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Diffusion of radionuclides in concrete/bentonite systems

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ABSTRACT

In a repository for nuclear waste, different construction materials will be used. Two important materials among these are concrete and bentonite clay. These will act as mechanical barriers, preventing convective water flow, and also retard transport due to diffusion of dissolved radionuclides by a combination of mechanical constraints and chemical interactions with the solid.

An important issue is the possible change of the initial sodium bentonite into the calcium form due to ion exchange with calcium from the cement. The initial leaching of the concrete has been studied using radioactive spiked concrete in contact with compacted bentonite.

The diffusion of Cs, Am and Pu into 5 different types of concrete in contact with porewater have been measured. The measured diffusivity for Cs agrees reasonable well with data found in literature. For Am and Pu no movement could be measured (less than 0.2 mm) even though the contact times were extremely long (2.5 y and 5 y, respectively).

This report gives also a summary of the previously published results about sorption and diffusion of radionuclides in cement performed in Prav/KBS/SKB projects 1980 - 1990.

SAMMANFATTNING

I ett förvar för radioaktivt avfall kommer olika konstruktionsmaterial att användas. Betong och bentonitlera är två av de mest betydelsefulla materialen i detta sammanhang. De kommer att retardera radionuklidernas vandring ut från förvaret på två sätt, i) mekanisk barriär som hindrar vattenflöde, ii)lägre diffusion av radionuklider beroende på kemiska interaktioner med materialet.

En viktig fråga att utreda är möjligheten av ett utbyte av natrium i bentoniten mot kalcium genom jonbyte med kalcium som lakas från betongen. Den initiala lakningen av betongen har studerats genom att använda radioaktivt märkt betong i kontakt med bentonit.

Diffusionen av Cs, Am och Pu i fem olika sorters betong i kontakt med betongporvatten har mätts. Diffusiviteten för Cs i betong stämmer väl överens med tidigare mätta diffusiviteter. För Am och Pu kunde ingen inträngning i betongen mätas (mindre än 0.2 mm) trots att långa kontaktider användes (2.5 respektive 5 år).

En sammanfattning av resultat uppnådda inom Prav/KBS/SKB-projekt under åren 1980-1990 ifråga om sorption och diffusion av radionuklider i betong är inkluderad i denna rapport.

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SUMMARY

This report gives a summary of the previously published results about sorption and diffusion of radionuclides in cement performed in Prav/KBS/SKB projects 1980 - 1990.

In addition recent results not previously published are presented. These include studies of diffusion of Cs, Am and Pu in cement and diffusion of Na, Cs and Ca from concrete into bentonite in contact with each other.

Sodium showed as expected very low retention both in the concrete and in the bentonite clay in contact with the concrete. The measured diffusivity ($\sim 5 \cdot 10^{-11}$ m^2/s) seems only dependent on the porosity and constrictivity of the solid. For calcium, an apparent diffusivity of $3 \cdot 10^{-11}$ m^2/s is obtained in the bentonite. This is the same value that is expected in the concrete [ALL91], and little sorption is obtained in both cases.

Cesium shows an apparent diffusivity of $3 \cdot 10^{-12}$ m^2/s in the bentonite. The same value is obtained with another type of experimental set-up [TOR85, ALB89]. This indicates that the measured D_a-values are independent of how the experiment is performed.

The apparent diffusivity of Cs in normal types of concrete is about $1 \cdot 10^{-13}$ m^2/s. This have been shown both in this investigation, by Andersson et al [AND81, AND83] and by Johnston et al. [JOH92].

For plutonium and americium no movement could be measured even though the contact times was extremely long (5 y and 2.5 y respectively. The maximum estimated diffusivity with at these contact times and this experimental setup is in both cases around 10^{-17} m^2/s. This low diffusion can be explained by the low content of carbonate in the pore water, giving little complexation of Am and Pu with carbonate. The low diffusivity also indicates that no negatively charged hydroxi species are formed.

1. INTRODUCTION

In a repository for nuclear waste, different construction materials will be used in the engineered barriers. Two important materials among these are cement and bentonite clay. These will act as mechanical barriers, preventing convective water flow, and also retard transport due to diffusion of dissolved radionuclides by a combination of mechanical constraints and chemical interactions.

In connection with studies of the long time behavior of the barriers the mechanical and chemical changes with time in the solids are of large interest, since the possible rate of diffusion of nuclides through the materials are strongly dependent on these properties.

One important issue is the degradation of the cement, a process that is initiated by an exchange of the initial pore solution against groundwater due to diffusion, followed by a dissolution of cement material.

Another important issue is the possible change of the initial sodium bentonite into the calcium form due to ion exchange with calcium from the cement. This change is of large importance, since the calcium bentonite does not have the swelling ability of the sodium form. In a contact surface between cement and bentonite, there will initially be a large amount of sodium available, in the cement pore water, which initially essentially is a sodium/potassium hydroxide solution. In a stage when the initial pore solution of the cement has disappeared, there will be an increasing concentration of calcium in the pore water, due to equilibrium with calcium hydroxide. In this stage an exchange into calcium bentonite may be possible.

The sorption and diffusion of various radionuclides in cement paste, concrete and bentonite clay has been studied at the Department of Nuclear Chemistry, Chalmers University of Technology (CTH) since 1980. The studies were initially funded by the former Prav (Swedish Research Council for Radioactive Waste) and thereafter by the SKB (formerly KBS). The results are published in Prav and KBS/SKB technical reports as well as in conference proceedings. Also some previously unpublished results are available.

The results of the previous studies of diffusion in bentonite have earlier been compiled in a KBS technical report [TOR83]. These will not be further discussed here.

This report gives a summary of the previously published results for concrete and cement paste. In addition recent results not previously published are presented. These include studies of diffusion of Cs, Am and Pu in concrete and concrete-bentonite in contact with each other.

2. SORPTION AND DIFFUSION OF RADIONUCLIDES IN CONCRETE AND CEMENT PASTE. REVIEW OF PUBLISHED RESULTS IN Prav/KBS/SKB PROJECTS. 1980 - 1990.

The early studies of sorption and diffusion in concrete and cement paste have been published in a number of conference proceedings and technical reports:

- sorption on concrete and cement paste: [ALL81], [ALL81:2], [ALL84], [ALL87], [AND81], [AND83], [HÖG85]

- diffusion in concrete: [AND81], [AND83].

2.1 PERFORMED EXPERIMENTAL STUDIES

Several different cement solids have been studied in the sorption experiments. Apart from the Standard Portland cement, concrete and cement paste of sulfate resistant cement, blast furnace slag cement, fly ash cement, aluminate cement, and silica cement have been used. Also three different samples of very old (ca 70 years) Portland concrete taken from hydropower dams have been included. Also the effect of organic additives to the concrete was studied for the actinides. The organics used were: tributylphosphate (TBP) and ethanolamine for all the actinides and also ethylene diamine tetraacetic acid (EDTA) and diethylene triamine pentaacetic acid (DTPA) for Am. In the diffusion experiments only standard Portland concrete was used.

Several different water phases were used, although the main part of the diffusion experiments were performed in synthetic concrete pore water and the sorption experiments in synthetic groundwater.

2.1.1 Technique for sorption studies

The experimental sorption studies were all performed as batch experiments, where a crushed solid was contacted with a aqueous phase. All handling of the samples was performed in a glovebox with nitrogen atmosphere in order to avoid carbonation of the concrete. Before contacting with the liquid phase, the crushed solid was sieved and a well defined size fraction was used.

The aqueous phase has been an artificial pore water in most cases, although synthetic groundwater as well as sea water and Baltic Sea water were used in some early measurements. The water compositions are given in Table 2.1.

TABLE 2.1 Composition of synthetic waters [ALL81].

Species	mg/l GW	PW1	PW2	PW3	PW4	PW5	PW6	SW	BW
Na^+	65	1600	460	4100	7000	10000	7000	10560	4980
K^+	3.9	6300	630	120	12000	22000	12000	380	179
Mg^{2+}	4.3	-	-	-	-	-	-	1270	599
Ca^{2+}	18	40	320	-	10	-	-	398	188
Sr^{2+}	-	-	-	-	-	-	-	13.3	6,3
Al^{+3}	-	-	-	2700	27	75	75	-	-
F^-	-	-	-	-	-	-	-	1.4	0.7
Cl^-	70	-	-	-	-	-	-	18980	8950
Br^-	-	-	-	-	-	-	-	64.5	30.4
HCO_3^-	123	-	-	-	-	-	-	140	140
SO_4^{-2}	9.6	-	-	-	400	400	400	2650	1250
BO_3^{3-}	-	-	-	-	-	-	-	24.7	11.6
SiOtot	12	-	-	-	15	20	15		
pH	8-8,2	<13.4	<12.7	<13.3	13.2	13.9	13.2	8	8

PW1, PW2, PW3, PW4 = Pore waters 1-4 in [ALL84].
PW5,PW6 = Pore waters 1 and 2 in [ALL83].
BW and SW= Baltic Sea water and sea water according to [ALL83].

The radionuclide spiked water phase and the solid was contacted for different times, typically 1 day, 1 week and 1-3 months, before sampling. The phase separation before sampling the water phase was performed using different centrifuges. In the first experiments a centrifuge giving 4 000 g was used. Later a 42 000 g centrifuge was used, thus reducing the risk of inclusion of radionuclide containing colloids in the supernatant liquid.

The sorption was evaluated in terms of a distribution coefficient, K_d, defined as:

$$K_d = \frac{q}{c}\left[\frac{m^3}{kg}\right] \qquad (1)$$

where q = concentration in solid phase, [mol/kg] and c = concentration in aqueous phase, [mol/m^3].

The experimental conditions in each sorption measurement are summarized in Table 2.2.

TABLE 2.2 Experimental conditions for determination of distribution coefficients. Prav/KBS/SKB studies 1981 - 1991

Ele-ment	Solid	Liquid	part. sizes, mm	Ratio, g/l	Contact time	Phase sep.,g	Reference
Am	AL concr	synth. pw3	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	FA concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	FP concr	$Ca(OH)_2$	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	FP concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	M concr	synth.pw1,4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	M paste	synth. pw4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	PI concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	PII concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	SI concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	SP concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	SP concr	synth. pw1	0.090-0.125	20	6h,1d,6w,4m,7m	42 000	ALL 87
Am	SR concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Am	T concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
^{14}C	SP concr.	synth. gw	0.063-0.090	20	3d, 1w, 5w	4 000	ALL 81, ALL 81:2
^{14}C	SP paste	synth. gw	0.063-0.090	20	3d, 1w, 5w	4 000	ALL 81, ALL 81:2
Cs	AL concr	synth. pw3	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Cs	FA concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Cs	FP concr	synth. pw4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Cs	M concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Cs	PI concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Cs	PII concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Cs	SI concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Cs	SP concr	synth. bw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
Cs	SP concr	synth. gw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
Cs	SP concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Cs	SP concr	synth.pw5,6	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
Cs	SP concr	synth. sw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
Cs	SP paste	synth. bw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
Cs	SP paste	synth. gw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
Cs	SP paste	synth. pw6	0.063-0.125	11,20	1d, 1w	4 000	
Cs	SP paste	synth. sw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
Cs	SR concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	
Cs	T concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
I	FA concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
I	FP concr	synth. pw4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
I	M concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
I	PI concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
I	SI concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
I	SP concr	synth. bw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
I	SP concr	synth. gw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
I	SP concr	synth.pw5,6	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
I	SP concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
I	SP concr	synth. sw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
I	SP paste	synth. bw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
I	SP paste	synth. gw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
I	SP paste	synth. pw6	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
I	SP paste	synth. sw	0.063-0.125	11,20	1d, 1w	4 000	AND 81, AND 83
I	SR concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
I	T concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	AL concr	synth. pw3	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	FA concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	FP concr	$Ca(OH)_2$	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85

TABLE 2.2, continued.....

Ele-ment	Solid	Liquid	part. sizes, mm	Ratio, g/l	Contact time	Phase sep., g	Reference
Np	FP concr	synth.pw1,4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	M concr	synth.pw1,4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	M paste	synth. pw4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	PI concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	PII concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	SI concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	SP concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	SR concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Np	T concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	AL concr	synth. pw3	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	FA concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	FP concr	$Ca(OH)_2$	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	FP concr	synth.pw1,4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	M concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	M paste	synth. pw4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	PI concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	PII concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	SI concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	SP concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	SR concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Pu	T concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Th	FP concr	$Ca(OH)_2$	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Th	FP concr	synth. pw4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Th	M concr	synth. pw4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
Th	M paste	synth. pw4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	AL concr	synth. pw3	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	FA concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	FP concr	$Ca(OH)_2$	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	FP concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	M concr	synth.pw1,4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	M paste	synth. pw4	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	PI concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	PII concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	SI concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	SP concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	SR concr	synth. pw2	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85
U	T concr	synth. pw1	0.090-0.125	20	1d,6d,1w,6w,3m	42 000	ALL 84, HÖG 85

2.1.2 Technique for diffusion studies

The diffusion studies were performed by letting a radionuclide diffuse into a concrete sample from a water solution of the nuclide. By covering all outer surfaces of the sample but one, one dimensional diffusion was obtained. After a certain contact time the sample was taken out of the solution and the concentration profile of the element was evaluated. This was performed by grinding off well defined amounts from the sample surface (typically 0,1 mm at a time) and measuring the radioactivity of the removed dust [AND83].

The diffusion experiments were performed in a glove box with nitrogen atmosphere in order to avoid carbonation of the samples.

The diffusivity was evaluated from the concentration profile by the equation:

$$\frac{c}{c_0} = erfc\left(\frac{x}{2\sqrt{D_a t}}\right) \qquad (2)$$

where c = concentration at distance x, c_0 = concentration at the phase boundary, x = distance [m], D_a = apparent diffusivity [m^2/s], and t = time [s]. The diffusion theory is further discussed in chapter 5, below.

The experimental conditions in each diffusion experiment is summarized in Table 2.3.

2.1.3 Studied Radionuclides

The sorption of the following radionuclides has been studied: ^{14}C, ^{125}I, ^{131}I, ^{134}Cs, ^{233}U, ^{234}Th, ^{235}Np, ^{239}Pu, and ^{241}Am.

2.2 SUMMARY OF RESULTS

The performed sorption measurements and the experimental conditions in these are summarized in Table 2.2. The distribution coefficients are listed in Table 2.4. The experimental conditions and results of the diffusion studies are listed in Table 2.3.

TABLE 2.3. Experimental conditions for determination of diffusion in Standard Portland paste and concrete and measured diffusivities.

Element	Solid	Liquid	Contact time	Diffusivity [m^2/s]	Reference
Cs	SP paste	synth. pw5	2 - 3 m	$(2 - 7)\cdot10^{-14}$	AND 81, AND 83
Cs	SP concr	synth. pw5	2 - 3 m	$(2 - 7)\cdot10^{-14}$	AND 81, AND 83

TABLE 2.4. Selected results of sorption measurements.

Element	Solid	Liquid	Contact time	log Kd [m^3/kg]),	Reference
Am	AL concr	synth. pw	3m	0.8 ± 0.2	ALL 84, HÖG 85
Am	FA concr	synth. pw	3m	1.4 ± 0.4	ALL 84, HÖG 85
Am	FP concr	$Ca(OH)_2$	3m	1.4 ± 0.2	ALL 84, HÖG 85
Am	FP concr	synth. pw	3m	1.6 ± 0.1	ALL 84, HÖG 85
Am	FP concr	synth. pw	3m	1.6 ± 0.2	ALL 84, HÖG 85
Am	FP concr	synth. pw[b]	3m	1.6 ± 0.1	ALL 84, HÖG 85
Am	FP concr	synth. pw[c]	3m	1.2 ± 0.2	ALL 84, HÖG 85
Am	M concr	synth. pw	3m	1.3 ± 0.3	ALL 84, HÖG 85
Am	M paste	synth. pw	3m	1.4 ± 0.3	ALL 84, HÖG 85
Am	PI concr	synth. pw	3m	1.0 ± 0.3	ALL 84, HÖG 85
Am	PII concr	synth. pw	3m	1.3 ± 0.4	ALL 84, HÖG 85
Am	SI concr	synth. pw	3m	1.2 ± 0.4	ALL 84, HÖG 85
Am	SP concr	synth. pw	3m	0.9 ± 0.1	ALL 84, HÖG 85
Am	SP concr	synth. pw[a]	6w	1.0 ± 0.5	ALL 87
Am	SR concr	synth. pw	3m	1.0 ± 0.3	ALL 84, HÖG 85
Am	T concr	synth. pw	3m	0.4 ± 0.2	ALL 84, HÖG 85
C	SP concr.	synth. gw	5w	>4	ALL 81, ALL 81:2
C	SP paste	synth. gw	5w	>4	ALL 81, ALL 81:2
Cs	AL concr	synth. pw	3m	-2.9 ± 0.2	ALL 84, HÖG 85
Cs	FA concr	synth. pw	3m	-2.9 ± 0.2	ALL 84, HÖG 85
Cs	FP concr	synth. pw	3m	-3.0 ± 0.1	ALL 84, HÖG 85
Cs	M concr	synth. pw	3m	-2.9 ± 0.1	ALL 84, HÖG 85
Cs	PI concr	synth. pw	3m	-2.9 ± 0.2	ALL 84, HÖG 85
Cs	PII concr	synth. pw	3m	-2.4 ± 0.1	ALL 84, HÖG 85
Cs	SI concr	synth. pw	3m	-2.3 ± 0.2	ALL 84, HÖG 85
Cs	SP concr	synth. bw	1w	-1.7 ± 0.1	AND 81, AND 83
Cs	SP concr	synth. gw	1w	-0.6 ± 0.01	AND 81, AND 83
Cs	SP concr	synth. pw	1w	no sorption obs.	AND 81, AND 83
Cs	SP concr	synth. pw	3m	-3.0 ± 0.2	ALL 84, HÖG 85
Cs	SP concr	synth. sw	1w	-1.7 ± 0.1	AND 81, AND 83
Cs	SP paste	synth. bw	1w	-2.7 ± 0.3	AND 81, AND 83
Cs	SP paste	synth. gw	1w	no sorption obs.	AND 81, AND 83
Cs	SP paste	synth. pw	1w	no sorption obs.	AND 81, AND 83
Cs	SP paste	synth. sw	1w	no sorption obs.	AND 81, AND 83
Cs	SR concr	synth. pw	3m	-3.0 ± 0.3	ALL 84, HÖG 85
Cs	T concr	synth. pw	3m	-2.6 ± 0.1	ALL 84, HÖG 85
I	FA concr	synth. pw	3m	-1.4 ± 0.1	ALL 84, HÖG 85
I	FP concr	synth. pw	3m	-1.0 ± 0.1	ALL 84, HÖG 85
I	M concr	synth. pw	3m	-2.5 ± 0.2	ALL 84, HÖG 85
I	PI concr	synth. pw	3m	-1.6 ± 0.1	ALL 84, HÖG 85
I	SI concr	synth. pw	3m	-1.8 ± 0.1	ALL 84, HÖG 85
I	SP concr	synth. bw	1w	-2.2 ± 0.08	AND 81, AND 83
I	SP concr	synth. gw	1w	no sorption obs.	AND 81, AND 83
I	SP concr	synth. pw	1w	-2.7 ± 0.3	AND 81, AND 83
I	SP concr	synth. pw	3m	-0.9 ± 0.1	ALL 84, HÖG 85
I	SP concr	synth. sw	1w	-2.8 ± 0.3	AND 81, AND 83
I	SP paste	synth. bw	1w	-2.1 ± 0.08	AND 81, AND 83
I	SP paste	synth. gw	1w	-2.1 ± 0.06	AND 81, AND 83
I	SP paste	synth. pw	1w	-1.8 ± 0.1	AND 81, AND 83

TABLE 2.4, continued...

Element	Solid	Liquid	Contact time	log Kd [m^3/kg]),	Reference
I	SP paste	synth. sw	1w	-3.0 ± 0.4	AND 81, AND 83
I	SR concr	synth. pw	3m	-0.8 ± 0.1	ALL 84, HÖG 85
I	T concr	synth. pw	3m	-1.5 ± 0.1	ALL 84, HÖG 85
Np	AL concr	synth. pw	3m	0.3 ± 0.4	ALL 84, HÖG 85
Np	FA concr	synth. pw	3m	1.0 ± 0.2	ALL 84, HÖG 85
Np	FP concr	$Ca(OH)_2$	3m	1.0 ± 0.2	ALL 84, HÖG 85
Np	FP concr	synth. pw	3m	0.7 ± 0.2	ALL 84, HÖG 85
Np	FP concr	synth. pw	3m	0.7 ± 0.2	ALL 84, HÖG 85
Np	FP concr	synth. pw[b]	3m	0.7 ± 0.2	ALL 84, HÖG 85
Np	FP concr	synth. pw[c]	3m	0.7 ± 0.2	ALL 84, HÖG 85
Np	M concr	synth. pw	3m	0.8 ± 0.2	ALL 84, HÖG 85
Np	M paste	synth. pw	3m	0.7 ± 0.2	ALL 84, HÖG 85
Np	PI concr	synth. pw	3m	1.5 ± 0.5	ALL 84, HÖG 85
Np	PII concr	synth. pw	3m	0.6 ± 0.1	ALL 84, HÖG 85
Np	SI concr	synth. pw	3m	0.7 ± 0.5	ALL 84, HÖG 85
Np	SP concr	synth. pw	3m	0.6 ± 0.5	ALL 84, HÖG 85
Np	SR concr	synth. pw	3m	0.8 ± 0.5	ALL 84, HÖG 85
Np	T concr	synth. pw	3m	1.5 ± 0.5	ALL 84, HÖG 85
Pu	AL concr	synth. pw	3m	0.2 ± 0.2	ALL 84, HÖG 85
Pu	FA concr	synth. pw	3m	0.1 ± 0.2	ALL 84, HÖG 85
Pu	FP concr	$Ca(OH)_2$	3m	1.1 ± 0.2	ALL 84, HÖG 85
Pu	FP concr	synth. pw	3m	0.7 ± 0.1	ALL 84, HÖG 85
Pu	FP concr	synth. pw[b]	3m	0.8 ± 0.1	ALL 84, HÖG 85
Pu	FP concr	synth. pw[c]	3m	0.6 ± 0.2	ALL 84, HÖG 85
Pu	M concr	synth. pw	3m	0.6 ± 0.2	ALL 84, HÖG 85
Pu	M concr	synth. pw	3m	0.1 ± 0.2	ALL 84, HÖG 85
Pu	M paste	synth. pw	3m	0.7 ± 0.1	ALL 84, HÖG 85
Pu	PI concr	synth. pw	3m	0.3 ± 0.1	ALL 84, HÖG 85
Pu	PII concr	synth. pw	3m	0.3 ± 0.1	ALL 84, HÖG 85
Pu	SI concr	synth. pw	3m	1.0 ± 0.3	ALL 84, HÖG 85
Pu	SP concr	synth. pw	3m	0.1 ± 0.2	ALL 84, HÖG 85
Pu	SR concr	synth. pw	3m	0.7 ± 0.2	ALL 84, HÖG 85
Pu	T concr	synth. pw	3m	0.4 ± 0.3	ALL 84, HÖG 85
Th	FP concr	$Ca(OH)_2$	3m	0.8 ± 0.2	ALL 84, HÖG 85
Th	FP concr	synth. pw	3m	0.7 ± 0.1	ALL 84, HÖG 85
Th	FP concr	synth. pw[b]	3m	0.8 ± 0.1	ALL 84, HÖG 85
Th	FP concr	synth. pw[c]	3m	0.6 ± 0.2	ALL 84, HÖG 85
Th	M concr	synth. pw	3m	0.5 ± 0.2	ALL 84, HÖG 85
Th	M paste	synth. pw	3m	0.6 ± 0.2	ALL 84, HÖG 85
U	AL concr	synth. pw	3m	-0.4 ± 0.2	ALL 84, HÖG 85
U	FA concr	synth. pw	3m	0.2 ± 0.2	ALL 84, HÖG 85
U	FP concr	$Ca(OH)_2$	3m	0.7 ± 0.2	ALL 84, HÖG 85
U	FP concr	synth. pw	3m	0.2 ± 0.2	ALL 84, HÖG 85
U	FP concr	synth. pw	3m	-0.6 ± 0.1	ALL 84, HÖG 85
U	FP concr	synth. pw[b]	3m	-0.6 ± 0.1	ALL 84, HÖG 85
U	FP concr	synth. pw[c]	3m	-0.7 ± 0.1	ALL 84, HÖG 85
U	M concr	synth. pw	3m	0.2 ± 0.2	ALL 84, HÖG 85
U	M concr	synth. pw	3m	-1.0 ± 0.2	ALL 84, HÖG 85
U	M paste	synth. pw	3m	-0.6 ± 0.1	ALL 84, HÖG 85

TABLE 2.4, continued...

Element	Solid	Liquid	Contact time	log Kd [m^3/kg])	Reference
U	PI concr	synth. pw	3m	0.2 ± 0.2	ALL 84, HÖG 85
U	PII concr	synth. pw	3m	-0.2 ± 0.2	ALL 84, HÖG 85
U	SI concr	synth. pw	3m	0.8 ± 0.3	ALL 84, HÖG 85
U	SP concr	synth. pw	3m	0.3 ± 0.2	ALL 84, HÖG 85
U	SR concr	synth. pw	3m	0.4 ± 0.3	ALL 84, HÖG 85
U	T concr	synth. pw	3m	0.1 ± 0.2	ALL 84, HÖG 85

[a] with EDTA or DTPA
[b] with TBP
[c] with ethanolamine

2.2.1 Sorption in cement paste and concrete

^{14}C

The observed sorption of carbon-14 in concrete is very high, most probably due to an incorporation of the radionuclide in the matrix by isotopic exchange.

^{125}I, ^{131}I

The sorption of iodide is low but significant in all experiments reported but one. The measured distribution coefficients are in the range 0.001 - 0.16 m^3/kg. The highest sorption was observed for the three months samples (only two samples, one Standard Portland and one sulfate resistant concrete). There does not seem to be any significant difference between cement paste and concrete.

^{134}Cs

For Cs the sorption was very low. In several cases, especially for cement paste no measurable sorption was observed. The highest distribution coefficient observed was 0.25 m^3/kg for one sample of Standard Portland concrete. The major part of the values were in the range between 0.001 and 0.01 m^3/kg. There is no tendency towards higher distribution coefficients at three months contact time than at one week. On the contrary, the lowest values were observed after three months.

^{233}U

The sorption of uranium is quite high. Distribution coefficients between 0.10 and 6.3 m^3/kg have been measured. The highest values were observed for silica concrete, while the lowest were observed for blast furnace slag concrete and fly ash concrete. Addition of strong complex formers did not affect the sorption.

^{234}Th

Also the sorption of thorium in concrete was quite high. The distribution coefficients are within the range 3.1 to 6.3 m^3/kg. The sorption of thorium was not affected by addition of strong complex formers.

^{235}Np

The sorption of neptunium was very high. Distribution coefficients between 2.0 and 30.0 m^3/kg were observed. No effect of strong complex formers was observed.

^{239}Pu

Also the sorption of plutonium is high. The maximum observed distribution coefficient is ca 12.5 m^3/kg, while the lowest value is 1.2 m^3/kg. No effect of strong complex formers was observed.

^{241}Am

The sorption of americium was also very high. The range in distribution coefficients is from 2.5 to 40 m^3/kg. No effect of strong complex formers was observed.

2.2.2 Diffusion in Cement Paste and Concrete

Quite few experimental determinations of the diffusivity of radionuclides in cement paste and concrete have been undertaken within the program. Only early measurements of the diffusion of ^{134}Cs in Portland concrete have been published [AND81], [AND83].

The apparent diffusion was high (around 10^{-13} m^2/s) for Cs in both the performed measurements

3. SOLIDS

3.1 CONCRETE

3.1.1 Solid Cement Phases

In the following discussion on cement chemistry, a number of generally accepted abbreviations for components in the cement solids used in cement chemistry will be used. These are:

C	CaO
S	SiO_2
A	Al_2O_3
F	Fe_2O_3
K	K_2O
M	MgO
N	Na_2O

The most common cement type in Sweden is Standard Portland cement. Other types that may be used are blast-furnace slag cement and possibly sulfate resistent cement. Some other types of cement with other properties were included in the experimental studies e.g. fly ash cement, silica cement and aluminate cement. All the cement types but aluminate cement are based on Portland cement with different added solids. The principal hydration products may be expected to be the same for the Portland cement based solids, although the proportions between the products may vary.

For a typical unhydrated Portland cement the following solid phases and corresponding concentration ranges are common (weight %) [AND86]:

C_3S	45 - 65 %
C_2S	10 - 30 %
C_3A	5 - 15 %
C_4AF	5 - 12 %

The hydration products of the main solid phases in Portland cement are [CZE69]:

C_3S: $2\ C_3S + 6\ H_2O = C_3S_2{\cdot}3H_2O + 3\ Ca(OH)_2$

C_2S: $2\ C_2S + 4\ H_2O = C_3S_2{\cdot}3H_2O + Ca(OH)_2$

C_3A: $C_3A + 6\ H_2O = C_3A{\cdot}6H_2O$

C_4AF: $4\ C_4AF + 2\ Ca(OH)_2 + 10\ H_2O = C_3A{\cdot}6H_2O + C_3F{\cdot}6H_2O$

When the cement is hydrated a gel with some crystalline components is formed. In the solid there is an initially water filled pore system,

constituting ca 10 - 15 % of the volume. The gel crystallizes slowly, and gel phases are found after very long time [LEA70].

Thus a considerable amount of calcium hydroxide is present in the hydrated cement. Also calcium silicate, calcium aluminate and calcium ferrite phases are formed.

The relative amount of free calcium hydroxide in slag cement depends on the amount of slag added to the Portland cement. (Decreases with increasing slag content) [LEA70].

Sulfate resistant cement is a name given cements with low contents of solid phases likely to react with sulfate, mainly C_3A [LEA70]. Although the chemical processes involved are not completely understood, it is evident that cements with less than 5 % C_3A show a higher sulfate resistance.

When cement is mixed with water various additives are used in order to improve the properties. An addition of a workability aid, eg sulfonated melamine-formaldehyde polymer, may be expected in all the cements used in a repository. The total amount of this is usually much less than 3 % of the cement weight [AND86].

Ballast are also incorporated in the concrete. These may be quartz sand or a similar material.

3.1.2 Pore Water

The pore water in a cement is initially an alkali hydroxide solution. In a repository this will eventually be washed away by contact with surrounding water, and equilibria with the solid cement phases will occur. As long as calcium hydroxide is present, it will buffer the equilibrium pH to ca 12.4. Later silicate phases will dissolve, buffering the pH at a value of about 10 [AND86].

A few experimental determinations of pore water composition have been made, mainly for Portland cement. One determination compares compositions for a number of different cements [AND83:2], [AND89]. There is also one determination of some pore water components of a degraded cement [GJØ82]. Since this cement comes from a hydropower dam, aged approximately 70 years, the initial composition of the cement is not known. Probably it is some type of Standard Portland cement. Porewater data for Portland cement, sulfate resistant cement, blast-furnace slag cement and the degraded cement are given in Table 3.1.

TABLE 3.1.Composition of cement pore waters. Ion concentrations in mg/l. Data for SPB, SRB, MB and FAB from [AND83]. Data for degraded cements from [GJØ82].

Cement[a]	SPB	SRB	MB	FAB	Degr. a1	Degr. b1
Age[b]	10 m	9 m	10 m	10 m	70 y	70 y
w/c[c]	0.5	0.5	0.5	0.5	-	-
Na	1500	600	3200	1600	1500	260
K	6300	600	6000	7500	1530	180
Ca	90	470	50	15	92	570
Mg	0.2	0.2	0.1	0.15	-	-
Al	<5	<5	5	15	-	-
Si	<6	<6	<6	<6	-	-
Fe	0.5	0.5	0.5	0.7	-	-
I, calc[d]	0.23	0.04	0.29	0.26		
Eh[e]	139	84	-377	106		
pH	13.4	13.1	13.5	13.4	12.9	12.6

[a] SPB = Standard Portland, SRB = sulfate resistant, MB = blast-furnace slag cement, FAB = fly ash cement, Degr. = two different samples of 70 years old, degraded cement.
[b] for "new" cements, months, for degraded, years
[c] w/c = water/cement ratio
[d] ionic strength, calculated according to $I = \frac{1}{2}\Sigma c_i z_i^2$
[e] potential measured with platinum electrode, mV

3.2 BENTONITE

Bentonite is the name for a mixture of several mineral phases, mainly Na-montmorillonite and quartz. The bentonite used in these studies, Wyoming bentonite MX-80, consists to a large extent (85%) of a size fraction that is smaller than 2 µm. The mineral composition of the bentonite is given in Table 3.2. An elemental analysis is given in Table 3.3.

TABLE 3.2 Mineralogical Composition of MX-Bentonite [MÜL83].

Mineral	Content (%)
Montmorillonite	75
Quartz	15.2
Feldspar	5-8
Carbonate	1.4
Mica	< 1
Kaolinite	< 1
Pyrite	0.3
Organic Carbon	0.4
Other Minerals	2

TABLE 3.3. Elemental composition of Na-bentonite. [RAN87]

Element	Content (ppm)
Ag	5.5 - 5.8
Al	109 000
Au	0.0029 - 0.0031
As	5.9 - 6.5
Ba	360 - 470
Br	0.32 - 0.49
Ca	11 000
Co	2.4 - 3.0
Cr	34.2 - 35.3
Cs	0.65 - 0.79
Fe	27 500 - 31 100
K	4 000
La	46 - 50
Lu	0.97 - 1.3
Mg	1 500

Element	Content (ppm)
Mo	2.2 - 3.4
Na	17 500 - 19 800
Ni	33.1 - 34.5
Rb	10.0 - 13.1
Sb	1.2 - 1.3
Sc	5.3 - 5.9
Si	275 000
Sm	7.8 - 8.3
Sn	85 - 99
Ta	2.7 - 3.1
Th	34.3 - 35.9
U	10 - 12
W	0.63 - 0.66
Zn	100 - 115

Montmorillonite is a layered silicate mineral that may be derived from a pyrophyllite structure by the insertion, between the triple layers of the pyrophyllite, of sheets of molecular water containing free cations, coupled with substitution of Al for Si in the framework sheets [HUR71]. The free cations in the water layer can be Na, K, Ca, and Mg. The ions in the interlayer positions are exchangeable and may influence the porewater composition. Weathering of the montmorillonite would give an addition of more strongly bound metal ions, e.g. silica, and aluminum, to the solution.

The bentonite is likely to buffer a water solution, at 25 °C, to a pH-value within the range of 8 to 10, and to an Eh value in the range from -200 to -300 mV [RAN87] [JOH]. Experimental values in these ranges have also been obtained by Rantanen, et al. [RAN87]. The low Eh values may be attributed to the content of pyrite and organic material, cf Table 3.2.

The equilibrium concentrations of carbonate, calcium and sodium, as well as the pH-value of water in contact with bentonite can be expected to be governed by several chemical reactions. It is of significance that bentonite contains carbonate minerals and some organic material. Dissolution of the carbonate minerals as well as microbial degradation of organic components may occur and cause an increase in dissolved carbonate. The calcium concentrations can also be controlled by the solubility of calcite ($CaCO_3$).

Another effect is the influence, both on the dissolved carbonate and on the sodium and calcium concentrations in the solution, of the ion exchange reaction coupled with calcite dissolution:

$$2Na(ex) + CaCO_3(s) + H_2O \text{ ----> } Ca(ex) + 2Na^+ + HCO_3^- + OH^-$$

Other cations, e.g. hydrogen, potassium and magnesium ions, may also participate in ion exchange reactions in the sodium bentonite.

4. EXPERIMENTAL

4.1 RADIONUCLIDES

The following radionuclides were used in the diffusion measurements: ^{22}Na, ^{45}Ca and ^{134}Cs for the concrete-bentonite systems and ^{134}Cs, ^{239}Pu and ^{241}Am for the concrete-water systems. The radionuclides used represent elements with valence states of +1 (Na(I), Cs(I)), +2 (Ca(II)), +3 (Am(III)) and +4 (Pu(IV)).

The main purpose of investigating the elements sodium and calcium in the concrete-bentonite system was to study the degradation of the concrete. This was done by measuring the amount of calcium and sodium leached from the concrete into the bentonite. The actinides and cesium are of interest due to long time hazard in radioactive waste.

The activity of the nuclides ^{22}Na and ^{134}Cs was measured using a NaI(Tl) well-type detector measuring the photo peaks at 1275 keV and 604.7 keV, respectively. ^{45}Ca, which is a β-emitter, was measured using liquid scintillation counting. Finally, the α-activity from plutonium and americium were measured on a ZnS(Ag) scintillation detector. The initial concentrations of plutonium and americium in the waterphase were around 10^{-8} M.

4.2 APPARATUS

The equipment used for the measurements of activity profiles in the clay is described by Torstenfelt [TOR85] and by Albinsson [ALB89].

The apparatus used for grinding the concrete is shown in Fig. 4.1. This equipment was placed inside a glovebox in order to avoid contamination from dust. This was especially important when americium or plutonium diffusion was studied.

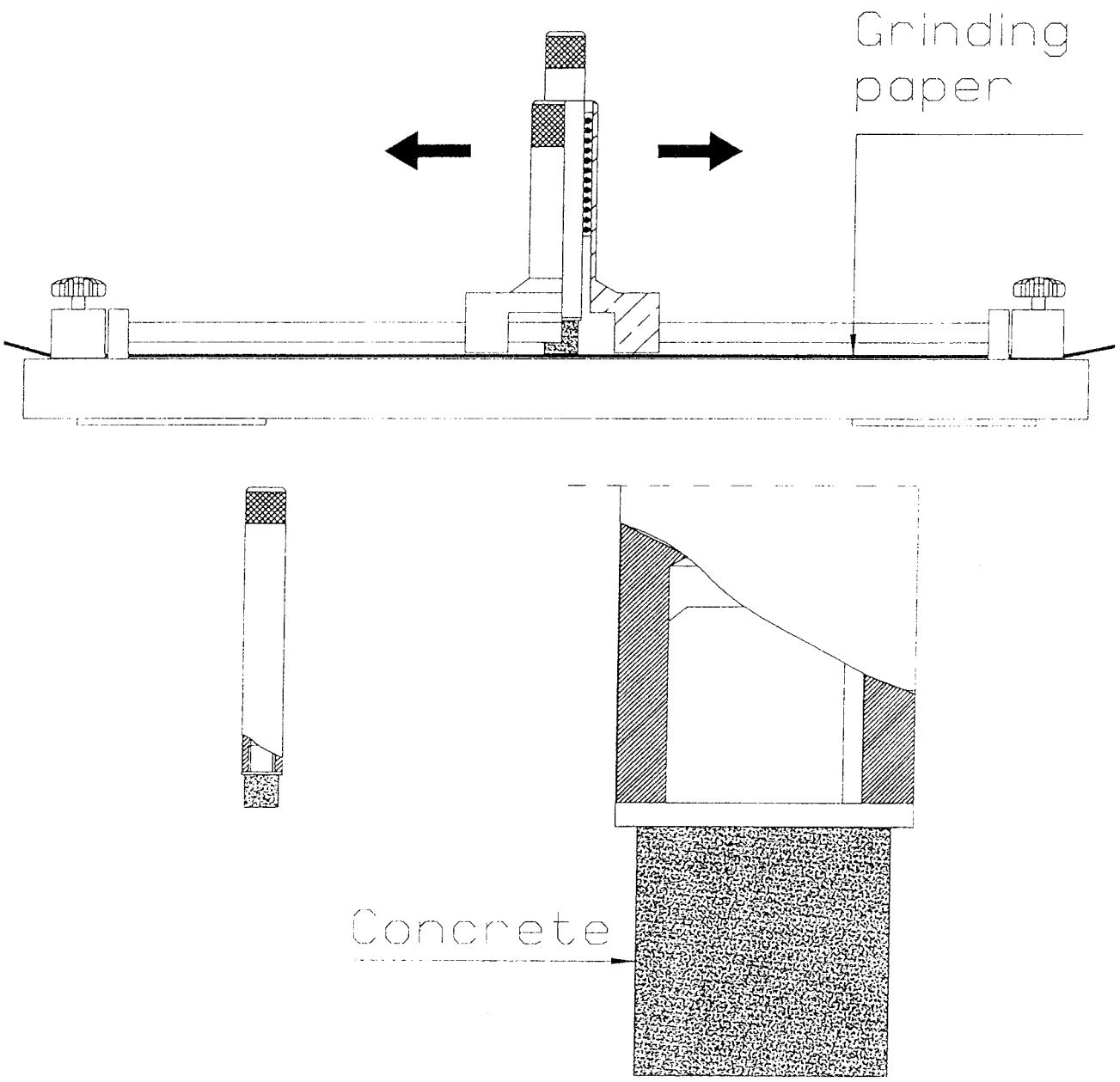

Figure 4.1. Grinding device for the study of concentration profiles in solids.

4.3 DIFFUSION MEASUREMENTS, CONCRETE/BENTONITE

The sodium bentonite used (MX-80, Wyoming bentonite) was preequilibrated for more than two weeks with an artificial groundwater [ALL79], after which it was centrifuged with an average centrifugal field of 22 000 g. The clay was then dried in an oven at 105 °C and finally ground in an agate mortar. The clay was then loaded into 25 mm long diffusion cells and compacted to 2000 kg/m^3. The entire cells were submerged in the artificial groundwater for more than one month, in order to wet/homogenise the clay completely. The water content/porosity was calculated to be 18% (equilibrated cell, dried at 105 °C, respectively).

Concrete samples (SPB) were prepared in plastic cylinders with an outer diameter of 25 mm, an inner diameter of 21 mm and a length of 25 mm. The concrete samples were spiked with ^{22}Na, ^{45}Ca or ^{134}Cs. The concrete paste to ballast ratio used was 0.62 and the water/solid ratio was 0.19. The concrete was only aged for some days, before it was contacted with the clay.

The concrete sample and the equilibrated clay was put together in a new diffusion cell with a length of 50 mm, and with no opening at the concrete end. This cell was then submerged in artificial groundwater for the appropriate time.

The clay was then sliced according to the procedure described by Albinsson et al. [ALB90] into ≈ 0.2 mm thick slices. The slices were air dried and the dry weight of each sample measured. The weight of the sample was then used to calculate a more precise thickness of each slice. In the case of sodium (^{22}Na) or cesium (^{134}Cs) the activity in the photopeaks at 1275 keV and 605 keV, respectively, was directly measured on a well-type NaI(Tl)-detector (Intertechnique CG 4000, France). The calcium (^{45}Ca, β-emitter) was extracted from the clay-slices with 1.5 ml 1 M HCl standing overnight. The tubes were centrifuged with a centrifugal field of about 300 g. One ml of the supertenant was mixed with 15 ml of liquid scintillation cocktail (Pico-Fluor 30, Packard instruments AB), and the samples were then measured on a liquid scintillation detector (Intertechnique SL-30, France).

In one case (^{22}Na, 6d contact time) the concrete sample was successively ground and the activity of each grinding paper was measured using a HPGe-detector (EG&G ORTEC., USA).

4.4 DIFFUSION MEASUREMENTS, CONCRETE

Six types of concrete pastes were used

* Standard portland paste (SPB): from Cementa AB, Slite, Sweden
* Sulphate resistant paste (SRB): from Cementa AB, Slite, Sweden
* Blastfurnace slag paste (MB): SPB + blastfurnace slag (mainly calcium silicate) Cementa AB, Slite, Sweden.
* Fly ash paste (FAB): SPB+30% fly ash
* Silica paste, Densit (SIB): SPB + 20% silica fume + 3.2% naphthalene
* High aluminia paste, Secar 80 (ALB): Titan AB, Upplands Väsby, Sweden

Conretes were prepared with cement paste, water and ballast and in a few cases also additives as described by Allard et al.[ALL84].

The concrete samples where moulded as cylinders with a diameter of about 20 mm. The samples where then aged in respective porewater [ALL84] for more than 12 months. After this they were sawn into samples with an approximate length of 25 mm. The concrete samples were covered with a thin resin on all sides except one and then fitted into metal holders. This holders were made to fit into a specially built grinding device (see Fig. 4.1). The samples were then put into a vessel with porewater spiked with a radionuclide. Only the uncovered side of the sample was dipped into the water. In order to have uniform concentration in the water a magnetic stirrer was introduced into the vessel. The vessels were closed to the surroundings to hold down evaporation losses of the water. All vessels was stored inside a glove-box with low air content, thus avoiding large uptake of carbonate.

After an appropriate contact time (Cs $\approx$ 0.7 y, Am $\approx$ 2.5 y and Pu $\approx$ 5 y) the concrete samples were taken out and ground according to the procedure described by Andersson et al. [AND81]. In each grinding a layer of between 0.1 to 0.7 mm was removed.

4.5 AUTORADIOGRAPHY

Autoradiograms were obtained using high speed film for direct X-ray exposures (REFLEX 15 single coated, CEA, Sweden). The autoradiograms were taken after each grinding in the beginning and after every second grinding in the end. The concrete samples were covered with a thin plastic film, and put on the photographic film. Exposure times were in the range 7 h to 72 days depending on sample activity. For americium autoradiograms were taken at five depths with the same exposure time, see Fig. 6.13, and this autoradiograms were later on used for estimation of the apparent diffusivity, c.f. paragraph 6.3. For plutonium only two autoradiograms were done, since almost all activity was removed after the first grinding, and no exposure could bee seen after 72 days exposure, see Fig. 6.15.

5. DIFFUSION THEORY

The diffusion rate of a trace element through a porous medium is mainly dependent on the molecular diffusion in the aqueous phase, on sorption phenomena, and on the pore constrictivity and the tortuosity of the solid. The general transport equation, which accounts for both advection and dispersion can be reduced to a diffusion equation which, if the relationship between the concentration of the solute on the solid

phase and the concentration of the solution is linear and reversible, can be written [CAR59]

$$\frac{dc}{dt} = \frac{d}{dx}(D_a \frac{dc}{dx}) \quad (3)$$

where D_a is the apparent diffusivity of a reactive solute in the medium. The solution of Eq.3, in one dimension, when Da is constant and independent of concentration, and with the prevailing initial and boundary conditions in the experiments, is [CRA75]

$$\frac{c}{c_0} = erfc(\frac{x}{2\sqrt{D_a t}}) \quad (4)$$

where c = concentration at distance x
c_0 = original concentration in the spiked cell
x = distance from the interface
t = time
D_a = apparent diffusivity

The diffusivity was calculated by fitting the experimental data to the equation with least square method.

6 RESULTS AND DISCUSSION

6.1 Na, Cs, Ca-DIFFUSION

The initial leaching of the concrete was studied using radioactive spiked concrete in contact with compacted bentonite.

6.1.1 Sodium

The sodium has shown a very low retention in the bentonite with an apparent diffusivity of $6 \cdot 10^{-11}$ m^2/s (cf. Fig. 6.1), which is more ore less exactly the same diffusivity as for TcO_4^- in pure bentonite with the same compactation [ALB89]. This indicates that there is no sorption of the sodium in the bentonite, and that the D_a measured is only dependent on the porosity and constrictivity of the solid. As can be seen from Fig. 6.2 is the D_a for sodium in the concrete of the same order as for the bentonite. This is in accordance with that the pore water content is of the same order in the bentonite (≈18%) as in the concrete (≈13%) [AND86, ALB89].

Another conclusion that can be drawn from the sodium experiments is that if too long contact time is used and the studied element is allowed to reach the bentonite/water interface, a too high diffusivity will be

measured in this experimental set-up. This is, however, not surprising since one of the conditions for eq. 2 is not fulfilled in this case.

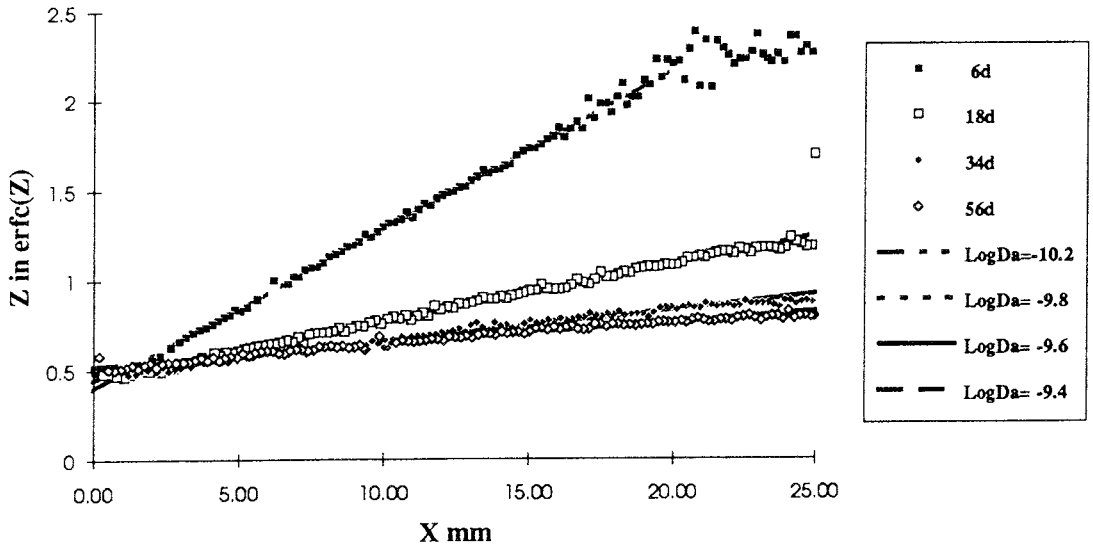

Figure 6.1. Diffusion of Na from a spiked SPB-concrete into compacted bentonite. Profile in the bentonite.

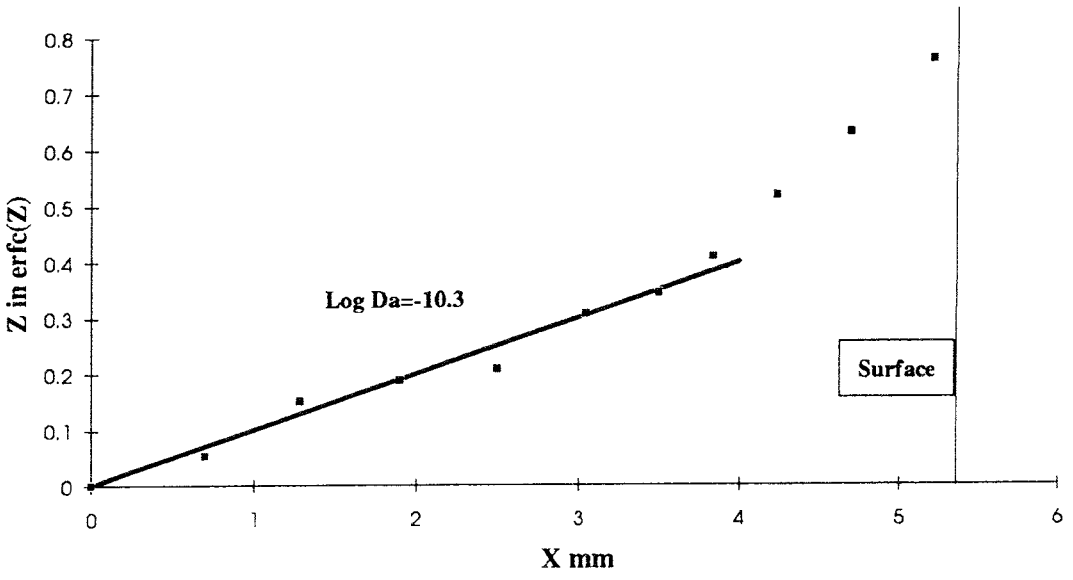

Figure 6.2. Diffusion of Na out of spiked concrete into bentonite. Concrete profile.

6.1.2 Calcium

For calcium only the activity profile in the bentonite was measured, see Fig. 6.3. The measured D_a-value in the bentonite is slightly lower than

for sodium, apparent diffusivity $3 \cdot 10^{-11}$ m^2/s, which is the same value that is expected in the concrete [ALL91].

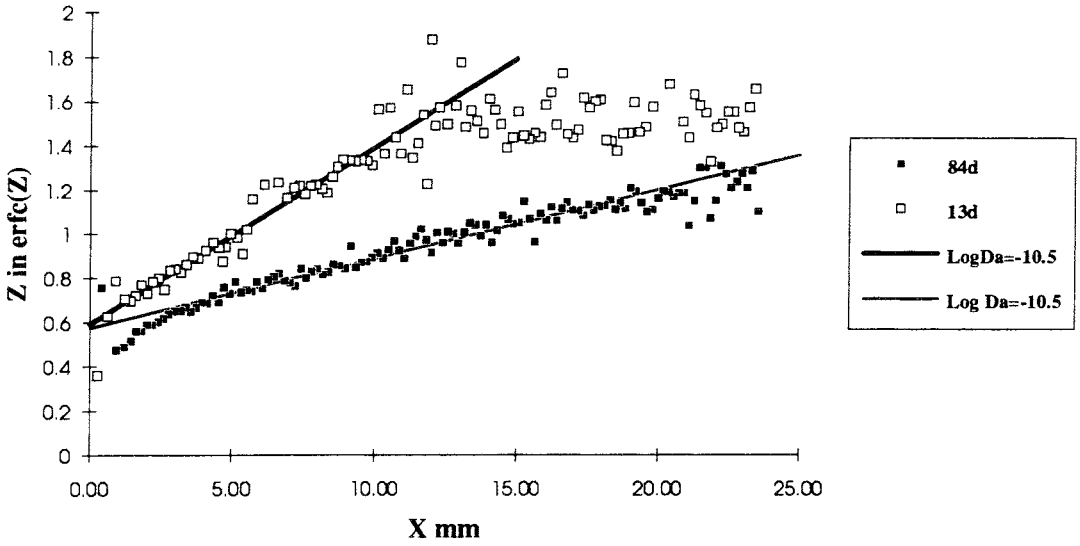

Figure 6.3. Diffusion of Ca from a spiked SPB-concrete into compacted bentonite. Profile in the bentonite.

6.1.3 Cesium

For Cs a D_a-value of about $3 \cdot 10^{-12}$ m^2/s is obtained in the bentonite. This is the same value that is measured with another experimental set-up [TOR85, ALB89] which is an indication that the obtained apparent diffusivities are independent of the experimental set-up.

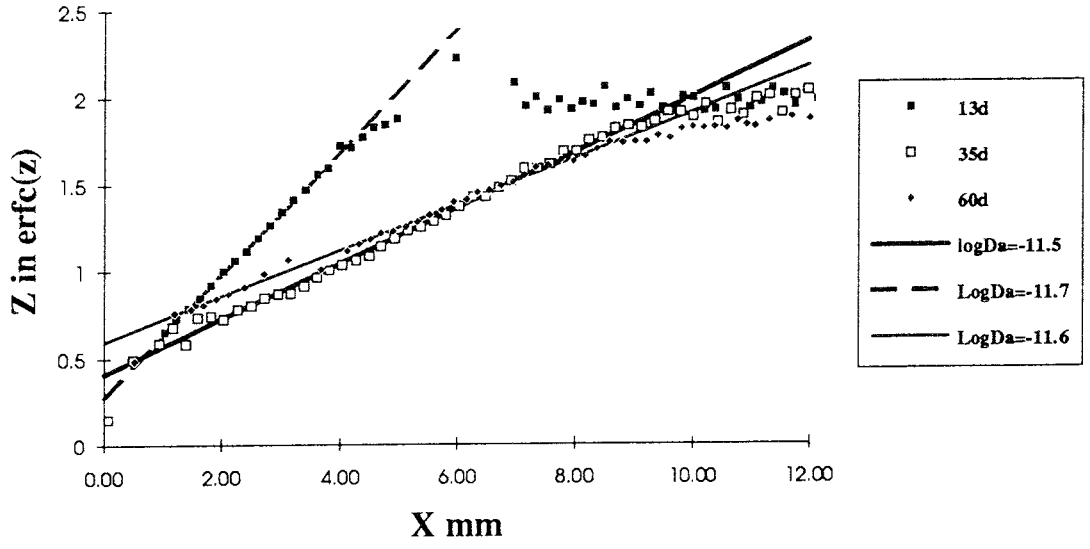

Figure 6.4. Diffusion of Cs from a spiked SPB-concrete into compacted bentonite. Profile in the bentonite.

6.2 Cs, Am, Pu DIFFUSION IN CONCRETE

6.2.1 Cesium

The diffusion of Cs in the different concrete's used are shown in Figures 6.5-6.10.

TABLE 6.1. Diffusion of Cs in different concrete's.

Concrete	Contact time, days	Log D_a, m^2/s
SPB	231	-13.1
SRB	231	-13.0
MB	260	-12.6
FAB	260	-12.8
SIB	260	-14.1
ALB	260	-11.9

As can be seen in Table 6.1, is the D_a for cesium in the concretes SPB, SRB, MB and FAB of the same order of magnitude and agrees reasonably well with data found in the literature [JOH92]. The lower diffusivity obtained in SIB can be explained by that this concrete has a

very dense structure and for ALB the high D_a-value is expected due to the low content of silica.

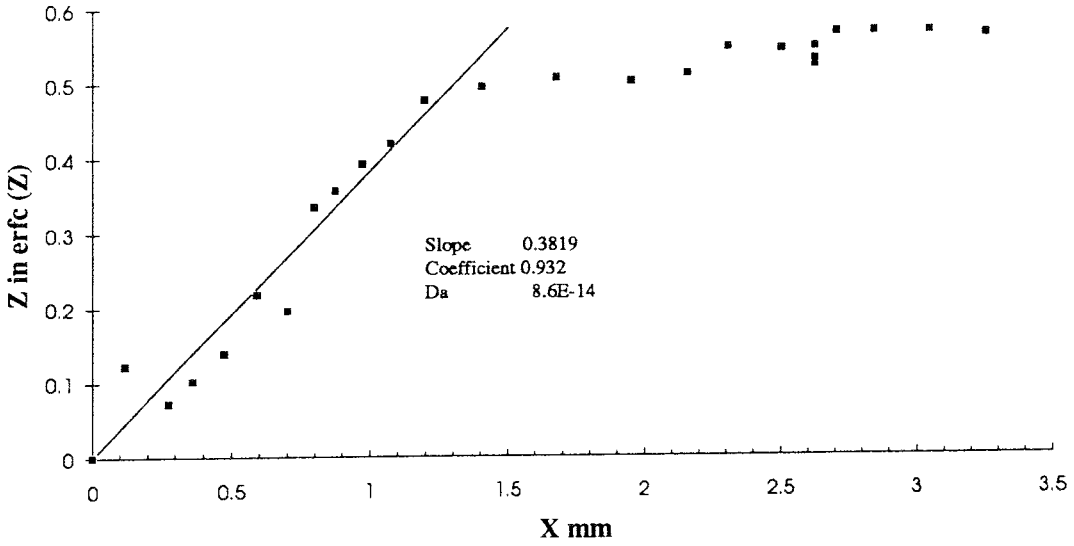

Figure 6.5. Diffusion of Cs in concrete (SPB). Diffusion time 231d.

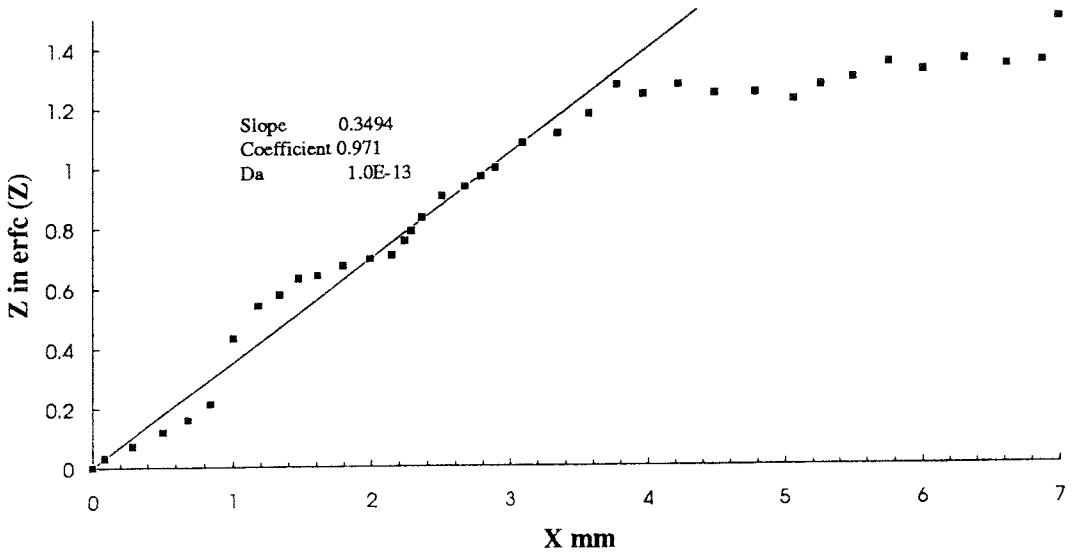

Figure 6.6. Diffusion of Cs in concrete (SRB). Diffusion time 231d

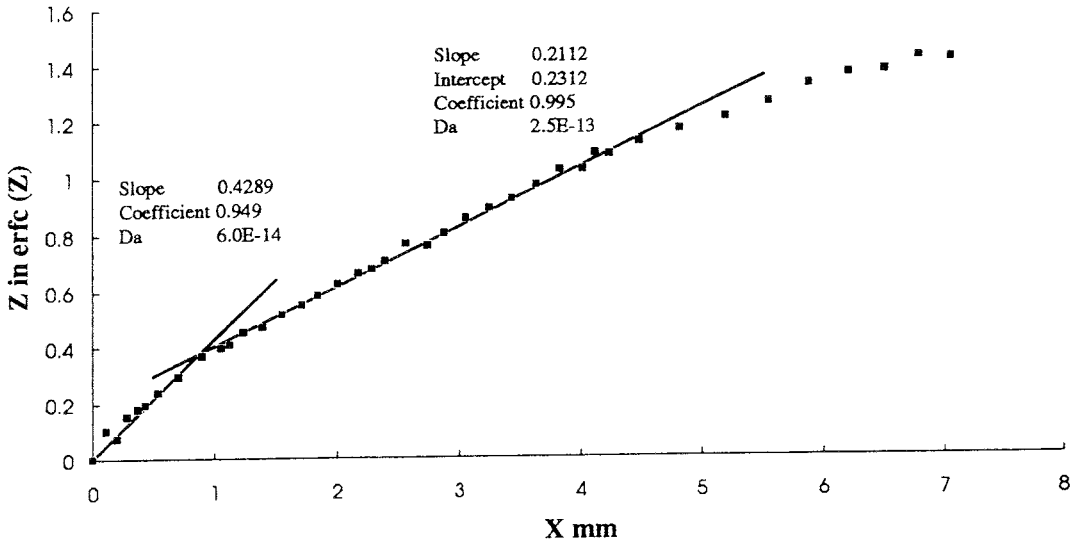

Figure 6.7. Diffusion of Cs in concrete (MB). Diffusion time 260d.

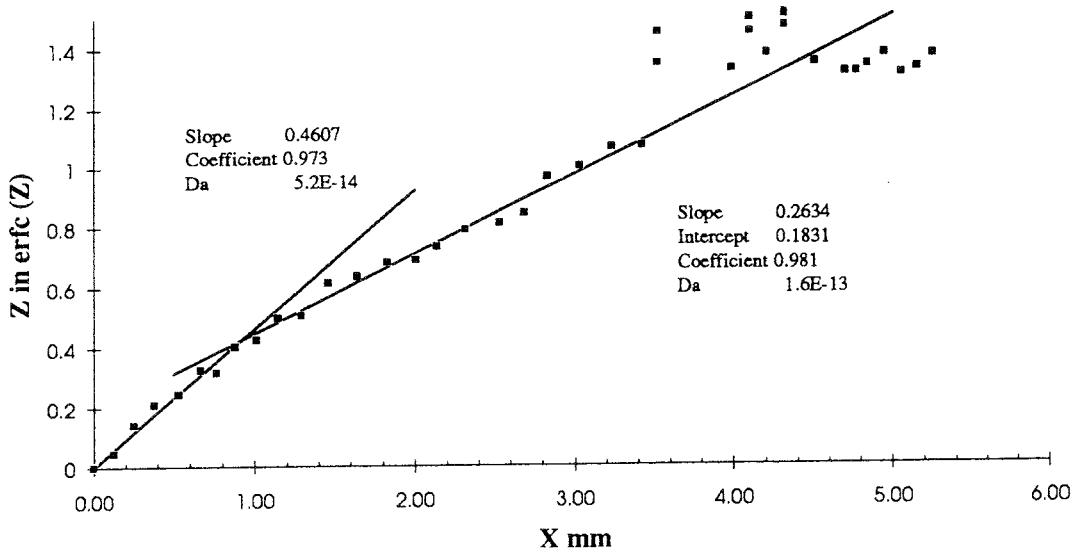

Figure 6.8. Diffusion of Cs in concrete (FAB). Diffusion time 260d.

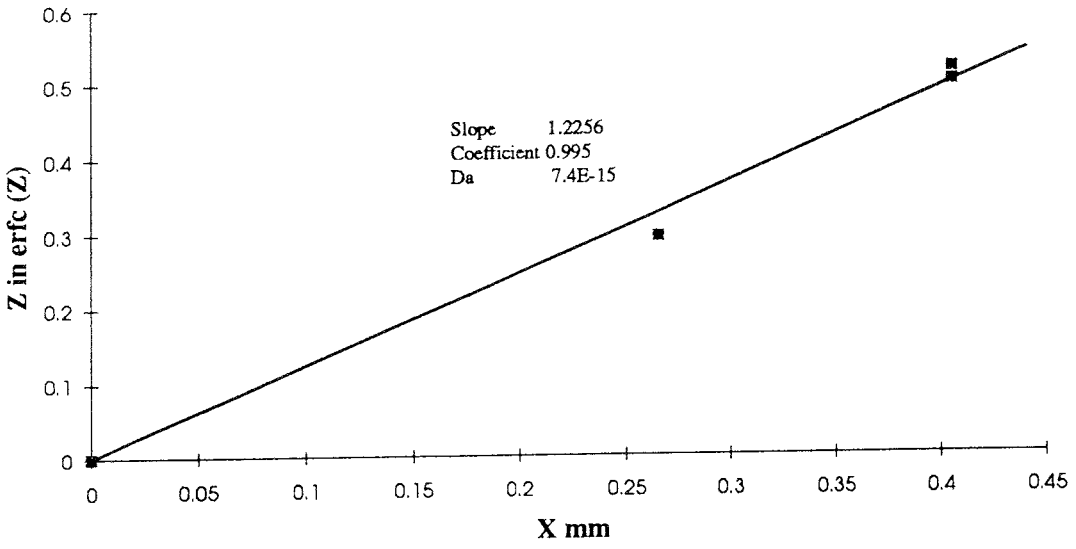

Figure 6.9. Diffusion of Cs in concrete (SIB). Diffusion time 260d.

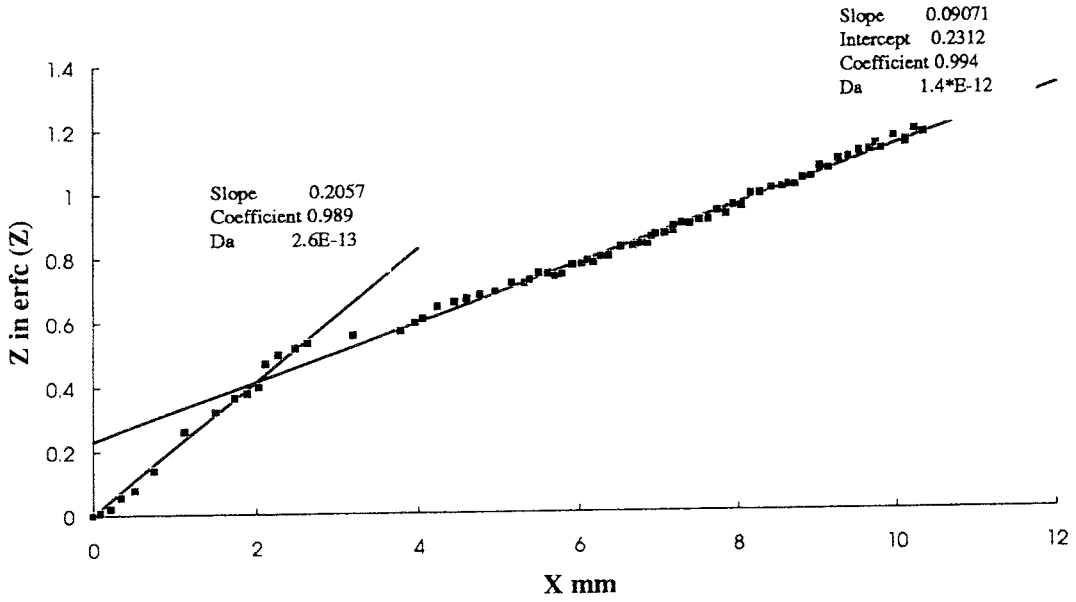

Figure 6.10. Diffusion of Cs in concrete (ALB). Diffusion time 260d.

For MB, FAB and ALB a lower apperent diffusivity can be observed in the first millimeter of the concrete. This can be due to cavities in the concrete and not completely planar samples, c.f. paragraph 6.2.2 resp. 6.2.3

6.2.2 Americium

The trivalent actinide Am is strongly sorbed on concrete, with K_d-values between 1 to 10 m^3/kg. [HÖG85] The anionic hydrolysis species

are supposed to dominate at this high pH and low carbonate concentration. The penetration of americium is for all the five concrete samples used (no SPB was available) extremely low even though a long contact time was utilised (912 d). Activity measurements gave to high diffusivity due to cavities in the concrete and not completely planar concrete samples, see Fig. 6.11 and Fig. 6.12.

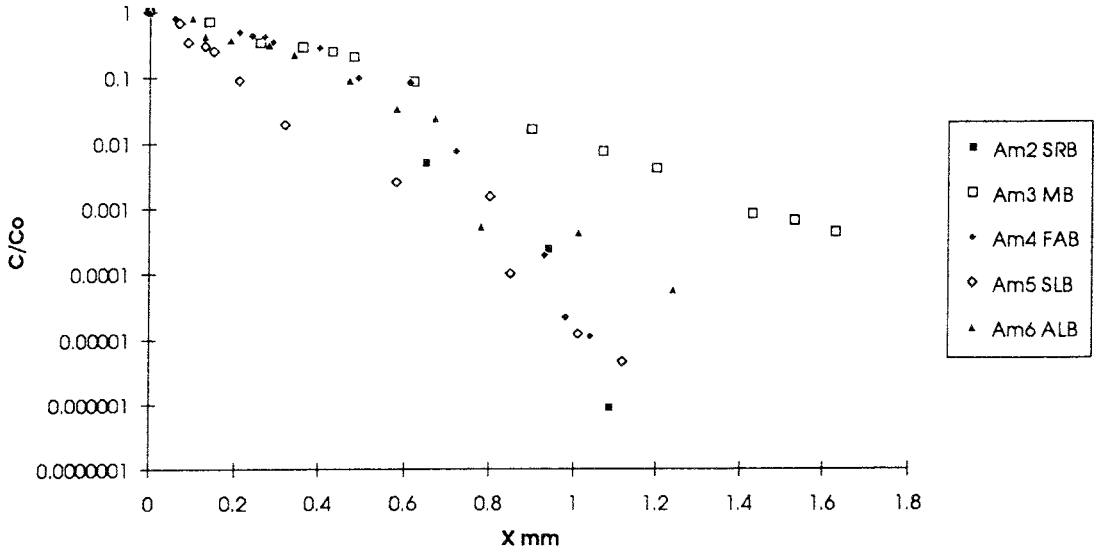

Figure 6.11. Americium penetration in concrete. Five concrete types.

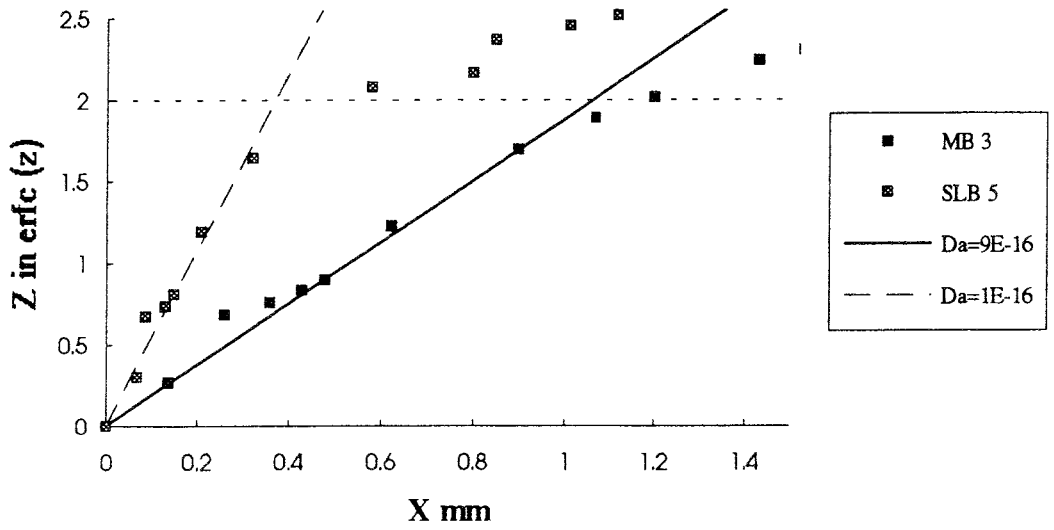

Figure 6.12. "Diffusion" of americium in concrete. The fastest (MB3) and slowest (SLB5) "diffusion" shown in figure. Dotted line shows estimated sensivity.

The estimated apparent diffusivity from activity measurement is $1\text{-}9 \cdot 10^{-16}$ m^2/s. However, the autoradiograms was used in order to evaluate a more accurate diffusivity. The estimation from autoradiograms was done comparing when a completely unexposed area was found after grinding and setting the activity to the same here

as when the whole concrete sample is planar and still some cavities gives exposure spots. In this case a countrate of below 500 cpm (background 1.2 cpm) gave a film with two small dots from cavities, and this was considered as the smallest activity that could be detected with autoradiogram using 7 h exposure time, see Fig. 6.13. This estimation gave a D_a<0.3-1.8·10^{-17} m^2/s, depending only on how deep the grinding was and not really on what type of concrete that was used.

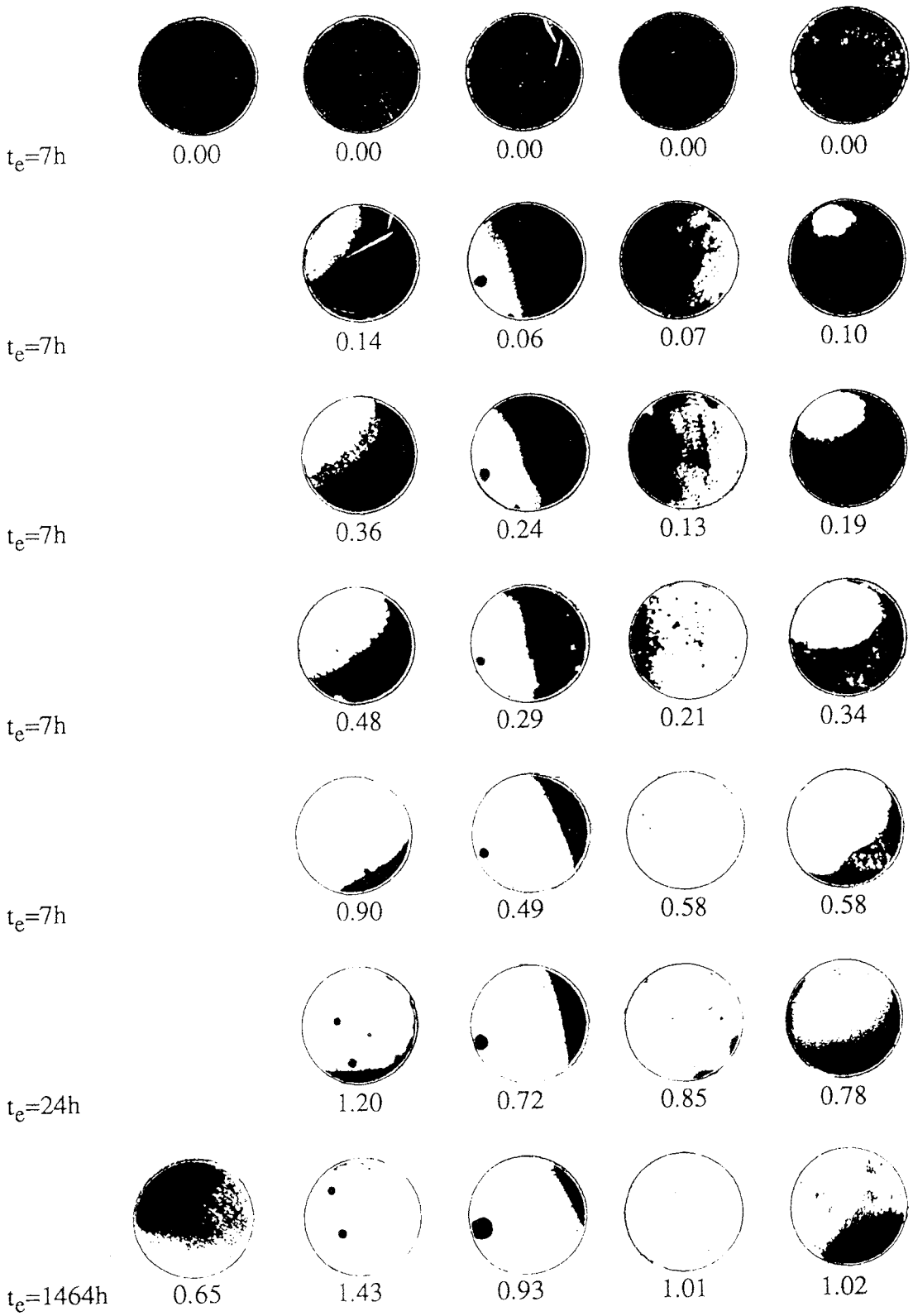

Figure 6.13. Autoradiographs showing penetration of americium in five different concrete samples, from left SRB, MB, FAB, SLB and ALB. Depths are given in mm, and t_e is the exposure time.

6.2.3 Plutonium

Several oxidation states are possible for plutonium. Oxic pore water conditions could favour the penta- and hexavalent oxidation states over the tetravalent, which is normally dominating under surface water conditions [ALL84]. The sorption is somewhat lower than for americium (K_d=1 to 5 m^3/kg) [HÖG85]. The estimated apparent diffusivity from activity measurement (Fig. 6.14) is <0.8-2.4·10^{-17} m^2/s, which as for Am is only dependent on cavities in the concrete. The relatively lower D_a values obtained for Pu compared with Am in the activity measurements is due to longer contact time (1850 d). No good estimation could be given from autoradiograms due to low starting activity (Fig. 6.15).

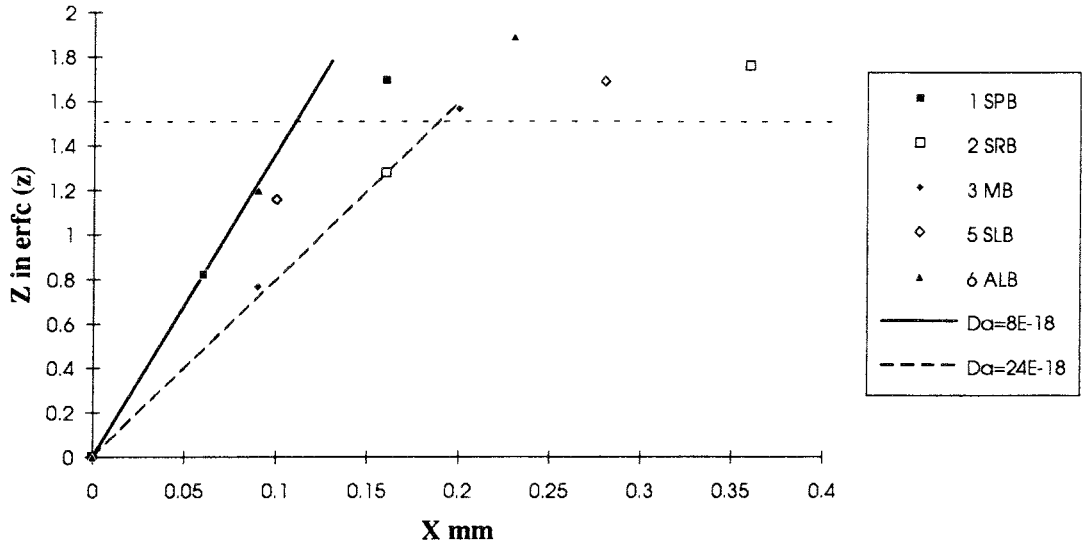

Figure 6.14. "Diffusion" of plutonium in concrete. Five types of concretes. Dotted line shows estimated sensivity.

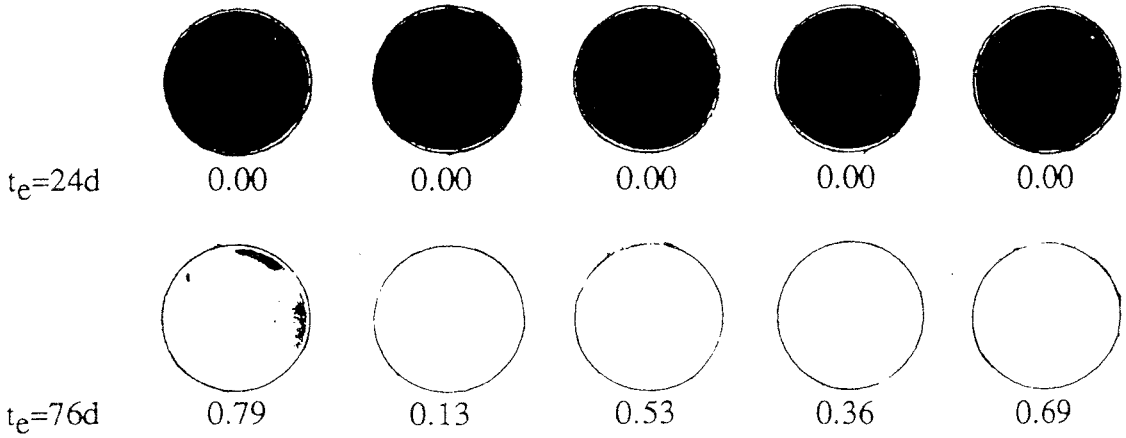

Figure 6.15. Autoradiographs showing penetration of plutonium in five different concrete samples, from left SPB, SRB, MB, SLP and ALB. Depths are given in mm, and t_e is the exposure time.

7 CONCLUSION

Sodium showed as expected very low retention both in the concrete and in the bentonite clay in contact with the concrete. The measured diffusivity ($\sim 5 \cdot 10^{-11}$ m^2/s) seems only dependent on the porosity and constrictivity of the solid. For calcium, an apparent diffusivity of $3 \cdot 10^{-11}$ m^2/s is obtained in the bentonite. This is the same value that is expected in the concrete [ALL91], and little sorption is obtained in both cases.

Cesium shows an apparent diffusivity of $3 \cdot 10^{-12}$ m^2/s in the bentonite. The same value is obtained with an other type of experimental set-up [TOR85, ALB89]. This shows that measured D_a-values are independent of how the experiment is performed.

The apparent diffusivity of Cs in normal types of concrete is about $1 \cdot 10^{-13}$ m^2/s. This has been shown both in this investigation, by Andersson et al. [AND81, AND83] and by Johnston et al. [JOH92].

For plutonium and americium no movement could be measured even though the contact times was extremely long (5 y and 2.5 y, respectively). The maximum estimated diffusivity with these contact times and this experimental setup is in both cases around 10^{-17} m^2/s. This low diffusion can be explained by the low content of carbonate in the pore water, giving little complexation of Am and Pu with carbonate. Also the low diffusivity indicates that no negatively charged hydroxi species are formed.

8 ACKNOWLEDGEMENT

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[2] CEA, France
[3] Etude Recherche Materiaux (ERM), France
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An approach to quality classification of deep groundwaters in Sweden and Finland

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TR 93-28

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Costs for management of the radioactive waste from nuclear power production

Swedish Nuclear Fuel and Waste Management Co
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