this analysis.)

$$\frac{dc}{dt} = D_a \frac{\partial^2 c}{\partial z^2} - \lambda c \quad (7)$$

for a decaying species.

The initial conditions is $c = 0 \quad z > 0 \quad t = 0$

The boundary condition at $z = 0$ is

$$N = N_U \cdot m_o/m_U \cdot e^{-\lambda t} = D_p \epsilon_p A \left. \frac{dc}{dz} \right|_{z=0} \quad (8)$$

which indicates that all released nuclide is transported into the backfill. The uranium dissolution rate N_U is assumed to have become stationary and is thus constant.

Equation (8) is rewritten

$$\left. \frac{dc}{dz} \right|_{z=0} = b e^{-\lambda t} \quad (9)$$

where $b = N_U m_o / (m_U A D_p \epsilon_p)$.

Taking the Laplace transform of equations (7) and (8) we obtain

$$\bar{c}_p = D_a \cdot \frac{d^2 \bar{c}}{dz^2} - \lambda \bar{c} \quad (10)$$

and

$$\left. \frac{d\bar{c}}{dz} \right|_{z=0} = - \frac{b}{p+\lambda} \quad (11)$$

Equation (10) is rewritten

$$\frac{d^2 \bar{c}}{dz^2} = \bar{c} (p+\lambda) \frac{1}{D_a} = \bar{c} d^2 \quad (12)$$

which has a solution

$$\bar{c} = A e^{-dz} + B e^{+dz} \quad (13)$$

We want \bar{c} to be limited as $z \rightarrow \infty$ which gives $B = 0$. This implies that we model an infinitely thick backfill.

Differentiating equation (13) gives

$$\frac{d\bar{c}}{dz} = -A d e^{-dz} \quad (14)$$

and equating equations (14) and (11) for $z = 0$ gives

$$A = \frac{b}{(p+\lambda)d} \quad (15)$$

Then from (15) and (13)

$$\bar{c} = \frac{b}{(p+\lambda)d} \cdot e^{-dz} \quad (16)$$

The property of the Laplace transform that if $\mathfrak{L}\{c_1\} = \bar{c}_1(p)$ then $\mathfrak{L}\{c_1\} = \mathfrak{L}\{e^{\lambda t} c\} = \bar{c}(p+\lambda)$ gives with

$$d = \sqrt{(p+\lambda) \frac{1}{D_a}} \quad (17)$$

$$\bar{c}_1 = \frac{b}{p \sqrt{\frac{p}{D_a}}} \cdot e^{-\sqrt{\frac{p}{D_a}} z}$$

The inverse can be obtained directly from the table of transforms (Carslaw and Jaeger p. 495)

$$c = b \cdot e^{-\lambda t} z \left\{ \frac{2}{\sqrt{\pi}} \frac{\sqrt{D_a t}}{z} \cdot e^{-\frac{z^2}{4D_a t}} - \operatorname{erfc} \left(\frac{1}{2} \frac{z}{\sqrt{D_a t}} \right) \right\} \quad (18)$$

and for the gradient we obtain $\frac{dc}{dz}$ from (14)

$$\frac{d\bar{c}}{dz} = \frac{-b}{(p+\lambda)} e^{-dz} \quad (19)$$

$$\frac{dc}{dz} = -b \cdot e^{-\lambda t} \cdot \operatorname{erfc} \frac{z}{2\sqrt{D_a t}} \quad (20)$$

and thus $N/z=z_0$ is

$$N/z=z_0 = b D_p \epsilon_p A \cdot e^{-\lambda t} \operatorname{erfc} \frac{z_0}{2\sqrt{D_a t}} \quad (21)$$

$$N/z=z_0 = N_U \cdot m_o/m_U \cdot e^{-\lambda t} \cdot \operatorname{erfc} \frac{z_0}{2\sqrt{D_a t}} \quad (22)$$

The rate of transport of radionuclide "i" past z_0 as a fraction of that entering the backfill at $z = 0$. and $t = 0$ is

$$N_{z_0}/N_0 = e^{-\lambda t} \operatorname{erfc} \frac{z_0}{2\sqrt{D_a t}} \quad (23)$$

Equations (18) and (23) consists of two parts. The first part accounts for the radioactive decay. The second part describes the change of concentration and flux of a stable species with time and distance. The convenient separability of the solution is due to the presence of the same decay term in equation (7) as well as in the boundary condition at the inlet. equation (8). If the importance of outlet boundary condition is deemphasized, i.e. mainly the early times are considered. we can study the nondecaying part of the solution independently of the decaying part.

The treatment above has considered an infinitely thick backfill barrier and linear transport of the diffusing species. The backfill in a repository will have finite length and cylindrical shape more than slab shape.

Because of the finite thickness the boundary condition at $z = z_0$ would better be described by a flux boundary condition $N_{z=z_0} =$

$$-D_p \epsilon_p \cdot A \cdot \frac{dc}{dz} / z=z_0 = Q_{eq} \cdot c_{z=z_0} \quad \text{where } Q_{eq} \text{ is the equivalent water flow}$$

rate which transports the nuclide into the flowing water in the bedrock. For a stable species a solution for this boundary condition is available (Carslaw and Jaeger p. 126). It is fairly complicated to evaluate and is not suited for calculations of the early breakthrough.

The same treatment has been applied to a cylindrically symmetric case with radial transport of the radionuclide which enters at r_1 and is observed at r_2 ($r_2 > r_1$) (Carslaw and Jaeger p. 332). This solution also includes the flux and concentration boundary conditions at r_1 as well as r_2 . It is also complicated to evaluate and not suited for calculations for short times.

An indication on the differences due to the outlet boundary conditions and due to geometry can be obtained for short times by comparing the time to obtain a given concentration at $z=z_0$ for a nondecaying species with a constant concentration at the inlet boundary $c_{z=0} = c_0$; $t > 0$. For a given ratio c_z/c_0 the results can be expressed as

$$t_{0.05} = \text{const}_{0.05} \cdot z_0^2 / D_a \quad (24)$$

The index 0.05 indicates that the concentration at z_0 is $c_z = 0.05 c_0$. For linear transport for the $c = 0$ boundary condition at infinity $\text{const}_{0.05} = 0.13$ (Carslaw and Jaeger p. 60) and for a zero flux boundary condition at $z = z_0$ $\text{const}_{0.05} = 0.10$ (Carslaw and Jaeger p. 101). For smaller concentrations at $z = z_0$ the differences decrease further.

For transport radially out in a cylinder with $r_2 = 2r_1$ and $z_0 = r_2 - r_1$. the constant in equation 24 for the $c = 0$ at infinity $\text{const}_{0.05} = 0.15$ (Carslaw and Jaeger p. 335) and for the zero flux boundary condition at $r = r_2$ the $\text{const}_{0.05} = 0.11$. (Numerical solution by integrated finite differences.)

For screening purposes and for first estimates it is deemed to be sufficient to use the solutions for any of the geometries and boundary conditions. For $c_z/c_0 < 0.05$ the results differ very little.

Some calculated results

Figure 1 shows N/N_0 for Pu^{239} as a function of time after start of leaching. The figure also shows the release in a case where there is no retardation in the buffer. The peak release (5.4 % of original maximum leach rate) takes place after 40 000 years. At this time decay accounts for a decrease by a factor of 5.3 and the retardation effect contributes with another factor 3.5. The latter is somewhat on the low side to assure the applicability of equation (23). It somewhat underestimates the peak height and the portion on the downhill side. Figures 2-6 show the transport curves for Pu^{240} , Sr^{90} , Cs^{137} , Am^{243} and Th^{229} .

For screening purposes and for visualization the entity breakthrough time " $t_{0.05}$ " is introduced. That is the time for which $\text{erfc}(z_0/2\sqrt{D_a t}) = 0.05$. It indicates the time after start of leaching for the nuclide flux at the outer edge of the backfill to become 5 % of the influx, provided the nuclide would not decay. This time can be expressed as a number of halflives. If the number of halflives is more than a given value e.g. 30, it can be said that the nuclide has decayed to insignificance in the backfill.

Table 4 shows breakthrough times $t_{0.05}$ through a cylindrical backfill barrier with $r_1 = 0.375$ m and $r_2 = 0.75$ m. The constant in equation 25 is 0.107 for this case.

The only nuclide of those considered which decays to insignificance is Am^{241} . Am^{243} , Pu^{239} , and Pu^{240} will be noticeably retarded. Sr^{90} and Cs^{137} will be somewhat retarded. Th^{229} comes predominantly from the Np^{237} chain which passes the backfill barrier and will thus not be influenced by the backfill.

Discussion and conclusions

Although there are still some effects that are not fully understood it can be concluded that Cs^+ and Sr^{2+} migrate much faster than can be explained by pore diffusion theory. By analogy this is also assumed to apply to other alkali and alkaline earth elements. Protactinium migration also seems to be enhanced by surface diffusion. Small anions are somewhat restricted in their movement through the pores. Large anions are severely restricted in their movement. This would apply to colloids as well, because of their size and low diffusivity in water. The movement of actinides Th, U, Np, Pu and Am can be explained by pore diffusion with a geometric factor of 0.05-0.2.

Only Am^{241} decays to insignificance in the barrier. Am^{243} has 4 residence times and thus may decay a few orders of magnitude. Due to the large uncertainties in the diffusivity values this is not deemed to be of large significance.

Pu^{239} , Pu^{240} , Cs^{135} and Sr^{90} decay somewhat in the barrier. Th^{229} originally present in fuel also decays somewhat. Most of the Th^{229} will be produced from the decay chain with Np^{237} which passes the barrier. The Th^{229} will thus not be influenced by the barriers.

Notation

A	cross section area	m^2
a	thickness of slab	m
c	concentration	mol/m^3
D_a	apparent diffusivity	m^2/s
D_p	pore diffusivity	m^2/s
D_v	diffusivity in unconfined fluid	m^2/s
K_d	equilibrium constant	m^3/kg
m	amount of tracer	mol
N	mass flow rate	mol/s
Q_{eq}	equivalent flow rate of water	m^3/s
r	radial distance	m
t	time	s
z	distance in flow direction	m
δ_D	constrictivity	-
ϵ_p	porosity of backfill	-
λ	decay constant	s^{-1}
ρ	density of backfill	kg/m^3
τ	tortuosity	-

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Compound	Reference	$D_a \cdot 10^{12}$ measured values m^2/s	K_d m^3/kg	$D_p \cdot 10^{12}$ from D_a m^2/s	$D_{p \epsilon_p} \cdot 10^{12}$ m^2/s	$D_v \cdot 10^9$ m^2/s	δ_D/τ^2 evaluated with $\epsilon_p = 0.35$
I^-	a)	4 ⁱ⁾	0	4	0.2 ^{s)}	1.5*	$0.38-2.7 \cdot 10^{-3}$
Cl^-	a)	6 ⁱ⁾	0	6	0.3 ^{s)}	1.5*	$0.57-4 \cdot 10^{-3}$
SH^-	c)	9 ⁱ⁾	-	(9)	0.018 ^{s)}	1.5*	$0.034 \cdot 10^{-3}$
H_2	c)		0		3.6 ^{s)}	10	$1.0 \cdot 10^{-3}$
	d)		0		18 ^{s)}		$5.0 \cdot 10^{-3}$
CH_4	d)		0		39 ^{s)}	4	0.028
Sr	a)	23	0.6	$30 \cdot 10^3$	$10 \cdot 10^3$		
	b)	8-14 ⁱ⁾	0.6	$10-14 \cdot 10^3$	$3-5 \cdot 10^3$	1.3*	8-51
	f)	2.6	2.9	$31 \cdot 10^3$	$11 \cdot 10^3$		
	g)	1.8-12	2.9	$10-67 \cdot 10^3$	$3-23 \cdot 10^3$		
Cs	a)	7.5	0.6	$9 \cdot 10^3$	$3 \cdot 10^3$		
	b)	1.5-3	0.6	$2-4 \cdot 10^3$	$0.7-1.5 \cdot 10^3$	2.0	0.1-4.5
	f)	0.9 ⁱ⁾	1.4	$1.8 \cdot 10^3$	$0.6 \cdot 10^3$		
	g)	1.4-2	1.4	$3.9-5.7 \cdot 10^3$	$1.5-2 \cdot 10^3$		
Tc	f)	53 ⁱ⁾ oxidizing conditions					
Th	f)	$4.6-8.5 \cdot 10^{-3}$ i)	≥ 6	55-100	20-35	1 ^{\epsilon}	0.055-0.1
Pa	f)	> 0.06 i)	5	$> 6 \cdot 10^3$	$2.1 \cdot 10^3$	1 ^{\epsilon}	6
U	f)	$0.58-0.82$ i)					
		(600 mg/l HCO_3^- in aq)	0.093	110-150	39-53	1 ^{\epsilon}	0.11-0.15
		$0.19-0.32$ i)					
	(10 mg/l humic acid in aq)	-	-				
	$0.57-0.94$ i)						
	(1 % $Fe_3(PO_4)_2$ in clay)	-					
Np	f)	$0.22-0.37$ i)	0.12	52-88	18-31	1 ^{\epsilon}	0.05-0.09
Pu	f)	$6.9-30 \cdot 10^{-3}$ i)	3.5	48-210	17-74	1 ^{\epsilon}	0.05-0.21

Table 1. Diffusivities of some substances in compacted Na-bentonite at about 25° C.

Compound	Reference	$D_a \cdot 10^{12}$ measured values m^2/s	K_d m^3/kg	$D_p \cdot 10^{12}$ from D_a m^2/s	$D_p \epsilon_p \cdot 10^{12}$ m^2/s	$D_v \cdot 10^9$ m^2/s	δ_D/τ^2 evaluated with $\epsilon_p = 0.35$
Am	f)	$4.0-14 \cdot 10^{-3}$ i)	6.6	53-180	19-63	$1^{\&}$	0.05-0.18
Eosin M=548	c)		-		$0.6 \cdot 10^{-3}$ s)		
AQS ⁻ M=240	c)		-		$2 \cdot 10^{-3}$ s)		
LS ⁻	c) M=5-30 $\cdot 10^3$	-	0		$> 10^{-4}$ - 10^{-3} s)		
	e) M=24 $\cdot 10^3$	-	0		< 0.003 s)		

$$D_a = \frac{D_p}{1 + K_d \rho}; \quad M = \text{Molecular weight}$$

$$\rho = 2000-2100 \text{ kg/m}^3, \quad \epsilon_p \approx 0.3-0.35$$

* Dilute solutions with Na as kation s) Obtained from stationary measurement

& General value for small ions i) obtained from instationary measurements

Table 1. Diffusivities of some substances in compacted Na-bentonite at about 25^o C.

Type of compound	$D_a \cdot 10^{12}$ m ² /s	$D_p \cdot 10^{12}$ m ² /s	$D_p \epsilon_p \cdot 10^{12}$ m ² /s	$D_v \cdot 10^9$ m ² /s
Dissolved gases e.g. CH ₃ .H ₂			4-40	
Small anions non interacting e.g. I ⁻ , Cl ⁻ , (SH ⁻), SO ₃ ²⁻ CO ₃ ²⁻ ..	4-9	4-9	0.2-0.3	~ 1.5
Large anions non interacting e.g. LS ⁻ , AQS ⁻ ..			(0.5-2) · 10 ⁻³	< 1
Colloids			(0.1-3) · 10 ⁻³	<< 1
Alkali metal ions e.g. Cs ⁺ , Na ⁺	1-8	(2-10) · 10 ³	(0.7-3) · 10 ³	~ 2
Alkaline earth metal ions e.g. Sr ²⁺ , Ca ²⁺ , Ra ²⁺	2-25	(10-70) · 10 ³	(3-25) · 10 ³	~ 1.5
The actinides Th, U, Np, Pu, Am		50-200	15-70	~ 1
Pa	> 0.6	> 6 · 10 ³	2 · 10 ³	~ 1
Tc oxidizing conditions	53	53	15	~ 1

Table 2. Estimated range of diffusivities for various types of compounds.

Compound	$D_a \cdot 10^{12}$ m^2/s
I ⁻	9
TcO ₄ ⁻	53
Cs ⁺	8
Sr ²⁺	25
Ra ²⁺	25
Pa	1
Th ¹⁾	10^{-2}
U ¹⁾	1
Np ¹⁾	0.4
Pu ¹⁾	$3 \cdot 10^{-2}$
Am ¹⁾	$1.5 \cdot 10^{-2}$

1) The dissolved species are probably carbonate, hydroxyl and mixed complexes, see Allard (Asilomar Sept 10-15, 1981).

Table 3. Upper limits of apparent diffusivity values of some nuclides for breakthrough calculations.

Nuclide	$D_a \cdot 10^{12}$ m^2/s	$t_{1/2}$ years	$t_{0.05}$ years	$t_{0.05}/t_{1/2}$	N/No max
I ¹²⁹	9	$2 \cdot 10^7$	53	i	1
Tc ⁹⁹	53	$2 \cdot 10^5$	9	i	1
Cs ¹³⁷	8	30	59.7	2.0	$10 \cdot 10^{-2}$
Sr ⁹⁰	25	28	19.1	0.68	$5.3 \cdot 10^{-2}$
Ra ²²⁶	25	$1.6 \cdot 10^3$	19.1	i	1
Th ²²⁹	10^{-2}	$7.3 \cdot 10^3$	$4.8 \cdot 10^4$	6.5	$2.9 \cdot 10^{-4}$
Pa ²³¹	1	$3.6 \cdot 10^4$	478	i	1
U ²³⁴	1	$2.5 \cdot 10^5$	478	i	1
U ²³⁵	1	$7.1 \cdot 10^8$	478	i	1
U ²³⁸	1	$4.4 \cdot 10^9$	478	i	1
Np ²³⁷	0.4	$2.0 \cdot 10^6$	$1.2 \cdot 10^3$	i	1
Pu ²³⁹	$3 \cdot 10^{-2}$	$2.4 \cdot 10^4$	$1.6 \cdot 10^4$	0.67	$5.3 \cdot 10^{-2}$
Pu ²⁴⁰	$3 \cdot 10^{-2}$	$6.6 \cdot 10^3$	$1.6 \cdot 10^4$	2.4	$6.6 \cdot 10^{-3}$
Am ²⁴¹	$1.5 \cdot 10^{-2}$	458	$3.2 \cdot 10^4$	69	0
Am ²⁴³	$1.5 \cdot 10^{-2}$	$7.4 \cdot 10^3$	$3.2 \cdot 10^4$	4.3	$1.3 \cdot 10^{-3}$

i) Insignificant retention time compared to halflife < 0.1.

Table 4. Time for radionuclide breakthrough in a 0.375 thick cylindrical backfill barrier.

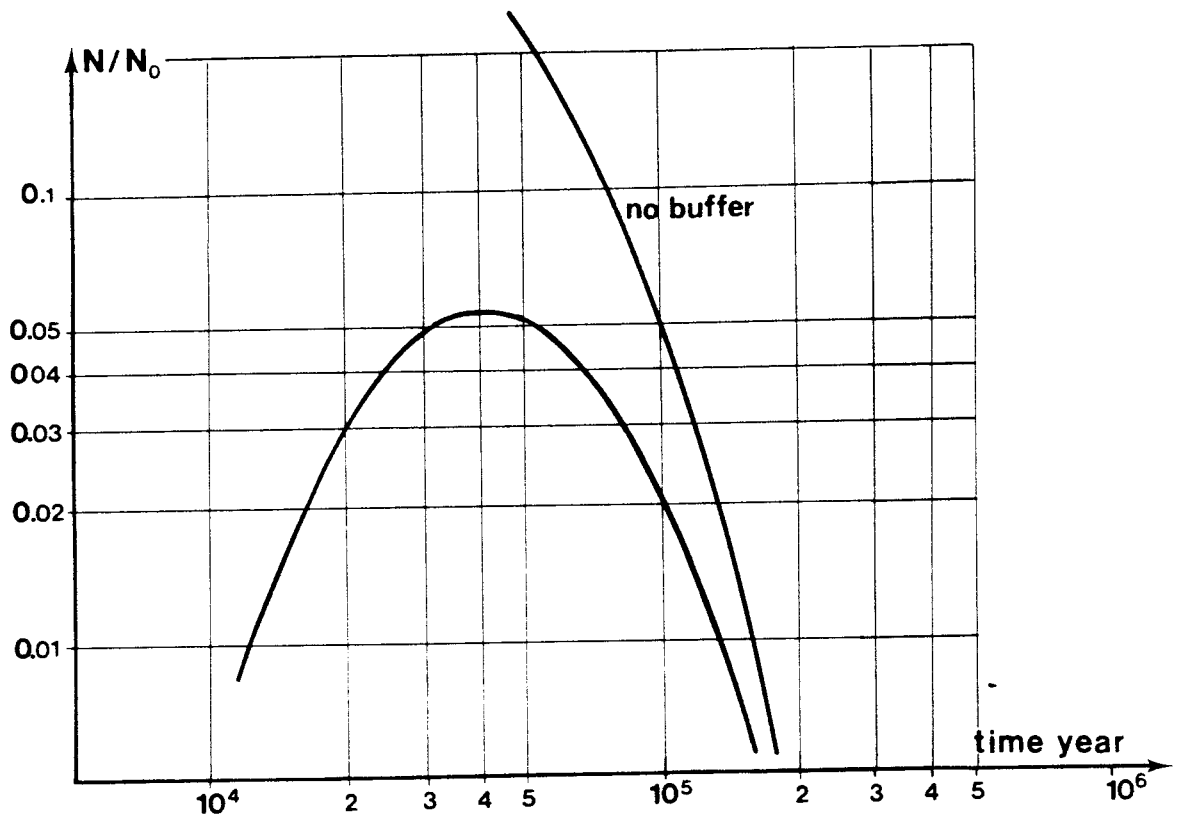


Figure 1. Breakthrough curve for Pu^{239}
 $D_a = 3 \cdot 10^{-14} \text{ m}^2/\text{s}$, $z_0 = 0.375 \text{ m}$

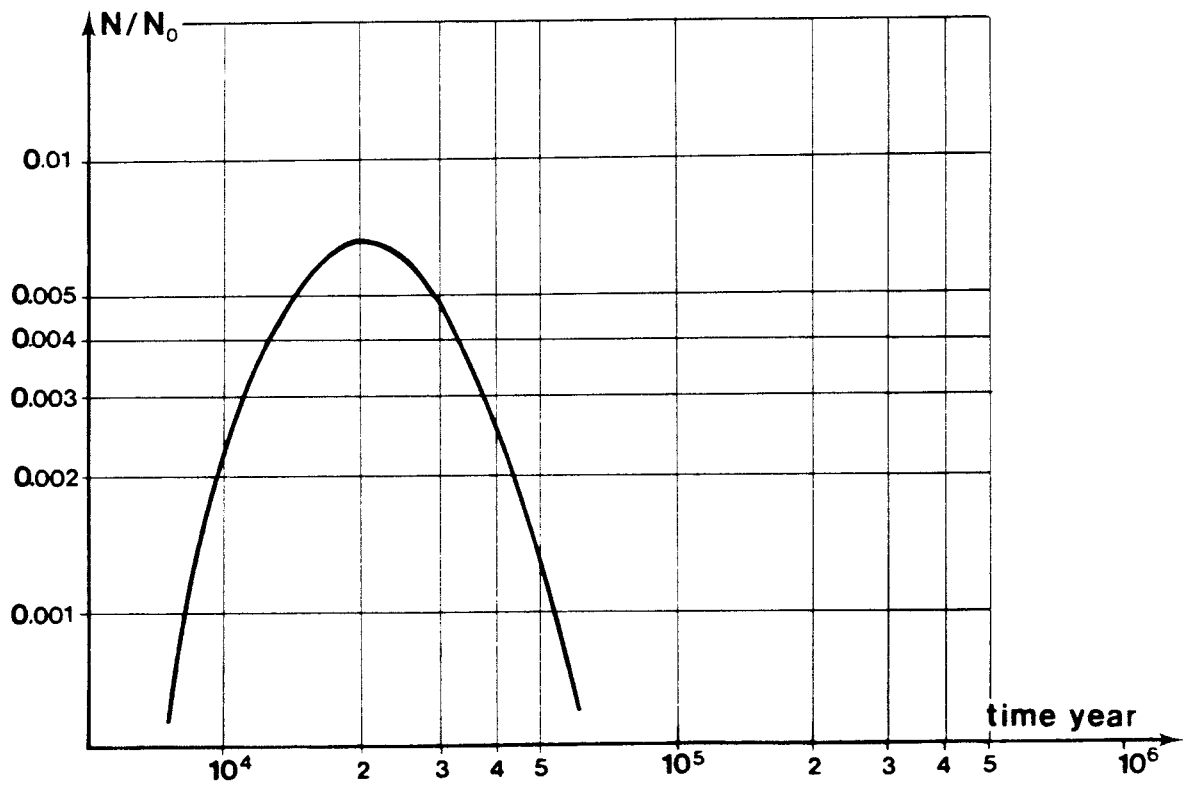


Figure 2. Breakthrough curve for Pu^{240}
 $D_a = 3 \cdot 10^{-14} \text{ m}^2/\text{s}$, $z_0 = 0.375 \text{ m}$

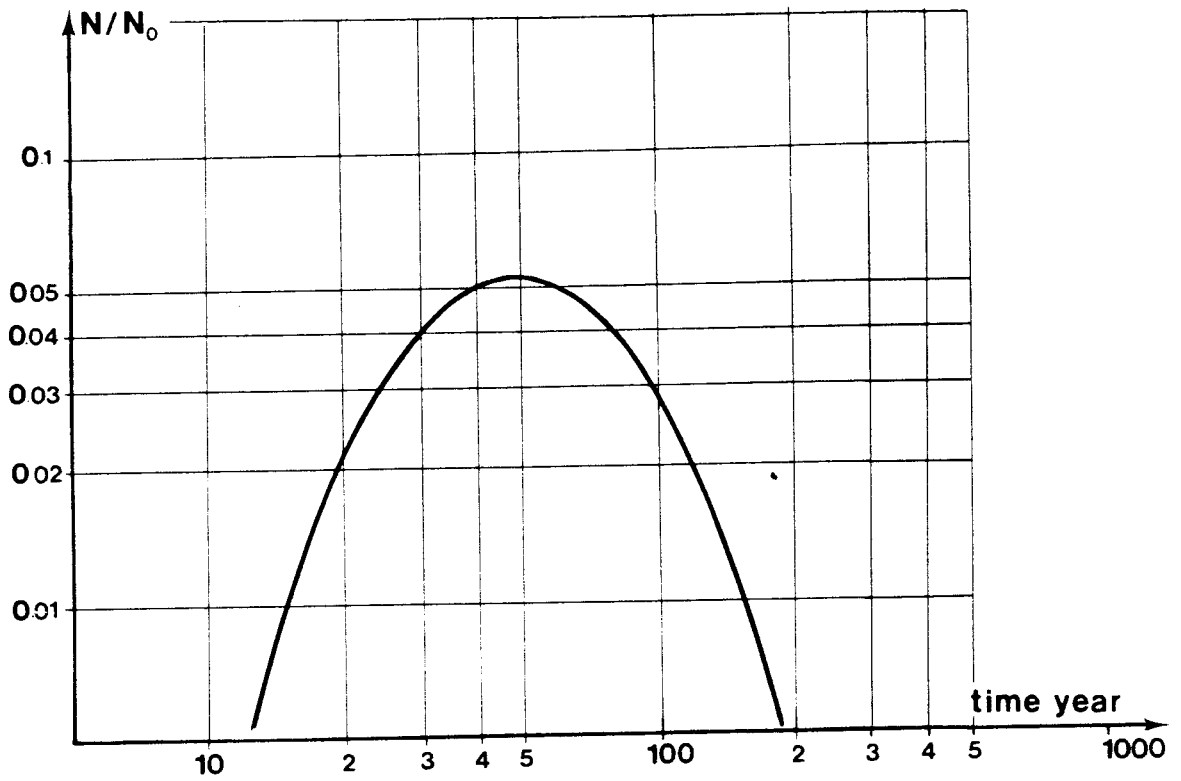


Figure 3. Breakthrough curve for Sr^{90}
 $D_a = 25 \cdot 10^{-12} \text{ m}^2/\text{s}$, $z_0 = 0.375 \text{ m}$

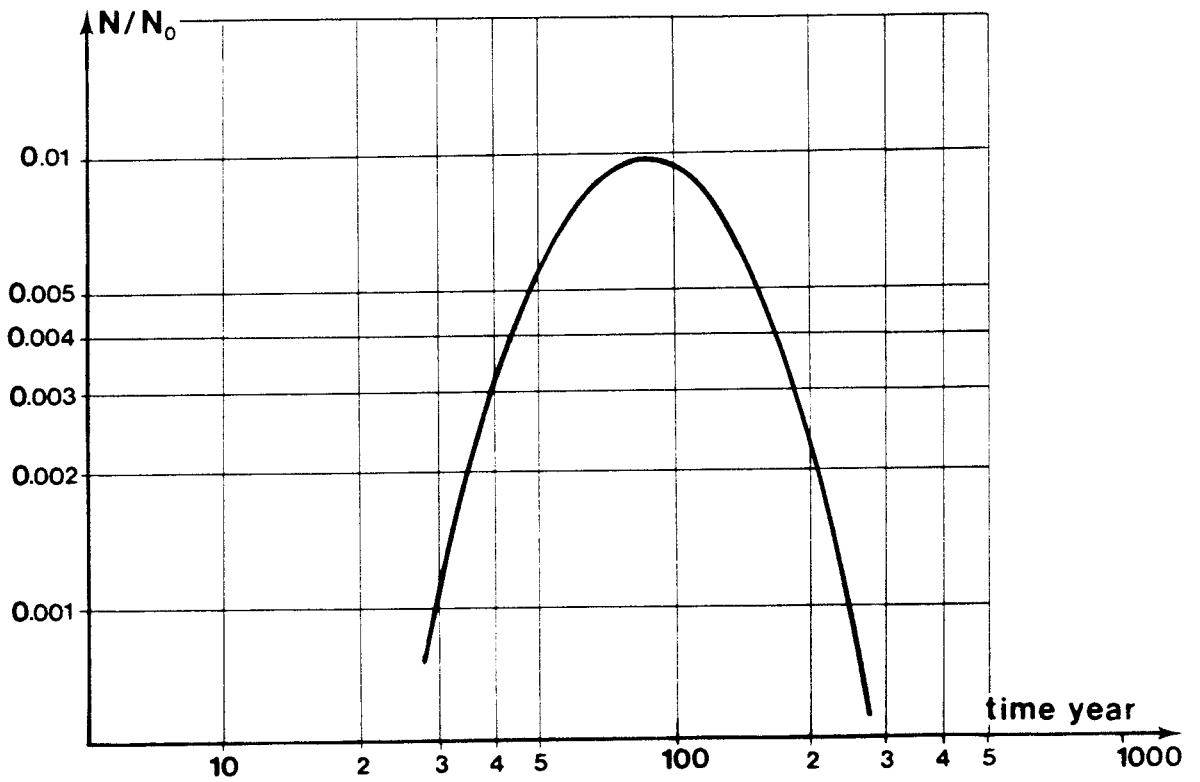


Figure 4. Breakthrough curve for Cs^{137}
 $D_a = 8 \cdot 10^{-12} \text{ m}^2/\text{s}$, $z_0 = 0.375 \text{ m}$



Figure 5. Breakthrough curve for Am^{243}
 $D_a = 1.4 \cdot 10^{-14} \text{ m}^2/\text{s}$, $z_o = 0.375 \text{ m}$

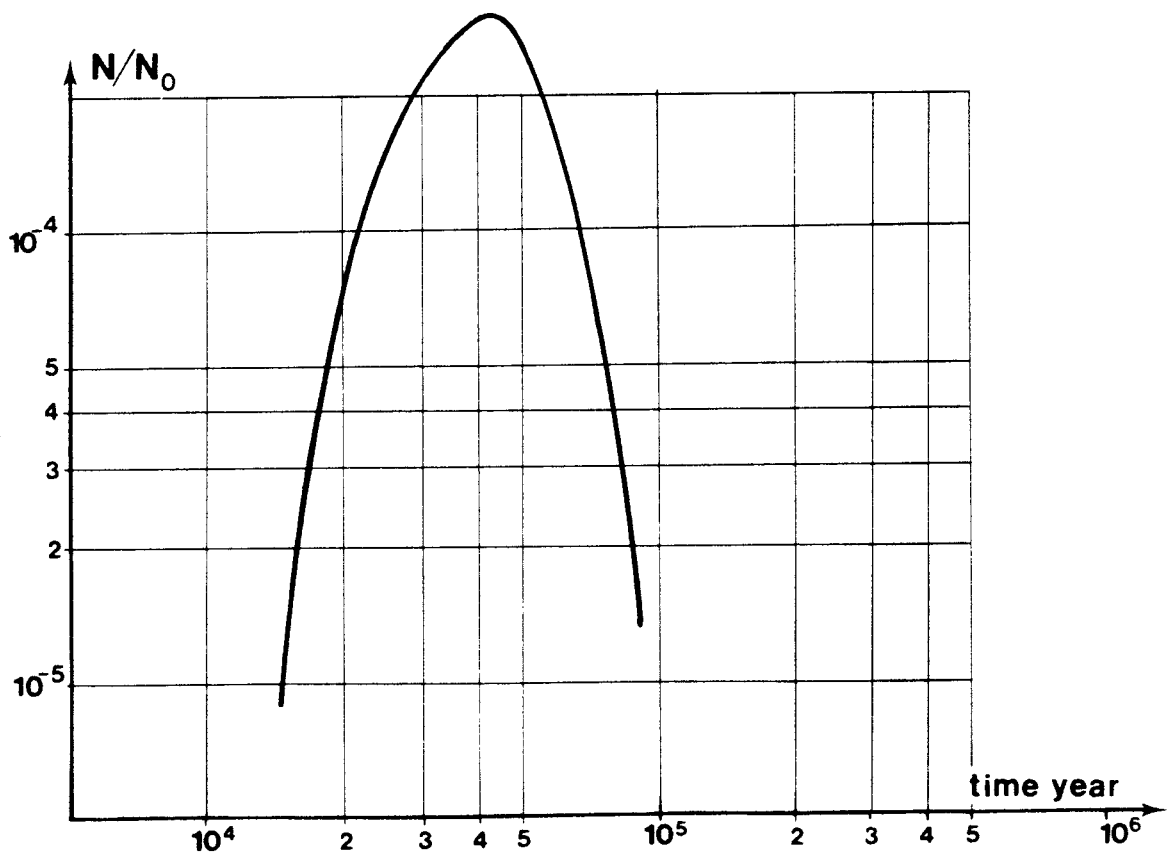


Figure 6. Breakthrough curve for Th^{229}
 $D_a = 9 \cdot 10^{-15} \text{ m}^2/\text{s}$, $z_o = 0.375 \text{ m}$

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