

**Diffusion and sorption properties of
radionuclides in compacted bentonite**

Ji-Wei Yu, Ivars Neretnieks

Dept. of Chemical Engineering and Technology,
Chemical Engineering, Royal Institute of Technology,
Stockholm, Sweden

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SVENSK KÄRNBRÄNSLEHANTERING AB

SWEDISH NUCLEAR FUEL AND WASTE MANAGEMENT CO

P.O.BOX 5864 S-102 40 STOCKHOLM SWEDEN

PHONE +46 8 665 28 00

FAX +46 8 661 57 19

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

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Department of Chemical Engineering and Technology
Chemical Engineering
Royal Institute of Technology
S 100 44 Stockholm, SWEDEN

Abstract

A literature review of diffusion and sorption properties of mainly ionic species in water saturated compacted bentonite has been made. Experimental methods used to determine diffusivities have been critically reviewed and sources of error analysed. The diffusion of charged species in narrow pores with charged surfaces and ion exclusion and surface diffusion effects are discussed. A detailed compilation of all found data is included. Recommended values for use in a performance assessment and the rationale for choosing them is given.

Kort sammanfattning

En litteraturundersökning rörande diffusion och sorption av främst små joner i vattenmättad bentonit har gjorts. Experimentella metoder som används för diffusionsmätningar har granskats och felkällor har beskrivits och analyserats. Diffusion av joner i trånga porer med laddade ytor och därav orsakad jonutträngning och ytdiffusion diskuteras. En stor mängd litteraturdata redovisas. Rekommenderade sorptions- och diffusionsdata för användning i en säkerhetsanalys ges och skälen till hur data valts presenteras.

CONTENTS

	Page
SUMMARY	3
1 INTRODUCTION	3
2 SORPTION PHENOMENA	9
3 DIFFUSION PHENOMENA	14
4 SORPTION EQUILIBRIUM DATA AND DIFFUSION COEFFICIENTS OF RADIONUCLIDES IN COMPACTED BENTONITE	35
5 DISCUSSION AND CONCLUSIONS	88
NOTATION	92
REFERENCES	93

SUMMARY

In this report, recent studies on sorption and diffusion of radionuclides in compacted bentonite have been reviewed. The sorption distribution coefficient and diffusion coefficient data obtained from experiments in the literature have been compiled. Based on these experimental data and the report SKB TR 91-16 (Brandberg and Skagius, 1991), this report proposes a set of sorption distribution coefficient and diffusion coefficient values for modelling purpose for safety analysis of nuclear waste repositories.

The variability and uncertainty of the diffusivity data span somewhat more than an order of magnitude up and down. Most of the nuclides have an effective diffusivity in around 10^{-10} m²/s.

Ion exclusion effects are observed for C, Cl and for Tc in oxidizing waters. Effective diffusivities are nearly two orders of magnitude lower for these elements and of the order of 10^{-12} m²/s. Surface diffusion effects are found for Cs, Ni, Pa, Pb, Ra, Sn, Sr and Zr. Effective diffusivities for these elements are of the order of 10^{-8} m²/s. The surface diffusion effect should decrease in saline waters which is seen for Cs and Sr where there are data available. It is also deemed that Ra will have this effect because of its similarity with Sr. The other nuclides should also show this decrease but no data are available.

Sorption and diffusion mechanisms in compacted bentonite are discussed in the report. In highly compacted bentonite, sorption and hence its distribution coefficient is not well-defined, and a pore diffusion coefficient or a surface diffusion coefficient is not well-defined either. Therefore, an apparent diffusion coefficient and a total concentration gradient should be more relevant in describing the diffusion process in compacted bentonite.

Different methods for diffusion coefficient measurements are discussed in the report.

1 INTRODUCTION

Compacted bentonite has been considered to be a back-fill material in repositories of nuclear waste. Bentonite is a good barrier for containment of radionuclides due to its sorption capability and low permeability. Safety analysis of the repository involves modelling of the radionuclide transport from the spent fuel to the environment around the repository, which requires the understanding of the diffusion and sorption processes of the nuclides in the compacted bentonite and reliable data for diffusion coefficients and sorption equilibria. In the technical report SKB TR91-16 (Brandberg and Skagius, 1991), diffusion and sorption data were provided for many radionuclides in bentonite. Since then, more studies have been conducted on this subject. The purpose of this report is to collect data from recent studies by literature review and to incorporate the new information with the contents in SKB TR91-16.

The parameters considered in this report are the sorption distribution coefficient K_d , the apparent diffusion coefficient D_a , and the effective diffusion coefficient D_e . The elements included in this report are Am, C, Ce, Cl, Cs, I, Nb, Ni, Np, Pa, Pb, Pd, Pu, Ra, Rn, Se, Sm, Sn, Sr, Tc, Th, U, and Zr.

Bentonite is a swelling clay due to its content of the smectite clay mineral montmorillonite. Montmorillonite has a considerable cation exchange capacity with generally sodium and/or calcium in the exchange positions. Bentonite also contains small amounts of other minerals, such as quartz, feldspars, other clay minerals, and trace amounts of pyrite and organic carbon (Wiborgh, 1995).

Two commercial bentonites, Wyoming MX-80 and Avonlea, are described below as examples.

The Wyoming MX-80 bentonite is a sodium bentonite. It consists to a large extent (85%) of a small size fraction that is smaller than $2 \mu\text{m}$ (Wiborgh, 1995). The average mineral composition of MX-80 bentonite is given in Table 1.

The water content of a 100% saturated Wyoming MX-80 bentonite with a bulk density of $2000\text{-}2100 \text{ kg/m}^3$ is 20-25% (Brandberg and Skagius, 1991). Muurinen et al. (1987) reported a water content of 40% in MX-80 bentonite for a wet density of 2000 kg/m^3 , and 25% for a dry density of 2100 kg/m^3 , which corresponds to a bulk density of 2350 kg/m^3 .

Table 1. Mineral composition of MX-80 Bentonite (Lehikoinen et al., 1996).

Mineral	Content (wt%)
Montmorillonite	65 - 80
Quartz	15.2
Feldspar	5-8
Mica	<1
Kaolinite	<1
Carbonate	1.4
Sulphate	0.34 - 0.8
Sulphide	0.1 - 0.3
Phosphate	0.1
Fluoride	0.1
Organic Matter	0.3 - 0.4

The Avonlea bentonite is from the Bearpaw Formation of Late Cretaceous age in southern Saskatchewan, Canada. The clay contains ~80 wt% montmorillonite, ~10 wt% illite, ~5 wt% quartz, and minor amounts of gypsum, feldspar and carbonate (Choi and Oscarson, 1996). It has a cation-exchange capacity of ~60 cmol/kg and a specific surface area of 480 m²/g. Na⁺ is the predominant exchangeable cation in the Avonlea bentonite (Oscarson et al., 1994).

The characteristic features of compacted bentonite are its large surface area, small pore size, the negative surface charge, exchangeable cations which balance the negative charge, the hydration of exchangeable cations, and the formation of a double layer on the surfaces. One special feature is the strong exclusion of anions due to the negative surface charge and the narrow interlamellar space. As a consequence of the anion exclusion, the accessible porosity of anions is much less than the water porosity (Muurinen et al., 1994).

The surface area of bentonite depends on the degree of packing. It was reported that, when non-compacted, the Wyoming bentonite had a specific surface area of 54 m²/g. The same material, compressed to form a plug with a porosity of 0.25, indicated an apparent specific surface area of 20 m²/g. The adsorption of nitrogen on water-dried aggregates with a porosity of 0.15 was found to be negligible and the specific surface area was less than 1 m²/g (Muurinen et al., 1994).

The microstructure of compacted bentonite is characterised by a domain-type arrangement of the clay platelets, which tends to become increasingly homogeneous in the course of the water saturation process (Pusch, 1992).

Na-bentonite consists of 10 Å elementary flakes forming stacks with a thickness of 50-200µm depending on the bulk density and porewater chemistry. The interlamellar space, the voids between the flakes within a stack, hosts "internal", ordered water forming three hydrates at maximum, while the voids separating the stacks contain water in various physical conditions ("external water") (Pusch and Hökmark, 1990). X-ray diffraction (XRD) analysis showed that the interlayer spacing of Na-bentonite was 0.30 nm (Kozai et al., 1993). After contacting with a solution of pH 2.5 at 20°C for 10 days, the interlayer spacing became larger (5.3 nm). Smectite's interlayer surfaces exert an influence on the water molecules within a 10-Å distance from the layers. Beyond that, water molecules behave just like in liquid water. The interlamellar water does not possess a rigid "ice-like" structure but a dynamic one. Water molecules interact with smectite's basal surfaces and with the interlayer cations within the interlayer space in a dynamic manner (Güven, 1990). Pusch and Hökmark (1990) proposed a basic model to describe the microstructure of compacted bentonite. The key feature of the microstructural model is that the dense grains constitute a basic network with continuous "external" voids of varying size. The grains contain internal space. At hydration of the system under confined conditions the grains expand and reduce the porosity, and the remaining voids become filled by soft clay gels that emanate from the dense grains.

The distribution of internal and external water is naturally determined by the bulk density. A rough estimation of the ratio of internal to external water in compacted bentonite was made on the basis of anion concentration in the diffusion measurements (Muurinen et al., 1994). The fraction of internal water of the total water content evaluated was 51% at a dry density of 1200 kg/m³ and 77% at a dry density of 1800 kg/m³. A theoretical relationship between the dry bulk density and the content of internal water was presented by Pusch et al. (1990). At a dry density of 1600 kg/m³, the content of internal water may reach 94% of the total water content for Na-bentonite and 84% for Ca-bentonite.

The sorption and diffusion behaviour of radionuclides in compacted bentonite depend on the water that is in contact with the bentonite. Table 2 gives the groundwater composition in the Finnsjön area in Sweden.

Table 2 Groundwater composition at some sites in Sweden.

Site	Äspö	Finnsjön	Finnsjön	Gideå
Bore hole	KAS02	BFI01	KFI07	KGI07
pH	7.7	7.0	7.9	9.3
Eh mV	-308	-	-250	-202
	mg/l	mg/l	mg/l	mg/l
Na ⁺	2100	1700	275	105
K ⁺	8	13	2	2
Ca ⁺⁺	1890	1650	142	21
Mg ⁺⁺	42	110	17	1
Sr ⁺⁺	35	21		
Fe ⁺⁺			1.8	
Mn ⁺⁺		0,8		
HCO ₃ ⁻	10	47	278	18
SO ₄ ²⁻	560	370	49	
Cl ⁻	6410	5500	555	178
Br ⁻	40	32		
F ⁻	1.5	1.2	1.5	3.2
Ionic strength	0.247	0.215	0.028	0.006

In granitic rock at depths where a potential nuclear fuel waste disposal vault would be located, groundwater generally has a relatively low redox potential (-100 to -200 mV) due to the absence of dissolved O₂ and the presence of reduced Fe, S, and Mn species (Sawatsky and Oscarson, 1991a). On the other hand, impurities in the clay barrier material and radiolysis effects may make the environment in a disposal vault more oxidizing.

Bentonite is likely to buffer a water solution, at 25°C, to a pH-value within the range of 8-10, and to an Eh-value in the range of -0.3 to -0.2V (Albinsson et al., 1996). The low Eh value may be attributed to the content of pyrite and organic matter (Wiborgh, 1995). Bentonite was elsewhere reported to exhibit a rather wide range of pH buffering effects, from pH 3 to pH 12 (Liang et al., 1993)

The hydrothermal stability of smectites ranges from 50°C to 850°C. Stability is not restricted to the chemical and structural survival of the smectite mineral in the repository environment but its continued performance of the buffer functions for a few million years. Temperature in such an environment may range probably from 200°C at the clay/waste container interface to the ambient temperature in the host rock/clay contact. Fe-rich smectites are known to be more easily altered and thermally less stable than the magnesian and aluminian varieties. Interlayer cations like Na⁺, Li⁺, Ca²⁺, and Mg²⁺, on the other

hand, enhance the stability of aluminous smectite above 300°C. Na⁺ is well-known for its role in greatly improving various physical properties of bentonites (Güven, 1990).

2 SORPTION PHENOMENA

Sorption mechanisms in bentonite

It is well-known that the surfaces of smectite clays carry a permanent negative charge, which is mainly a result of isomorphous substitution of lower-valency atoms for higher-valency atoms within the clay lattice. This charge deficiency is manifested at the surface of clay platelets where charge neutralisation takes place by exchangeable cations. When clay is water-saturated, the exchangeable cations are hydrated and may form electric double layers at the water-solid interface (Muurinen and Lehtikoinen, 1995).

The charge-neutralising cations are readily exchangeable, which means that they can be replaced by other cationic species in the solution in contact with the clay surface. The sorption reactions of cations in montmorillonite can, to a reasonable extent, be described by ion-exchange reactions and modelled by thermodynamic equilibrium constants or selectivity coefficients (Muurinen and Lehtikoinen, 1995). In the experiments by Abou-Jamous (1992), the sorption for Cs and Sr increased with pH, as would be expected by an ion exchange process. The strong ionic strength dependence of sorption was more effective for Sr than Cs, which also indicated that ion exchange was a dominating sorption mechanism (Abou-Jamous, 1992). However, the sorption data obtained from experiments often seem to be somewhat less than the idealized ion-exchange model would predict. Many complexing agents, e.g., CO_3^{2-} , in the water-bentonite system can form anionic complexes with cations to decrease their sorption. In equilibrium modelling, the major competing reactions should thus be considered (Muurinen and Lehtikoinen, 1995).

Radionuclides may also be sorbed by reactions with surface functional groups. A surface functional group is a chemically reactive molecular unit which is in such a place in a structure that it can be reached by a molecule in solution. Because the functional groups are fixed to the solid structure, their reactivity is a cooperative property which depends on the neighbouring groups and their reaction state. When a surface functional group reacts with a molecule dissolved in a surrounding solution to form a stable unit, a surface complex is said to exist and the formation reaction is termed surface complexation. Two categories of surface complexes can be distinguished. If there is no water molecule between the surface functional group and the bound molecule, the complex is called an inner-sphere complex. If at least one water molecule is between the functional group and the bound molecule, the complex is called an outer-sphere complex. As a rule, outer-sphere surface complexes involve electrostatic bonding mechanisms and therefore are less stable than inner-sphere surface complexes, which necessarily involve either ionic or

covalent bonding. The principal kinds of surface functional groups found in clays are siloxane ditrigonal cavities and the hydroxyl groups (Muurinen et al., 1994).

Physical adsorption occurs when the species attach to the clay surfaces by van der Waals forces (e.g., a dispersion or a dipolar interaction). Van der Waals interactions have a long range but are weak. Physical adsorption is reversible. The energy released when a species is physically adsorbed is small, which is insufficient to lead to bond breaking, and so a physically adsorbed species retains its identity, although it might be distorted by the presence of the surface.

Factors influencing sorption in compacted bentonite

Various experiments have shown that the sorption of cations on bentonite depends on the concentration of the competing ions in the solution (Muurinen and Lehtikoinen, 1995).

Competitions of sorption among the ions on bentonite have been investigated (Horyna and Dlouhy, 1988). For the sorption from the water solution containing 0.001 M Na, 0.002 M Ca, and 0.001 M Mg, the K_d values of Sr are one to two orders of magnitude lower than those of Cs. This was believed to be due to low selectivity of the bentonite for Sr^{2+} in the presence of its chemical analogue - Ca^{2+} due to its chemical similarity. From the results of Horyna and Dlouhy (1988), it was evident that the most competitive ion for Co^{2+} and Sr^{2+} is H^+ , while for Cs^+ , the most competitive ion is K^+ .

Experiments have shown that easily soluble compounds in bentonite can readily dissolve upon saturation with water leading to an increase in the ionic strength of the porewater and being a potential reason for the decreased sorption of radionuclides by ion exchange (Lehtikoinen et al., 1996). Pusch et al. (1989) indicated that protons of exposed lattice hydroxyls are exchangeable, which may yield considerable changes in the pH of porewater. Speciation in the porewater, sorption and diffusion will influence each other. If one species preferably diffuses or is sorbed, the speciation in the porewater is re-established.

Sorption properties are closely related to the surface charge of bentonite, which varies with the exchangeable cations. Güngör and Tulun (1994) measured the zeta potential of both Na-bentonite and Ca-bentonite. Ca-bentonite was found to have lower zeta potential than Na-bentonite. The zeta potential of Na-bentonite ranged from -4.24 mV to -4.67 mV, while the zeta potential of Ca-bentonite ranged from -3.54 mV to -3.72 mV.

In many studies, only ion exchange arising from the layer charge has been considered, where displacement by the proton is negligible at $\text{pH} > 4$ (Wanner et al., 1992). However, the edge surface, which consists of hydrolysed AlOH and SiOH functional groups, also carries charges which depend on pH and can be positive or negative. In the alkaline pH range, deprotonation of the OH groups at the edge face of a montmorillonite platelet will cause negative charge and the edge surface is considered to constitute 10 to 15% of the total montmorillonite surface charge (Wanner et al., 1992). Cations, including protons, compete for these negatively charged edge sites.

Experimental results showed that compacted clays preferred monovalent over divalent cations while in loose clays the situation was just the opposite. This kind of selectivity could be due to the difference in the structure of the tactoids. In compacted clays, the tactoids cannot open because of the lack of space (Muurinen and Lehtikoinen, 1995).

The nuclear waste will contain certain amounts of organic material, such as ion exchange resins, plastic, rubber, and cellulose, etc. The degradation of the organic components under the conditions in the repository will generate a fraction of soluble organic components that may influence the solubility and adsorption behaviour of the radionuclides and thereby their releases and migration out of the repository. The conclusion from a number of studies (Wiborgh, 1995) is, nevertheless, that the degradation products from ion-exchange resins, various natural and synthetic polymers, plasticizers, and hydrocarbons would have only a minor influence on the releases of radionuclides from the repository, with the exception of certain categories of extremely strong complexing agents like the aminopolycarboxylic acids (NTA, EDTA, DTPA). However, these complexing agents are expected to be present in very limited quantities in the nuclear waste.

The influence of irradiation on the sorption capacity of bentonite was studied by Nikiforov et al. (1991). The results showed that irradiation of the bentonite specimen at 300 K up to absorbed doses in the range 108 - 109 Gy altered the sorption capacity a little with respect to cesium, while a combination of irradiation and raised temperature to 473 - 573 K resulted in a rapid decrease in the amount of sorbed cesium. After the most severe radiation-thermal stresses, the sorption capacity of bentonite with respect to cesium diminished by more than 70%, and the sorption of Cs occurred only by ion exchange and physical adsorption mechanisms, i.e. without fixation at all. This was attributed to the decrease in the negative charge on the layer and of the concentration of OH groups on the edges of the crystals (subject to the most intensive radiolysis) as a result of the rearrangement of the structure with the transfer of some of the cations from the exchange to the non-exchange positions under the radiation-thermal conditions. The decrease in the

negative charge would cause the irreversible compression of the aluminosilicate layers and hence decrease the accessibility of the interlayer gaps of the bentonite to cesium (Nikiforov et al., 1991).

Sorption in compacted bentonite depends also on the density of the bentonite. Oscarson et al. (1994) measured the sorption of cesium on compacted Na-bentonite. Over the clay density range from 500 to 1500 kg/m³, K_d values for Cs⁺ with compacted bentonite were about one-half to one-third the value of those with loose bentonite.

Sorption equilibrium modelling

In most studies concerning radionuclide transport in the nuclear waste repositories, the sorption equilibrium is commonly described by a linear relation between the concentration in the sorbed phase and the concentration in the solution phase, characterised with a distribution coefficient K_d , which is defined as:

$$K_d = \frac{q}{C_w} \quad (1)$$

where q is the concentration in the sorbed phase and C_w is the concentration in the solution phase. At low concentrations of the species in the solution, this linear approximation should describe the sorption equilibrium satisfactorily. Recently, attempts have been made to describe the sorption equilibrium with multiparameter models, e.g., the Freundlich isotherm, the Langmuir isotherm, and the ion-exchange model.

Sorption data from the literature show that K_d varies with the salt concentration in the solution. K_d also varies with the concentration of the radionuclide itself. This indicates that the sorption equilibrium cannot always be represented by a single K_d . Non-linear, multicomponent sorption equilibrium models may give better results.

Puigdomènech and Bergström (1994) proposed that, to reflect the correct dependence on chemical parameters like pH, etc., plausible K_d may be estimated by surface complexation models when the complexation equilibrium constants in the model are obtained under well-controlled laboratory conditions on well determined minerals.

As some K_d values vary at different conditions, when one uses the K_d value under the condition outside the experimental scope, one should see if the sorption is overestimated because in such cases the retardation effect of the sorption will also be overestimated.

Especially when one uses a K_d value obtained from a low concentration experiment for high concentration conditions, one would probably overestimate the sorption.

The sorption isotherm of Tc in bentonite under reducing condition showed a remarkable change in the slope (Ito and Kanno, 1988), which could suggest the precipitation of Tc. To model the partition between the solution phase and the surface phase under such conditions, it could be important to consider describing mathematically the sorption and the precipitation separately, because the former has a continuous isotherm while the latter has a discontinuity in the equilibrium relation.

Diffusion measurements, whose primary goal is to determine apparent diffusion coefficients, are not suitable to determine reliable K_d values because a couple of uncertain parameters (D_p and ϵ) are involved (Wanner et al., 1994).

It has been stated that care should be taken when applying sorption data to compacted bentonite. Most of the water content in compacted bentonite is the internal water. The interlamellar space would contain maximum 3 water layers (Pusch and Hökmark, 1990) and therefore no distinct solution phase and sorbed phase would exist. Consequently, the sorption equilibrium relation established in the batch experiments would no longer apply to the interlamellar space, where K_d is not well-defined. Nevertheless, in some carefully conducted experiments (Eriksen and Jansson 1996) no differences were found between S_r and C_s K_d -values in batch experiments with suspended bentonite particles and in compacted bentonite with a dry density of 1800 kg/m^3 . The authors of the present report suspect that in many cases where differences are found this may be due to errors in measurement techniques. See the below section on measurement techniques especially the discussion of "Method I".

3 DIFFUSION PHENOMENA

Diffusion equations

The diffusion process in a porous medium is described as follows, according to Fick's first law:

$$J = -D_p \varepsilon \frac{dC_p}{dx} \quad (2)$$

where J is the diffusive flux, D_p is the diffusion coefficient in the pore solution, ε is the porosity of the porous medium, C_p is the concentration of the diffusing substance in the solution, and x is the spatial coordinate.

The pore diffusion coefficient, D_p , is less than the diffusion coefficient in an unconfined solution, D_w . This is because of the tortuous winding of the pores, which increases the length of the diffusion path, and because the pores may be constricted in some parts. The relation between D_p and D_w is:

$$D_p = D_w \frac{\delta}{\tau^2} \quad (3)$$

where δ is the constrictivity and τ^2 is the tortuosity of the pores.

For sorbing species, there are experimental indications of an additional diffusion mechanism, surface diffusion. The cause of surface diffusion could be that the tracer cations, which are sorbed among the exchangeable cations, are hydrated in the water saturated clay, and can change place by diffusion. They are, however, bound to the proximity of the bentonite surface, and in that sense are sorbed by ion exchange (Muurinen and Lehtikoinen, 1995).

Fick's second law, or the diffusion equation, describes the concentration of a diffusing species as a function of time and space. For diffusion of a sorbing species in a porous material, this is mathematically given by:

$$\varepsilon \frac{\partial C_p}{\partial t} + \rho \frac{\partial q}{\partial t} = \varepsilon D_p \frac{\partial^2 C_p}{\partial x^2} + \rho D_s \frac{\partial^2 q}{\partial x^2} \quad (4)$$

where ρ is the bulk density of the material, and D_s is the surface diffusion coefficient. If the linear sorption isotherm is assumed, the equation can be written as:

$$\frac{\partial C_p}{\partial t} = \frac{D_e}{\varepsilon + K_d \rho} \frac{\partial^2 C_p}{\partial x^2} = D_a \frac{\partial^2 C_p}{\partial x^2} \quad (5)$$

where D_e is the effective diffusion coefficient, and D_a is the apparent diffusion coefficient, which are defined as:

$$D_e = \varepsilon D_p + K_d \rho D_s \quad (6)$$

and

$$D_a = \frac{D_e}{\varepsilon + K_d \rho} \quad (7)$$

As the linear sorption isotherm is assumed, the solution concentration in Equation 5 can even be replaced by the total concentration in the porous material by multiplying $\varepsilon + K_d \rho$ on the both sides of the equation, which leads to:

$$\frac{\partial C}{\partial t} = D_a \frac{\partial^2 C}{\partial x^2} \quad (8)$$

Where C is the total concentration. Equation 8 is the diffusion equation widely used in diffusion studies for radionuclides in bentonite. For the diffusion of non-sorbing species in bentonite, D_a is equal to D_p .

In the compacted bentonite, most of the water content is the internal water. As indicated previously, no distinct solution phase and sorbed phase would exist and K_d is not well-defined in the interlamellar space in the compacted bentonite. Therefore, it is appropriate, in our opinion, to use Equation 8 for diffusion in the compacted bentonite with the apparent diffusion coefficient and the total concentration instead of Equation 5.

Surface diffusion

In many studies, contradictions between the apparent diffusion coefficients of cations and their distribution coefficients have been considered. The measured diffusion coefficients have been greater than what the sorption values would suggest on the basis of the

traditional diffusion-sorption theory. Two principal reasons have been offered as explanations for the obtained contradiction: surface diffusion and a decrease in the distribution coefficient in compacted bentonite compared to that in batch experiments (Muurinen and Lehtikoinen, 1995).

Surface diffusion has been suggested to be an important migration mechanism (Neretnieks, 1982; Rasmuson and Neretnieks, 1983). Lehtikoinen et al. (1996) compiled the information available from the open literature and concluded based on the compiled data that surface diffusion most probably plays a significant role in the transport of cations in bentonite clays. Pusch et al. (1989) concluded that uranium migrates in bentonite by both pore diffusion and surface diffusion. The latter mechanism is dominant, especially in dense clay.

Surface diffusion can be explained by cationic diffusion in the electric double layer. Unlike in the solution phase where the migration of the ions is decreased by the ionic atmosphere, in the double layer, the lack of anions diminishes the effect of the ionic atmosphere on cations, which may partly compensate the interaction from the mineral surface and hence allow the cations to migrate quite freely in the double layer. The electrostatic field from the negatively charged surface gives the cations a force perpendicular to moving direction along the surface, while the effect of the ionic atmosphere is always against the migration of the ions. The sorbed but mobile species form an additional route with a much higher concentration gradient than in pore water, although the diffusion coefficient near the surface can be lower than in the pores.

The measurements on the apparent diffusion coefficients of cations in compacted bentonite and the sorption measurements with loose and compacted samples have shown that the effect of solution ionic strength on the apparent diffusion coefficient is small. This means that although the ionic strength changes due to dissolving compounds in bentonite, the effect on the apparent diffusion coefficient is small and cannot explain the high diffusion coefficients obtained. Through-diffusion measurements and electrical conductance measurements in clays offer further evidence for the surface diffusion (Muurinen and Lehtikoinen, 1995).

Cheung (1989) discussed the time dependence of the apparent diffusion coefficient and attributed this to the difference in diffusion coefficients of pore diffusion and surface diffusion. It was found that in the diffusion experiments for Na^+ in bentonite, D_a decreased with time during the first 12 hours and then remained constant. The results of anion I^- diffusion in compacted bentonite also indicated the effect of the heterogeneous

diffusion. However, in the study of Miyahara et al. (1991), the diffusion time did not have a significant effect on the apparent diffusion coefficient of Cs.

Kim et al. (1993) concluded from their Cs, Sr and Cl diffusion experiments that migration of cations through the compacted Na-bentonite was controlled by the surface diffusion due to the sorption. Kim et al. (1993) observed that, although K_d values of Sr and Cs cations sorbing onto Na-bentonite were rarely different, the diffusion coefficient of Sr obtained from their diffusion experiment was up to 30 times larger than that of Cs with increasing bentonite density. Kim et al. (1993) further indicated that it cannot be concluded that the cation having a higher K_d value always migrates through the compacted clay slower than the cation having a lower K_d value.

It has been emphasised (Eriksen and Jacobsson, 1984) that for compacted bentonite the water content corresponds roughly to 2-4 water molecule layers. There is therefore really no "free water" and the measured diffusion coefficient can be regarded as corresponding approximately to the diffusion coefficient within the sorbed phase (Eriksen and Jacobsson, 1984).

There are very few systematic studies on surface diffusion. One notable exception is that by Eriksen and Jansson (1996). They studied diffusion and sorption of Cs and Sr in bentonite compacted to 1800 kg/m^3 in through diffusion experiments. Two different ionic strengths were used, 0.218 and 0.018. In addition, sorption was measured in batch experiments over a large concentration range. Concentration profiles in the compacted bentonite samples as well as the flux of tracer through the samples were used to evaluate diffusivities and sorption coefficients. No difference in sorption capacity was found between clay in suspension and compacted clay. Their own data, those of Muurinen (1994) and Oscarson (1992) clearly showed that surface diffusion was very important at high K_d 's. The D_a and D_e values vary considerably with K_d as this changes with the ionic strength of the solution. These data were used to evaluate the pore and the surface diffusion coefficients by curve fitting. This is equivalent to plotting D_e vs. K_d as can be seen in Equation 6. The D_p and D_s values so obtained were constant in the studied range.

Sorption effect on diffusion

An alternative way to explain the excess diffusion coefficients of the cations is that the distribution coefficient in the compacted bentonite is less than that obtained from batch experiments for loose bentonite (Muurinen and Lehtikoinen, 1995). One possible explanation for this phenomenon is that all the surfaces in the compacted clay are not

accessible to the cations and therefore the distribution coefficient decreases (Oscarson, 1994). Another explanation is that the actual ionic strength in the compacted clay is higher than that in the batch experiments with loose bentonite, which decreases the sorption (Muurinen and Lehtikoinen, 1995).

The D_a values for the diffusants were reported to be inversely proportional to K_d (Choi and Oscarson, 1996), while Miyahara et al. (1991) concluded that K_d determined from batch experiments with high solution/bentonite ratio is not applicable to predict D_a .

The batch K_d 's of Cs^+ and Sr^{2+} determined for compacted MX-80 bentonite in contact with NaCl solutions of varying concentrations (0.001-1.2 M) were more or less equal to those for loose samples and, thus, cannot explain the high apparent diffusion coefficients obtained (Lehtikoinen et al., 1996).

Impact of compaction of bentonite on diffusion

The transport rate of radionuclides through compacted bentonite is influenced by the degree of compaction. Whether this is due to variation in porosity or variation in sorption is uncertain (Wanner et al., 1994).

The diffusion coefficients of carbon in Avonlea bentonite tended to decrease with an increase in clay density (Oscarson and Hume, 1994). The decrease in D_a of iodide with increasing density of bentonite was also observed, which was attributed to an increase in the tortuosity as the density increased. For iodide, there appeared to be a critical dry density value for Avonlea bentonite of about 1400 kg/m³, beyond which the flux of iodide from the clay could be markedly decreased (Oscarson et al., 1992).

The apparent diffusion coefficients were reported to decrease with increasing dry density of bentonite for 3H , ^{90}Sr , ^{99}Tc , ^{129}I , ^{137}Cs , ^{237}Np , and ^{241}Am (Sato et al., 1992). The decrease in D_a of Tc with an increase in clay density was largely attributed to an increase in the anion exclusion volume in the clay with increasing clay density, which would likely decrease the effective size of the pores (Sawatsky and Oscarson, 1991a).

The amount of external water depends on the compaction of the bentonite. With increasing compaction the amount of external water decreases, and so the water volume, where the diffusion of anions can occur, decreases (Muurinen, 1994).

Ramebäck et al. (1994) correlated the apparent diffusion coefficient of Sr in Na-bentonite Wyoming MX-80 to the clay dry density with an exponential relation, which correlated their measured data well. Relyea et al. (1986) correlated the effective diffusion coefficient of Cl to the clay bulk density and the temperature. The relation between the effective diffusion coefficient and the bulk density was also exponential. An exponential correlation between the apparent diffusion coefficient of Tc and the dry density of bentonite was presented by Sawatsky and Oscarson (1991a).

Kim et al. (1993) compared the dependency of the diffusion coefficients of Cs and Sr on the bentonite density. With increasing bentonite density, the diffusion coefficient of Cs decreased more rapidly than that of Sr. This phenomenon may be explained by the increase in the fraction of immobile unhydrated cation adsorbed on the clay surface with the increase in the density of the bentonite. Using the electric double layer theory, it was verified that the fraction of immobile unhydrated cation specifically adsorbed on the clay surface increased rapidly with increasing clay density. The fraction of mobile hydrated cation decreased considerably (Kim et al., 1993). This effect was greater when the cation has a lower valence and a larger ionic radius. Therefore, it was thought that Cs, which is a monovalent cation and has an ionic radius larger than that of Sr, may respond to the increase in clay density more than Sr which is a divalent cation. Kim et al. (1993) concluded that it is important to consider the hydration status of cation sorbed on the surface of clay rather than the distribution coefficient.

The effective porosities ($\epsilon_e = D_e/D_a$) calculated from the diffusion measurements for tritium by Kato et al. (1994) were found to be very similar to the theoretical total porosities, which implies that all the pores in compacted bentonite are actually available for mass transfer of neutral species (Muurinen and Lehtikoinen, 1995).

Ion exclusion

As noted by Muurinen (1992), it seems likely that there are different diffusion and sorption mechanisms and also different diffusion routes in compacted bentonite depending on the speciation of the diffusing element. The ion exclusion arises when the compaction of a clay suspension induces an overlap of double layers of adjacent platelets or particles (Lehtikoinen et al., 1996). Anions cannot squeeze into the narrow interlamellar pores of compacted bentonite due to the electrostatic forces between the negatively charged bentonite surfaces and the anion. This ion-exclusion decreases the porosity available for diffusion, especially in compacted bentonite. In saline conditions the effect of ion-exclusion is smaller and diffusion rates will be higher. In sand-bentonite mixture,

the effect of ion-exclusion has been evaluated to be of smaller importance and is neglected (Muurinen, 1992).

Cheung (1989) conducted the diffusion experiments for Cs^+ and I^- in compacted bentonite by using Method III (described later). It was observed that the flux for I^- was much less than that for Cs^+ .

The anion-exclusion volume is affected by the salinity of the pore solution of the bentonite: the greater the salinity, the thinner the electric double layer, and hence the less the anion-exclusion volume. The effective diffusion coefficient for anions should, therefore, increase with increasing salinity (Oscarson et al., 1992). With highly compacted bentonite, however, the salinity would have little effect on anion-exclusion volume because most of the anions are already excluded due to the compressed interlayer space. This is not born out by experiments. See below.

Cho et al. (1993) indicated that in clay-water systems, electroneutrality must be maintained in any macroscopic region and so no ionic species can move independently. The flux of ions with one sign of charge must be comparable to the flux of the opposite charge (Cho et al., 1993). However in diffusion experiments usually only one mobile ion is monitored and there is no information on how the other ions in the system move to maintain the electroneutrality. Eriksen and Jansson (1996) found that the effective porosity for I^- diffusion decreased from between 0.06-0.11 for a high ionic strength water (0.218) to 0.006 for an ionic strength of 0.018 in a compacted bentonite with a dry density of 1800 kg/m^3 . The real porosity then was 0.32.

Influence of exchangeable ions

The interlamellar space varies with the exchangeable cations. Sodium bentonite flakes may be completely separated and the distance between the platelets is then determined by diffuse double layer interaction. Calcium montmorillonite exists in stacks consisting of 3-9 lamellae with a 4-5 Å hydrate in each interlamellar space (Muurinen et al., 1994). The calcium bentonite does not have the swelling ability of the sodium form (Albinsson et al., 1996).

Choi and Oscarson (1996) examined the effect of exchangeable cation Na^+ and Ca^{2+} on the diffusive transport of I^- , Sr^{2+} and ^3H in compacted bentonite using a through-diffusion method. They observed that the effective diffusion coefficients determined by concentration profiles for all diffusants were 2 to 6 times higher in the Ca- than Na-

bentonite. They attributed this to the larger quasicrystal, or particle, size of Ca- compared to Na-bentonite. Hence, Ca-bentonite had a greater proportion of relatively large pores. This was confirmed by Hg intrusion porosimetry. This meant that the diffusion pathways in Ca-bentonite were less tortuous than those in Na-bentonite (Choi and Oscarson, 1996).

As Ca^{2+} gradually replaces Na^+ on the exchange complex of bentonite in a disposal vault, the diffusion coefficients for radionuclides in a bentonite barrier would be expected to increase accordingly. The increase, though, would only be about a factor of 5, at most (Choi and Oscarson, 1996). On the other hand, when compacted, clay layers have restricted mobility. Hence they may be unable to assume other arrangements, for example, more unit layers per quasicrystal as the proportion of Ca^{2+} on the exchange complex increases. If so, compacted clay would likely maintain the diffusivity of that initially emplaced in a disposal vault regardless of how the exchangeable cation composition evolves (Choi and Oscarson, 1996).

Dependence on the diffusant

Different species of the same element may have different diffusion coefficients. Christiansen and Torstenfelt (1988) found two diffusion coefficients for Ni, one probably as $\text{Ni}(\text{OH})_2$ (or possibly as NiOH^+) and the other as Ni^{2+} . Also Sr was found to migrate with two diffusion coefficients, one probably as SrCO_3 and the other as Sr^{2+} (Christiansen and Torstenfelt, 1988). This behaviour with two diffusion coefficients was not observed for strontium diffusion in a neutral aqueous phase, as in groundwater (Torstenfelt, 1986a). The reason for the much lower diffusion coefficient of part of Sr in the high pH was probably the formation of neutral SrCO_3 (Christiansen and Torstenfelt, 1988).

The apparent diffusion coefficients for I^- , Cl^- and TcO_4^- were found to be similar (Oscarson et al., 1992). The fact that the apparent diffusion coefficients for these monovalent anions are similar is not surprising given that they are not strongly sorbed on bentonite and that their effective hydrated radii are all essentially the same: 0.331 nm for I^- , 0.332 nm for Cl^- , and ~0.35 nm for TcO_4^- (Oscarson et al., 1992).

The diffusion coefficients of different cations deviate from each other (Muurinen and Lehtikoinen, 1995), although the effective hydrated radii of most common inorganic ionic species are similar - ranging from 0.33 to 0.45 nm (Cho et al., 1993). The probable explanation for this could be that only part of the sorbed cations are in the diffuse layer

and are mobile, while the rest of the sorbed cations are in the Stern layer and immobile (Muurinen and Lehtikoinen, 1995).

Redox-dependence of diffusion

Albinsson et al. (1990b) measured the apparent diffusion coefficients of U, Pu, Np, and Tc in compacted bentonite mixed with 1% Fe(0), Fe(II), Cu(0) or Cu(II) and for Np and Tc with 1 or 10% respectively of vivianite ($\text{Fe}_3(\text{PO}_4)_2$), magnetite or fracture fillings (mainly epidot and chlorite) in a nitrogen atmosphere. The results indicated, especially in the case of Fe(0) or Fe(II), reduction from the higher oxidation states Np(V), U(VI) and Tc(VII) to lower oxidation states probably Np(IV), U(IV) and Tc(IV). The apparent diffusion coefficients decreased by several orders of magnitude. For Tc with Fe(0) addition, no migration could be measured, while with Cu(I) addition, the effect was not so pronounced. Albinsson et al. (1990b) concluded that the material for a canister containing nuclear waste would, for Tc, U and Np, give a significant decrease of the transport through the bentonite barrier and with elementary iron in the bentonite barrier it would be sufficient to completely retain Tc in the bentonite buffer.

Interaction between the diffusing species and the clay matrix, possibly by hydrogen bonding, caused by OH-groups, was demonstrated in Tc diffusion experiments (Eriksen and Jacobsson, 1984). Under oxic conditions the dominating species was the heptavalent TcO_4^- having a high diffusion coefficient whereas on reduction of TcO_4^- the dominating soluble species would be $\text{TcO}(\text{OH})_2$. The diffusion coefficient of $\text{TcO}(\text{OH})_2$ was found to be at least two orders of magnitude lower than that of TcO_4^- (Eriksen and Jacobsson, 1984).

Experimental methods

In choosing experimental methods great care must be taken in defining the experimental constraints. When measuring the diffusive transport through a clay section one must consider the effect of two clay-solution interfaces. If the clay is confined by metallic filters, the diffusive resistance of these may quite often be the limiting factor (Eriksen and Jacobsson, 1984).

The methods used to determine apparent diffusion coefficients in compacted bentonite-water systems were assessed by Cheung (1989). The measured apparent diffusion coefficient could vary between the methods. The measured diffusion coefficients would

often vary by several orders of magnitude (Cheung, 1990). It was noted (Cheung, 1989) that the measured apparent diffusion coefficient under transient conditions was not numerically the same as that at steady-state conditions.

Three fundamentally different tracer methods have been frequently used in various studies to determine the apparent diffusion coefficient in compacted bentonite-water systems.

Method I

This method is a transient method, sometimes called in-diffusion method. In this method, air-dried, tracer-free bentonite is compacted in a stainless-steel ring prior to being placed in a cylindrical diffusion cell. The bentonite is then subjected to a selected water solution for saturation for a certain period. After this period of time, a solution with a tracer is circulated through a metallic filter, which is in contact with the bentonite specimen (Figure 1). The tracer concentration of solution is adjusted to keep constant during the diffusion experiment. After a predetermined diffusion period, the bentonite is removed from the diffusion cell, and sectioned into thin slices. The tracer concentration profile in the bentonite specimen is then determined.

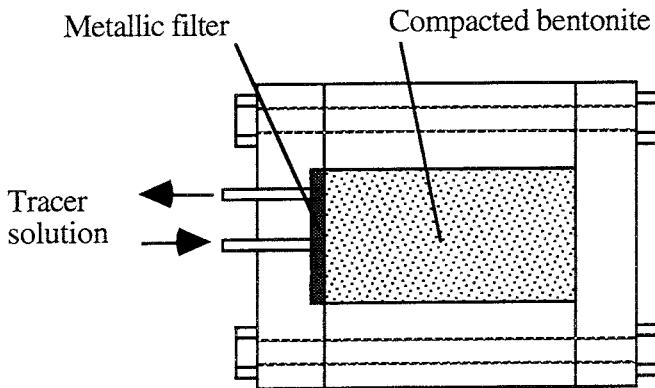


Figure 1. Diffusion cell for Method I.

The solution to the diffusion equation for the initial and boundary conditions in this method has been given (Crank, 1975), provided that the tracer does not penetrate through the bentonite specimen:

$$\frac{C}{C_0} = \operatorname{erfc} \frac{x}{2\sqrt{D_a t}} \quad (9)$$

where C is the tracer concentration at a distance x from the solution-bentonite interface at time t , C_0 is the concentration at the interface, and erfc denotes the complementary error function. C_0 is generally considered by using the measured average concentration in the bentonite slice adjacent to the tracer solution (Cheung, 1990)

In this method, the requirement of a constant tracer concentration at the solution-bentonite interface is probably not satisfied for cationic diffusion due to the high sorption capacity in the bentonite (Cheung, 1989) and the mass transfer resistance in the metallic filter. For the radionuclide with high sorption on bentonite, the resistance in the filter could result in erroneous measured parameters. This can be shown in the following calculations.

The boundary condition for the diffusion equation in bentonite in Method I is:

$$k_f(C_{wo} - C_{wb}) = -D_a \frac{\partial C}{\partial x} \Big|_{x=0} \quad (10)$$

where k_f is the mass transfer coefficient for the filter, C_{wo} and C_{wb} are the tracer concentrations in the bulk water solution and in the pore water solution at the filter-bentonite boundary respectively, and C is the total concentration in the bentonite. These concentrations are illustrated in Figure 2.

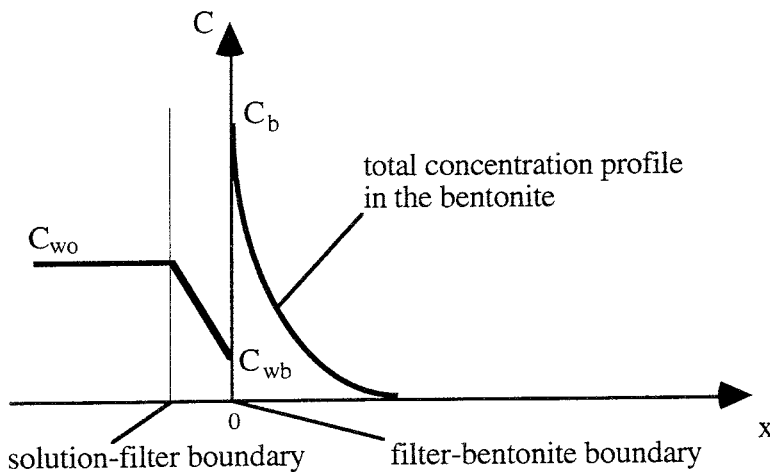


Figure 2. Illustration of the concentration profiles in diffusion experiments by Method I.

The mass transfer coefficient for the filter is related to the diffusion coefficient in the bulk solution, the porosity of the filter and the thickness of the filter approximately by the following equation:

$$k_f = \frac{D_w \varepsilon_f}{L_f} \quad (11)$$

where D_w is the diffusion coefficient in the bulk solution, ε_f the porosity of the filter and L_f the thickness of the filter. At equilibrium the total concentration in the bentonite, C , is related to the concentration in the solution phase by the distribution coefficient K_d :

$$C = K_d \rho C_w \quad (12)$$

where ρ is the density of the bentonite and C_w is the concentration in the solution phase.

With this boundary condition (Equation 10) and the initial condition $C = 0$ at $t = 0$, the diffusion equation (Equation 8) has an analytical solution (Carslaw and Jaeger, 1959):

$$\frac{C}{C_o} = 1 - \operatorname{erf}\left(\frac{x}{2\sqrt{D_a t}}\right) - \exp(hx + h^2 D_a t) \operatorname{erfc}\left(\frac{x}{2\sqrt{D_a t}} + h\sqrt{D_a t}\right) \quad (13)$$

where C_o is the concentration in equilibrium with C_{wo} according to Equation 12, i.e. $C_o = K_d \rho C_{wo}$, and h is defined here as

$$h = \frac{k_f}{K_d \rho D_a} \quad (14)$$

The following three cases demonstrate the deviations of this solution from the solution without filter resistance (Equation 9).

Case 1. The bentonite thickness is 0.1 m, $D_w = 3 \cdot 10^{-9}$ m²/s, $\varepsilon_f = 0.333$, $L_f = 0.002$ m, $K_d = 0.2$ m³/kg, $\rho = 2000$ kg/m³, $D_a = 1 \cdot 10^{-11}$ m²/s. The concentration profiles obtained with and without consideration to the filter resistance at $t = 365$ d are shown in Figure 3.

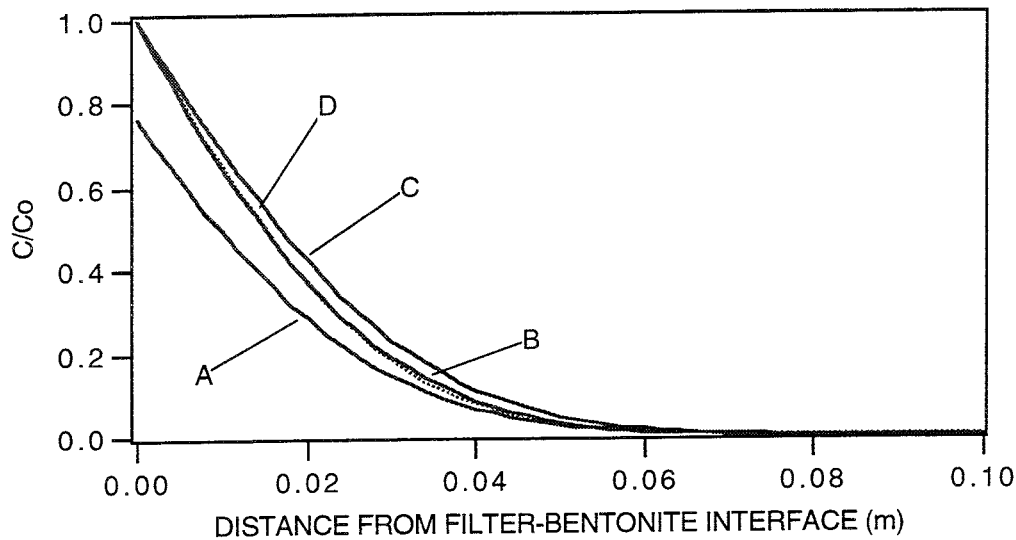


Figure 3. Concentration profiles in the bentonite at 365 d in Case 1. A: C/C_0 , profile accounting for filter resistance; B: C/C_b , profile accounting for filter resistance, normalized with C_b ; C: profile neglecting filter resistance; D: dotted line, profile neglecting filter resistance with $D_a = 8 \cdot 10^{-12} \text{ m}^2/\text{s}$.

From Figure 3, we can see that the concentration profile considering the filter resistance (Curve A) and the concentration profile disregarding the filter resistance (Curve C) are different. The concentration profile normalised by the filter-bentonite boundary concentration C_b (Curve B) still deviates from Curve C. If the apparent diffusion coefficient would be determined by fitting Equation 9 to Curve B, the obtained value would be about $8 \cdot 10^{-12} \text{ m}^2/\text{s}$ instead of $1 \cdot 10^{-11} \text{ m}^2/\text{s}$, a 20% deviation. If K_d would be determined by the measured C_b and C_{w0} , i.e., neglecting the filter resistance, the obtained value would be over 20% less than the real value.

Case 2. $K_d = 0.5 \text{ m}^3/\text{kg}$, the other parameters are the same as in Case 1. The concentration profiles obtained with and without consideration to the filter resistance at $t = 365 \text{ d}$ are shown in Figure 4.

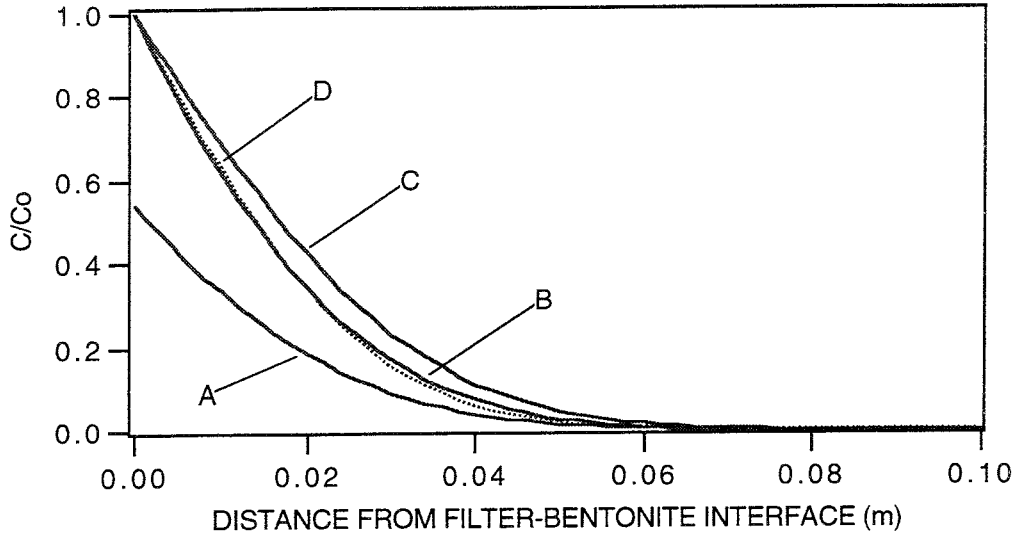


Figure 4. Concentration profiles in the bentonite at 365 d in Case 2. A: C/C_0 , profile accounting for filter resistance; B: C/C_b , profile accounting for filter resistance, normalized with C_b ; C: profile neglecting filter resistance; D: dotted line, profile neglecting filter resistance with $D_a = 7 \cdot 10^{-12} \text{ m}^2/\text{s}$.

From Figure 4, we can find more discrepancy between the concentration profile considering the filter resistance (Curve A) and the concentration profile disregarding the filter resistance (Curve C). The concentration profile normalised by the filter-bentonite boundary concentration C_b (Curve B) also deviates more from Curve C. If the apparent diffusion coefficient would be determined by fitting Equation 9 to Curve B, the obtained value would be about $7 \cdot 10^{-12} \text{ m}^2/\text{s}$ instead of $1 \cdot 10^{-11} \text{ m}^2/\text{s}$, a 30% deviation. If K_d would be determined by the measured C_b and C_{wo} , the obtained value would be over 40% less than the real value.

Case 3. The bentonite thickness is 0.02 m, $K_d = 1 \text{ m}^3/\text{kg}$, the other parameters are the same as in Case 1. The concentration profiles obtained with and without consideration to the filter resistance at $t = 40 \text{ d}$ are shown in Figure 5.

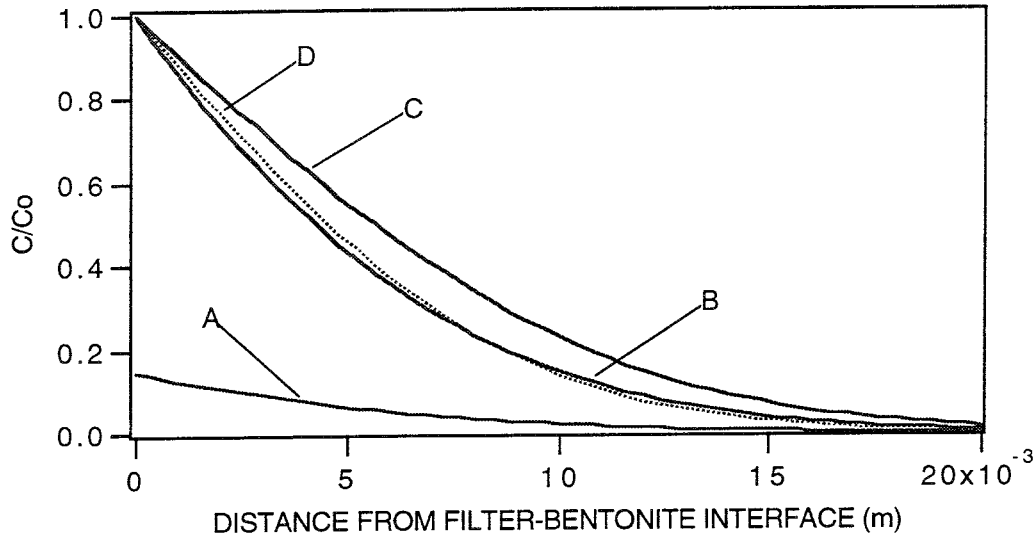


Figure 5. Concentration profiles in the bentonite at 40 d in Case 3. A: C/C_0 , profile accounting for filter resistance; B: C/C_b , profile accounting for filter resistance, normalized with C_b ; C: profile neglecting filter resistance; D: dotted line, profile neglecting filter resistance with $D_a = 6.5 \cdot 10^{-12} \text{ m}^2/\text{s}$.

From Figure 5, we can find that for shorter length of bentonite specimen and higher sorption effect, the deviation becomes larger. The apparent diffusion coefficient obtained from fitting Equation 9 to Curve B would be about $6.5 \cdot 10^{-12} \text{ m}^2/\text{s}$ instead of $1 \cdot 10^{-11} \text{ m}^2/\text{s}$, resulting in a 35% deviation. If K_d would be determined by the measured C_b and C_{w0} , the obtained value would be reduced by a factor of 5 due to the filter resistance.

From the above demonstration, it is advisable to try to avoid the error caused by the filter mass transfer resistance when using this method for apparent diffusion coefficient measurement, especially where the values of the sorption distribution coefficient and the apparent diffusion coefficient are high.

Method II

This method is also a transient method in which the tracer source is introduced to the tracer-free bentonite either as a thin layer or as an extended section of tracer-tagged bentonite. Thus the method is commonly divided into two methods.

Method IIa, impulse source

In this method a thin layer of tracer source is placed between two compacted bentonite sections (Figure 6). The bentonite is initially tracer-free and saturated with a selected water solution. After a predetermined diffusion period, the bentonite specimen is removed from diffusion cell and sectioned into slices. Subsequently the concentration profile in the bentonite is determined (Torstenfelt, 1986a).

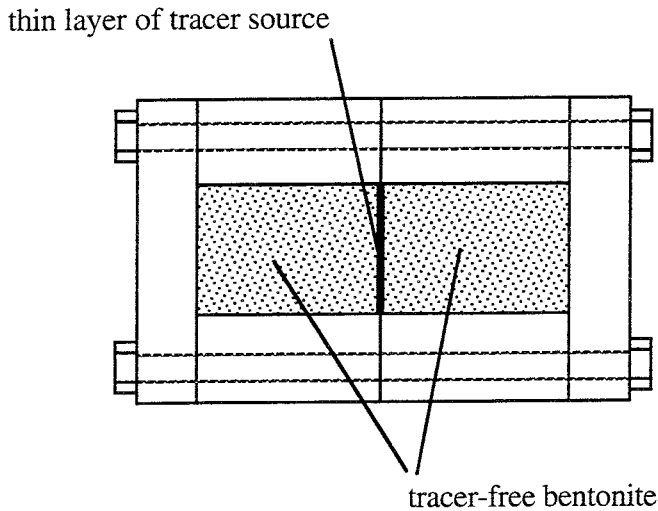


Figure 6. Diffusion cell for Method IIa

For the initial and boundary conditions in this method, the diffusion equation (Equation 8) has the following solution provided that the tracer does not penetrate the bentonite specimen (Crank, 1975, Torstenfelt, 1986a).

$$\frac{C}{M} = \frac{1}{2\sqrt{\pi D_a t}} \exp\left(\frac{-x^2}{4D_a t}\right) \quad (15)$$

where M is the total amount of tracer added to the source layer per unit area and x is the distance from the interface.

In this method, the filter mass transfer resistance in Method I is avoided. However, care must be taken with the placing angle and the shape of the clay plate initially containing all activity. If the plate is not placed at right angle to the axis of the measuring cell, the slices near the center will contain fractions of the initial radioactive plate and this will give a positive bias to the measured activity (Albinsson and Engkvist, 1989), which thus

questions the validity of the diffusion coefficient values obtained for the almost immobile radionuclides, especially plutonium.

Method IIb, extended source

In this method, two sections of bentonite are saturated with a selected water solution. The tracer is added to one of the bentonite sections. These two specimens are compacted to the same density and brought into contact with each other in the diffusion cell, as shown in Figure 7. After a predetermined diffusion period, the bentonite specimens are removed from the diffusion cell and sectioned into slices. Subsequently, the concentration profile in the bentonite is determined (Robin et al., 1987).

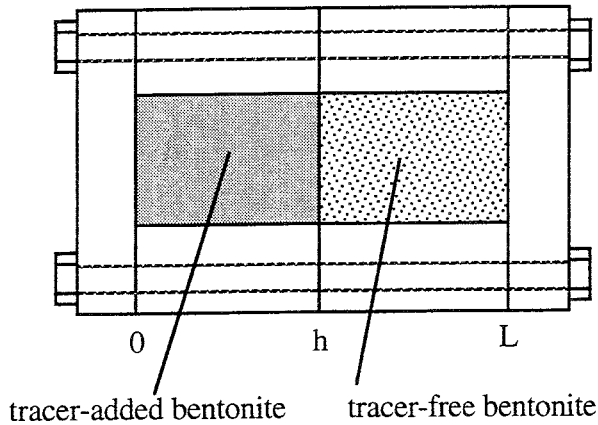


Figure 7. Diffusion cell for Method IIb.

For the initial and boundary conditions in this method, the diffusion equation (Equation 8) has the following solution (Robin et al., 1987). This is useful for the "long" times i.e. when the profile reaches the bottom of the cell.

$$\frac{C}{C_0} = \frac{h}{L} + \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{1}{\theta} \exp\left(-\frac{\theta^2 \pi^2 D_a t}{L^2}\right) \cos\left(\frac{\theta \pi x}{L}\right) \sin\left(\frac{\theta \pi h}{L}\right) \quad (16)$$

where C_0 is the initial tracer concentration in the tagged bentonite, h the thickness of the tagged bentonite, L the total length of the diffusion cell, θ the water content of the bentonite, and x the distance from the tracer end of the diffusion cell.

Equation 8 has also the solution (Crank, 1975) useful for "short" times:

$$\frac{C}{C_o} = \frac{1}{2} \operatorname{erfc}\left(\frac{x-h}{2\sqrt{D_a t}}\right) \quad (17)$$

Another solution is given in the literature (Muurinen et al., 1994)

$$\frac{C}{C_o} = \frac{1}{2} \left\{ \operatorname{erf}\left[2(h-x)\sqrt{D_a t}\right] + \operatorname{erf}\left[2(h+x)\sqrt{D_a t}\right] \right\} \quad (18)$$

This method also avoids the filter mass transfer resistance in Method I.

It is generally observed in diffusion experiments of this type that there is significantly more variability in the tracer concentration in the initially tagged clay section relative to the untagged clay section (Sawatsky and Oscarson, 1991b). This could be attributed to insufficient mixing preparation of the initially tagged clay section.

Method III

This method is often referred to as through-diffusion method or steady-state method, which means that the tracer will penetrate the bentonite specimen and the concentration profile and thereby the diffusion flux will reach a steady state. In this method, a water solution containing tracer with constant concentration is circulated through a filter which is in contact with one end of the bentonite specimen. Another water solution which is initially free of tracer is circulated through a filter in contact with the other end of the bentonite specimen, as shown in Figure 8. The bentonite is initially saturated with a selected water solution and compacted to a certain density before being placed in the diffusion cell. The transient tracer concentration in initially tracer-free solution is monitored. This provides a cumulative amount of tracer diffused through the bentonite specimen. After a predetermined diffusion period, the bentonite specimen is removed from the diffusion cell and sectioned into slices. Subsequently the concentration profile in the bentonite is determined (Eriksen and Jacobsson, 1984).

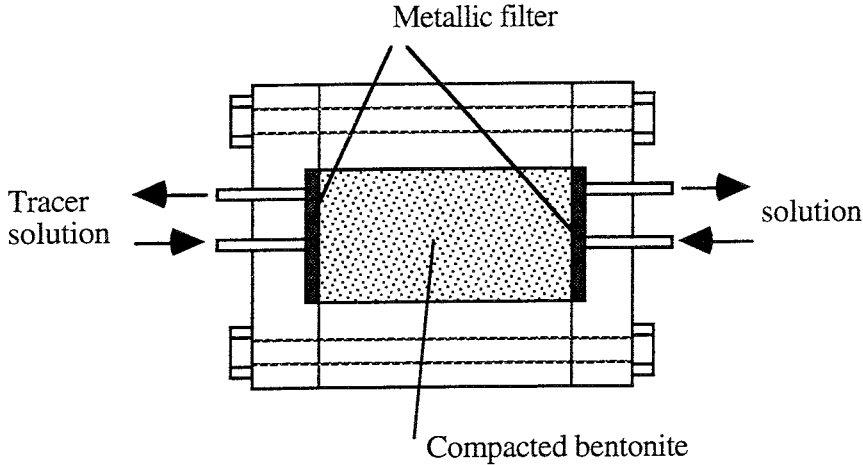


Figure 8. Diffusion cell for Method III.

When steady-state has been reached the apparent diffusion coefficient can be calculated by the equation (Eriksen and Jacobsson, 1984):

$$D_a = \frac{QL}{A(C_1 - C_o)} \quad (19)$$

where Q is the total amount of tracer diffused through the specimen per unit time, L the thickness of the bentonite specimen, A the surface area perpendicular to the diffusion flux, C_1 is the total tracer concentration in the bentonite at the interface to the tracer source solution and C_o is the total tracer concentration in the bentonite at the interface to the initially tracer-free solution.

Care should be taken in evaluating C_1 and C_o . If C_1 and C_o are calculated from the tracer concentrations in the bulk solutions circulating through the filters based on the sorption distribution coefficient, the values thus obtained would deviate from those directly obtained from the profile in the bentonite specimen because of the filter mass transfer resistance, as demonstrated in Method I, even though the deviation in the steady-state case would be less than that in the transient case. The D_a value thus obtained would be underestimated.

The concentration monitored in the initially tracer-free solution gives a breakthrough curve. When a steady state is reached, the breakthrough curve becomes linear. The intercept of the asymptote of the breakthrough curve at the time axis gives the time lag, which can also be used to calculate D_a (Eriksen and Jacobsson, 1984):

$$D_a = \frac{L^2}{6t_l} \quad (20)$$

where t_l is the time lag.

This method has also been used to calculate the effective diffusion coefficient by the equation (Eriksen and Jacobsson, 1984):

$$D_e = \frac{QL}{A(C_{w1} - C_{w0})} \quad (21)$$

where C_{w1} and C_{w0} are the tracer concentrations in the tracer source solution and in the initially tracer-free solution respectively.

Again, when using this equation, caution should be taken regarding the filter resistance.

Lehikoinen et al. (1996) pointed out that the interference caused by the filter plates appeared difficult but could be avoidable through the use of bentonite samples long enough to minimise the contribution of filter resistance to the overall diffusion resistance. The extension of the bentonite sample is, however, limited by the experimental time.

In the experiments of uranium diffusion in compacted bentonite (Muurinen, 1990), the steady state was clearly reached in the samples where the density was 600 to 900 kg/m³. With higher densities the steady state was not so obvious. The apparent diffusion coefficients obtained from the concentration profile were systematically higher by a factor of 5 to 10 than those obtained by the time-lag method. The apparent diffusion coefficients obtained from the concentration profile and obtained by time-lag were also found to vary by less than a factor of four in the diffusion measurement of carbon in compacted bentonite. This degree of variability is not uncommon in diffusion experiments of this type (Oscarson and Hume, 1994). Helfferich (1962) has noted that the time-lag technique is not particularly accurate.

Summary on how we view diffusion in compacted bentonite

Equations (1-8) are based on the assumption that there is a pore water concentration that can be defined. Because of the narrow pore spaces and the electrical double layer near the surfaces of the clay particles this is not a viable concept for charged diffusing species. Nevertheless the basic ideas can be applied on a macroscopic scale if the concentration C_p

in the equations instead is thought of as that in a free bulk water outside the clay that would be in local equilibrium with a point in the compacted clay, C_w . Such a concentration would e.g. result if equilibrium would be attained in the experiment in Figure 1. The effective diffusivity, D_e is now seen as that defined by a gradient in C_w . The total equilibrium concentration in the compacted bentonite is C which is related to C_w by $C=C_w \cdot (\varepsilon+K_d\rho)$. In this way the relation between the effective and apparent diffusivities, Equation (7) becomes well defined. $D_a = \frac{D_e}{\varepsilon + K_d\rho}$. As will be found and

described later in chapter 5, Discussion and Conclusions, the effective diffusivity D_e is the entity that is most invariant for the nuclides that do not have the surface diffusion or ion exclusion effects. For very high salinity waters this would apply to all nuclides because the electrical double layer then is much suppressed. The apparent diffusivity D_a on the other hand is commonly the most direct entity evaluated from the experiments when concentration profiles are observed. We therefore give recommended D_a values for the individual nuclides. These must be taken together with the recommended value for the sorption coefficient K_d . The D_a value thus is bound to one specific K_d value. The "invariant" D_e is used to obtain D_a values for other sorption coefficients by Equation (7). The effects of ion exclusion and surface diffusion are accounted for specifically. This is described in detail later together with the discussion of the recommended procedure and data. Somewhat simplified it may be stated that in high ionic strength waters these effects are not important and all species have similar effective diffusivities. In low ionic strength waters on the other hand ion exclusion and surface diffusion effects become quite important for some species.

For nuclides where no experimental diffusion data have been found the recommended data have been selected with some bias to the high side in order not to underestimate the rate of transport.

4 SORPTION EQUILIBRIUM DATA AND DIFFUSION COEFFICIENTS OF RADIONUCLIDES IN COMPACTED BENTONITE

In this section literature data on the different radionuclides are compiled in tables. The data for every nuclide is commented upon and recommended K_d , and D_a -values are given for all the nuclides. ρ_b and ρ_d indicate the compacted clay densities for bulk wet and bulk dry density respectively. Where there is no subscript it has not been possible to ascertain which of the cases is meant.

The recommended data for K_d are chosen from lower end of the observed span. This is done to ensure that the transport rate through the buffer is not underestimated.

We have found that the methods to determine D_a are more reliable than those used to determine D_e . The latter, if and when needed, are determined by Equation (7). Ion exclusion effects are seen in the decrease in apparent porosity e for the compacted clay.

As will be seen in the tables of diffusion data there are very large variations in data from different investigations and also, but somewhat less within one and the same investigation. Often it has not been possible to determine what the authors mean by dry compacted bentonite, if it means air dry then the powder has an unknown amount of adsorbed water already. An increased compaction will decrease the diffusivity but in the range of interest, 1600-2000kg/m³, the variations are so large that a value is chosen that is representative of the whole range.

Americium

Americium is expected to be present only in the Am(III) oxidation state in aerated solution and the most abundant complexes formed in ground water and pH 8.2 are $\text{Am}(\text{OH})_2^+$, $\text{Am}(\text{OH})_2^{2+}$, AmCO_3^+ , and $\text{Am}(\text{CO}_3)_2^-$ (Eriksen and Jacobsson, 1984). If, as expected, the pH in the clay is higher (~9.5) and the CO_3^{2-} concentration is lower than in the ground water $\text{Am}(\text{OH})_3$ will be the dominating species.

The speciation of Am was calculated for the Am-carbonate system at an ionic strength of 0.1 M and at a CO_2 partial pressure of $10^{-4.5}$ atm (Nagasaki et al., 1994). Am^{3+} is dominant at $\text{pH} < 7$. The monohydroxo complex AmOH^{2+} begins forming at $\text{pH} < 4.5$. The bis-hydroxo complex $\text{Am}(\text{OH})_2^+$ and the monocarbonate complex AmCO_3^+ begin to

form at pH = 6.5. The bis- and tris-carbonate complexes $\text{Am}(\text{CO}_3)_2^-$ and $\text{Am}(\text{CO}_3)_3^{3-}$ are dominant at pH > 9.

In the compilation of sorption data for bentonite made by Oscarson et al. (1984), K_d -values for Am in the system montmorillonite and groundwater were reported as 0.16 m^3/kg at pH 5 and pH 8, and 1.4 and 9.2 m^3/kg at pH 9.9 in Na-montmorillonite and Ca-montmorillonite, respectively.

Nagasaki et al. (1994) observed that, in the solution equilibrated with the sodium bentonite Kunigel V1, a great part of Am formed Am-bearing colloids, which suggested that the water-bentonite interaction could not be appropriately described by K_d alone.

Experimentally determined sorption and diffusion coefficient data of Am are given in Table 3.

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A realistic K_d -value of 3 m^3/kg is proposed and a conservative value of 1 m^3/kg is proposed, as proposed in SKB TR 91-16.

On the basis of the experimentally obtained diffusion data a D_a -value of $1 \cdot 10^{-14} \text{ m}^2/\text{s}$ is proposed for Am in compacted bentonite.

Table 3. Experimentally determined diffusion coefficient and sorption data for americium from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Wyoming MX-80 Na-bentonite, SGW				6.2 - 6.6	batch sorption	Allard et al., 1982
Na-bentonite pH 13.3	ρ _d 1100	2.7·10 ⁻¹⁰			IIa	Christiansen and Torstenfelt, 1988
bentonite, DW, pH 2.0				6.8	batch sorption	Konishi et al., 1988
bentonite, DW, pH 6.0				23	- " -	- " -
bentonite, SGW, pH 2.0				3.8	- " -	- " -
Na-bentonite DW	ρ _d 200	7.2·10 ⁻¹⁴			IIa	Sato et al., 1992
- " -	ρ _d 600	1.9·10 ⁻¹⁴			- " -	- " -
- " -	ρ _d 1550	9.0·10 ⁻¹⁶			- " -	- " -
Wyoming MX-80 Na-bentonite, SGW	ρ _d 1900 - 2000	4·10 ⁻¹⁵ 1.4·10 ⁻¹⁴			transient diff.	Torstenfelt et al., 1982
Wyoming MX-80 Na-bentonite, SGW	ρ _d 1900 - 2300	2·10 ⁻¹⁵ 7.3·10 ⁻¹⁵			- " -	Torstenfelt et al., 1983
Wyoming MX-80 Na-bentonite, SGW 600 ppm HCO ₃ ⁻	- " -	2.8·10 ⁻¹⁵ 9.5·10 ⁻¹⁵			- " -	- " -
Wyoming MX-80 Na-bentonite, SGW	ρ _d 2000	1.3·10 ⁻¹⁴			- " -	Torstenfelt and Allard, 1986
Wyoming MX-80 Na-bentonite, SGW 600 ppm HCO ₃ ⁻	- " -	2.8·10 ⁻¹⁶			- " -	- " -

Carbon

Dissolved inorganic carbon occurs in groundwater in the carbon-bicarbonate-carbonate system. The speciation of carbon is related to the pH (Hakanen and Hölttä, 1992).

A K_d -value of 0 for inorganic carbon (HCO_3^-) in Na-montmorillonite was given by Allard et al. (1981). The effective diffusion coefficient of dissolved CH_4 in bentonite saturated with synthetic groundwater was reported by Neretnieks and Skagius (1978). The bentonite was compacted to a density of 2350 kg/m^3 , and the measurement was performed at 50°C . The effective diffusion coefficient, D_e , was determined to be $3.9 \cdot 10^{-11} \text{ m}^2/\text{s}$.

Oscarson and Hume (1994) indicated that to ensure a slow migration of ^{14}C through a clay-based barrier material, the barrier must remain saturated. If the barrier becomes unsaturated and interconnected air-filled pores are formed, the transport rate of ^{14}C would be expected to increase markedly because of the migration of $^{14}\text{CO}_2$ in the gas phase - gaseous diffusion is generally about four orders of magnitude greater than that in aqueous solution (Oscarson and Hume, 1994).

Experimentally determined diffusion coefficient data of carbon are given in Table 4.

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A K_d -value of 0 is proposed, as proposed in SKB TR 91-16.

On the basis of the experimentally obtained diffusion data, a D_a -value of $5 \cdot 10^{-12} \text{ m}^2/\text{s}$ is proposed for C in compacted bentonite.

Table 4. Experimentally determined diffusion coefficient data for carbon from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Avonlea Na-bentonite, SGW	ρ _d 960	3.8·10 ⁻¹¹ (1.7·10 ⁻¹¹ time-lag)	1.1·10 ⁻¹¹		III	Oscarson and Hume, 1994
— " —	ρ _d 890	4.2·10 ⁻¹¹ (2.1·10 ⁻¹¹ time-lag)	9.6·10 ⁻¹²		— " —	— " —
— " —	ρ _d 920	6.5·10 ⁻¹¹ (1.9·10 ⁻¹¹ time-lag)	1·10 ⁻¹¹		— " —	— " —
— " —	ρ _d 1180	7.1·10 ⁻¹² (1.7·10 ⁻¹¹ time-lag)	3.0·10 ⁻¹²		— " —	— " —
— " —	ρ _d 1180	1.2·10 ⁻¹¹ (1.7·10 ⁻¹¹ time-lag)	8.0·10 ⁻¹²		— " —	— " —
— " —	ρ _d 1250	7.2·10 ⁻¹² (5.4·10 ⁻¹² time-lag)	1.3·10 ⁻¹²		— " —	— " —
— " —	ρ _d 1310	6.6·10 ⁻¹² (5.7·10 ⁻¹² time-lag)	4.7·10 ⁻¹²		— " —	— " —
— " —	ρ _d 1430	2.6·10 ⁻¹² (8.2·10 ⁻¹² time-lag)	5.5·10 ⁻¹³		— " —	— " —
— " —	ρ _d 1410	2.4·10 ⁻¹² (5.5·10 ⁻¹² time-lag)	2.7·10 ⁻¹³		— " —	— " —
— " —	ρ _d 1610	1.3·10 ⁻¹² (5.9·10 ⁻¹² time-lag)	4.9·10 ⁻¹³		— " —	— " —
— " —	ρ _d 1630	2.2·10 ⁻¹² (5.1·10 ⁻¹² time-lag)	6.5·10 ⁻¹³		— " —	— " —

Cerium

Cerium will probably be hydrolysed in the bentonite porewater and thus strongly sorbed onto the material (Brandberg and Skagius, 1991).

Sorption data for cerium in bentonite were given in a compilation of sorption data made by Oscarson et al. (1984). K_d -values reported ranged from 0.3 to 2.0 m³/kg in bentonite saturated with a solution containing sodium, calcium, chloride, and carbonate with an ionic strength of 0.0055 M and pH 8.5. No diffusion coefficient data have been found.

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). Ce is a lanthanide and can be expected to behave similarly to Am. A realistic K_d -value of 1 m³/kg is proposed and a conservative value of 0.2 m³/kg is proposed, as proposed in SKB TR 91-16.

A D_a -value of $1 \cdot 10^{-13}$ m²/s is proposed for Ce in compacted bentonite.

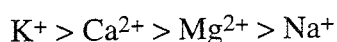
Cesium

Cesium is largely present as unhydrated Cs⁺ ion under most groundwater conditions (Cho et al., 1993). Sorption and diffusion behaviour of Cs in bentonite have been extensively studied.

Hsu and Chang (1994) conducted sorption and desorption experiments for Cs on Ca-saturated Wyoming Bentonite. The water in contact with the bentonite was initially deionized water. Cs was rapidly sorbed by the bentonite, reaching equilibrium within 24 h. The K_d obtained in the sorption experiment was 2.97 m³/kg, while the K_d obtained in the desorption experiment was 19.4 m³/kg, indicating that Cs has, to a large extent, been sorbed to the interlayer sites of the bentonite in an irreversible manner (Hsu and Chang, 1994). Nikiforov et al. (1991) concluded that the main factor determining the fixation of cesium cations was the low hydration energy.

Sorption of Cs on bentonite was studied by Khan et al. by using batch technique (1994). It was found that K_d varied with the Cs concentration, the concentration of other cations, and the pH value in the solution as well as the solution to bentonite ratios. Sorption data were well interpreted in terms of Freundlich, Langmuir and Dubinin-Radushkevich isotherms. The mean free energy (~14.5 kJ/mol) for cesium sorption showed that ion-exchange is the predominant mode of sorption at 0.01-0.5 molar cesium concentration.

The desorption studies with 0.01 M CaCl₂ and ground water at low cesium loading on bentonite showed that about 95% of the element is irreversibly sorbed probably due to the fixation of the cesium by isomorphous replacement in the crystal lattice of the bentonite. The results of Khan et al. (1994) showed that the rate of sorption of cesium on bentonite was rapid and equilibrium was reached instantly after mixing. The instantaneous uptake of cesium by the bentonite also indicated that ion exchange may be the main sorption mechanism (Khan et al., 1994). The presence of other cations was found to be able to depress the sorption of cesium in the following order:



Muurinen et al. (1985, 1987) measured K_d -values and apparent diffusion coefficients in Wyoming MX-80 bentonite contacted with solutions of different salinity. The K_d -values obtained decreased with increasing sodium concentration in the solution. This was attributed to that the sorption mechanism for cesium under the experimental condition was mainly ion exchange.

Kawaguchi et al. (1989) measured K_d -values for Cs in Na-bentonite contacted with solutions containing sodium concentrations from $1.45 \cdot 10^{-3}$ M to 0.446 M. The K_d -values decreased from 2.12 to 0.007 m³/kg with the increase in sodium concentration. They also measured sorption of Cs in Na-bentonite contacted with distilled water and obtained K_d -values between 1.72 and 2.12 m³/kg.

Liang et al. (1993) measured the sorption of Cs on Wyoming MX-90 bentonite. A modified Freundlich isotherm was used to describe the sorption equilibrium. It was observed that the exponent in the Freundlich isotherm was near unity when the initial radionuclide concentration was <0.01 M, suggesting a linear isotherm.

Oscarson et al. (1994) measured the sorption of cesium on compacted Na-bentonite. Over the clay density range from 500 to 1500 kg/m³, K_d values for Cs⁺ with compacted bentonite were about one-half to one-third the value of those with loose bentonite. The lower sorption on compacted bentonite was largely attributed to that Cs⁺ could not enter the small and occluded pores and thus it could not access the entire volume, or all the sorption sites, of the compacted bentonite. Oscarson et al. (1994) concluded that differences in solution composition was not the main reason for lower sorption in compacted bentonite.

Miyahara et al. (1991) measured the apparent diffusion coefficient of Cs in Funipia F bentonite by using Method IIa. D_a decreased with increasing dry density in the range of 200 to 2000 kg/m³.

Low surface diffusivity of cesium was observed. An explanation was that a significant fraction of the Cs⁺ ions was irreversibly sorbed in the Stern layer and immobile (Lehikoinen et al., 1996).

Recently Eriksen and Jansson (1996) measured the apparent diffusivity and sorption of Cesium in compacted clay at different ionic strengths and cesium concentrations. They used the through diffusion method. Their experimental technique permitted them to separately evaluate the surface diffusion coefficient and the pore diffusion coefficient. They found that the lower the ionic strength was, the higher was the sorption and also the transport in the sorbed phase. In summary the apparent diffusion coefficient D_a in low ionic strength water was $1.0 \cdot 10^{-12}$ m²/s and $2.3 \cdot 10^{-12}$ m²/s in saline water respectively. The effective diffusivities D_e were $6.4 \cdot 10^{-10}$ m²/s and $3.5 \cdot 10^{-10}$ m²/s in low ionic strength and saline water respectively.

Experimentally determined sorption and diffusion coefficient data of Cs are given in Table 5.

Many new sorption data have been reported since SKB TR 91-16 (Brandberg and Skagius, 1991). However, the K_d -values reported vary quite much from investigation to investigation. A realistic K_d -value of 0.5 m³/kg is proposed and a conservative value of 0.2 m³/kg is proposed for sorption in fresh groundwater. A realistic K_d -value of 0.05 m³/kg is proposed and a conservative value of 0.005 m³/kg is proposed for sorption in saline water, as proposed in SKB TR 91-16.

On the basis of the experimentally obtained diffusion data, a D_a -value of $5 \cdot 10^{-12}$ m²/s is proposed for Cs in compacted bentonite.

Chlorine

Chlorine occurs in groundwater as chloride ion Cl⁻. In many waters it is the most abundant anion.

Muurinen et al. (1989) performed measurements of the sorption and diffusion of chloride in Wyoming MX-80 bentonite in contact with a synthetic groundwater. No sorption of chloride was measured in the experiments.

Experimentally determined diffusion coefficient data of chlorine are given in Table 6.

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A K_d -value of 0 is proposed, as proposed in SKB TR 91-16.

On the basis of the experimentally obtained diffusion data, a D_a -value of $2 \cdot 10^{-11}$ m²/s is proposed for Cl in compacted bentonite. An effective porosity of 0.05 is proposed accounting for ion exclusion effects.

Table 5. Experimentally determined diffusion coefficient and sorption data for Cs from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite	ρ _d 2000	3·10 ⁻¹²			IIa	Albinsson et al., 1993
– " –	– " –	5·10 ⁻¹³			IIa	Albinsson et al., 1990a
Na-bentonite + 0.5% Cu	– " –	5·10 ⁻¹³			– " –	– " –
Na-bentonite + 0.5% Fe	– " –	5·10 ⁻¹³			– " –	– " –
Na-bentonite + 1% vivianite	– " –	1·10 ⁻¹²			– " –	– " –
Na-bentonite SGW	ρ _d 2000	3·10 ⁻¹²			IIb	Albinsson et al., 1996
Avonlea Na-bentonite	1250	2·10 ⁻¹¹			I	Cheung et al., 1986
– " –	1500	1.5·10 ⁻¹¹			– " –	– " –
– " –	1750	3·10 ⁻¹¹			– " –	– " –
Avonlea Na-bentonite low- and high-salinity water	ρ _d 1250	1·10 ⁻¹¹			III	Cheung, 1989
Avonlea Na-bentonite low-salinity water	ρ _d 1750	1·10 ⁻¹⁰			– " –	– " –
Avonlea Na-bentonite SGW	ρ _b 1360	5.4·10 ⁻¹³	1.3·10 ⁻¹¹	0.1	III batch sorption	Cho et al., 1993
Ca-bentonite	ρ _d 1520	6.7·10 ⁻¹³		8.998	IIa batch sorption	Choi et al., 1992
– " –	ρ _d 1780	1.7·10 ⁻¹³		10.538	– " –	– " –
Ca-bentonite heat treated at 100 °C	ρ _d 1590	8.5·10 ⁻¹³		12.487	– " –	– " –
Ca-bentonite heat treated at 150 °C	ρ _d 1670	6.0·10 ⁻¹³		17.116	– " –	– " –
Na-bentonite pH 13.3	ρ _d 1100	1.3·10 ⁻¹¹			IIa	Christiansen and Torstenfelt, 1988

Table 5. Experimentally determined diffusion coefficient and sorption data for Cs from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite 25 °C				1.37 - 1.46	batch sorption	Eriksen et al., 1981
Na-bentonite 25 °C	ρ _b 2100	7.5·10 ⁻¹²			transient diff.	- " -
Na-bentonite 70 °C	- " -	4.0·10 ⁻¹¹			- " -	- " -
Ca-bentonite 25 °C	- " -	3.4·10 ⁻¹²			- " -	- " -
Ca-bentonite 70 °C	- " -	4.0·10 ⁻¹²			- " -	- " -
Na-bentonite	ρ _b 2100	2·10 ⁻¹² - 2.8·10 ⁻¹²			IIb	Eriksen and Jacobsson, 1981
- " -	- " -	1.5·10 ⁻¹²			IIa	- " -
Ca-bentonite	ρ _b 1600	6·10 ⁻¹³ - 7.5·10 ⁻¹³			- " -	- " -
- " -	ρ _b 2100	6·10 ⁻¹³			- " -	- " -
Na-bentonite MX-80 Ionic Str. 0.018	ρ _d 1800	1.0·10 ⁻¹²	6.4·10 ⁻¹⁰	0.36	III	Eriksen & Jansson 1996
Na-bentonite MX-80 Ionic Str. 0.218	ρ _d 1800	2.3·10 ⁻¹²	3.5·10 ⁻¹⁰	0.085	- " -	- " -
montmorillonite GW				3.5	batch sorption	Erten et al., 1988
bentonite, water 0.001M Na, 0.002 M Ca, 0.001 M Mg				2.1-65 (med. 15)	batch sorption	Horyna and Dlouhy, 1988
Ca-saturated Wyoming bentonite				2.97 sorption	batch sorption and desorption	Hsu and Chang, 1994
- " -				19.4 desorp.	- " -	- " -
Na-bentonite 0 mmol/l Na				1.72-2.12	batch sorption	Kawaguchi et al., 1989
Na-bentonite 1.45 mmol/l Na pH 8.6				2.12	- " -	- " -
Na-bentonite 1.54 mmol/l Na pH 8.6				1.72	- " -	- " -

Table 5. Experimentally determined diffusion coefficient and sorption data for Cs from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite 2.01 mmol/l Na pH 9.3				1.94	batch sorption	Kawaguchi et al., 1989
Na-bentonite 3.36 mmol/l Na pH 9.0				1.56	– " –	– " –
Na-bentonite 5.11 mmol/l Na pH 8.8				0.83	– " –	– " –
Na-bentonite 13.6 mmol/l Na pH 8.7				0.54	– " –	– " –
Na-bentonite 44.4 mmol/l Na pH 8.8				0.26	– " –	– " –
Na-bentonite 152 mmol/l Na pH 9.5				0.14	– " –	– " –
Na-bentonite 439 mmol/l Na pH 9.3				0.1	– " –	– " –
Na-bentonite 466 mmol/l Na pH 9.2				0.07	– " –	– " –
Na-bentonite	1000	1.62·10 ⁻¹¹			transient diff.	– " –
– " –	– " –	1.33·10 ⁻¹¹			– " –	– " –
– " –	1200	1.45·10 ⁻¹¹			– " –	– " –
– " –	1400	1.18·10 ⁻¹¹			– " –	– " –
– " –	1850	6.5·10 ⁻¹²			– " –	– " –
– " –	– " –	6.6·10 ⁻¹²			– " –	– " –
Wyoming Na- bentonite, DW				0.662	batch sorption	Kim et al., 1993
– " –	ρ _d 1060	1·10 ⁻¹¹ 9.13·10 ⁻¹²			IIa	– " –
– " –	ρ _d 1270	7.15·10 ⁻¹² 6.87·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1580	2.57·10 ⁻¹² 3.24·10 ⁻¹²			– " –	– " –

Table 5. Experimentally determined diffusion coefficient and sorption data for Cs from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Wyoming Na-bentonite, DW	ρ _d 1780	3.5·10 ⁻¹³ 4.7·10 ⁻¹³			IIa	Kim et al., 1993
– " –	ρ _d 1960	1.13·10 ⁻¹³			– " –	– " –
bentonite, DW pH 2.4				0.14	batch sorption	Konishi et al., 1988
bentonite, DW pH 9.3				1.5	– " –	– " –
bentonite, SGW pH 2.4				0.16	– " –	– " –
bentonite, SGW pH 9.3				1.1	– " –	– " –
Na-bentonite ionic strength 10 ⁻³ M, pH 8.5				3.4	batch sorption	Miyahara et al., 1991
Kunipia F Na-bentonite	ρ _d 200	1.5·10 ⁻¹¹			IIa	– " –
– " –	ρ _d 400	1.1·10 ⁻¹¹			– " –	– " –
– " –	ρ _d 600	7.9·10 ⁻¹²			– " –	– " –
– " –	ρ _d 800	5.9·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1000	4.4·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1200	3.7·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1400	2.2·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1600	1.4·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1800	7.9·10 ⁻¹³			– " –	– " –
– " –	ρ _d 2000	3.9·10 ⁻¹³			– " –	– " –
Na-bentonite 1.2 M NaCl aerobic	ρ _d 2030 diff.exp	6.5·10 ⁻¹³		0.05	transient diff. batch sorption	Muurinen et al., 1985
Na-bentonite 0.6 M NaCl aerobic	ρ _d 1980 diff.exp	6.9·10 ⁻¹³		0.09	– " –	– " –

Table 5. Experimentally determined diffusion coefficient and sorption data for Cs from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite 0.2 M NaCl aerobic	ρ _d 1870 diff.exp	8.5·10 ⁻¹³		0.21	transient diff. batch sorption	Muurinen et al., 1985
Na-bentonite 0.02 M NaCl aerobic	ρ _d 1910 diff.exp	3.5·10 ⁻¹³		0.85	– " –	– " –
Na-bentonite 0.003 M NaCl aerobic	ρ _d 1940 diff.exp	2.5·10 ⁻¹³		1.8	– " –	– " –
Na-bentonite gw salt 10 g/l aerobic	ρ _d 2000 diff.exp	8·10 ⁻¹³		0.393	– " –	– " –
Na-bentonite 0.001 M NaCl				2.66	batch sorption	Muurinen et al., 1987
Na-bentonite 0.01 M NaCl				1.025	– " –	– " –
Na-bentonite 0.1 M NaCl				0.334	– " –	– " –
Na-bentonite 0.5 M NaCl				0.11	– " –	– " –
Na-bentonite 1 M NaCl				0.073	– " –	– " –
Na-bentonite 0.01 M NaCl	ρ _d 1750			4.173	sorption on compacted bentonite	– " –
Na-bentonite 0.1 M NaCl	– " –	4·10 ⁻¹² (1·10 ⁻¹² time-lag)	4.5·10 ⁻⁹	0.834	III sorption on compacted bentonite	– " –
Na-bentonite 0.5 M NaCl	– " –	4.5·10 ⁻¹² (1.1·10 ⁻¹² time-lag)	1.5·10 ⁻⁹	0.198 - 0.33	– " –	– " –
bentonite, SGW				0.34	batch sorption	Neretnieks and Skagius, 1978
bentonite, SGW	ρ _b 2350	5.4·10 ⁻¹¹				– " –

Table 5. Experimentally determined diffusion coefficient and sorption data for Cs from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-Ca-montmorillonite SGW 55400 ppm salt 25 °C				0.021 (init. Cs 10 ⁻⁵ M)	batch sorption	Oscarson et al., 1987
- " -				0.0356 (init. Cs 10 ⁻⁶ M)	- " -	- " -
- " -				0.0605 (init. Cs 10 ⁻⁷ M)	- " -	- " -
- " -				0.103 (init. Cs 10 ⁻⁸ M)	- " -	- " -
Na-Ca-montmorillonite SGW 55400 ppm salt 80 °C				0.0098 (init. Cs 10 ⁻⁵ M)	- " -	- " -
- " -				0.0167 (init. Cs 10 ⁻⁶ M)	- " -	- " -
- " -				0.0284 (init. Cs 10 ⁻⁷ M)	- " -	- " -
- " -				0.0482 (init. Cs 10 ⁻⁸ M)	- " -	- " -
Na-Ca-montmorillonite SGW 11700 ppm salt 25 °C				0.0481 (init. Cs 10 ⁻⁵ M)	- " -	- " -
- " -				0.078 (init. Cs 10 ⁻⁶ M)	- " -	- " -
- " -				0.127 (init. Cs 10 ⁻⁷ M)	- " -	- " -

Table 5. Experimentally determined diffusion coefficient and sorption data for Cs from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-Ca-montmorillonite SGW 11700 ppm salt 25 °C				0.205 (init. Cs 10 ⁻⁸ M)	batch sorption	Oscarson et al., 1987
Na-Ca-montmorillonite SGW 11700 ppm salt 80 °C				0.0131 (init. Cs 10 ⁻⁵ M)	- " -	- " -
- " -				0.0218 (init. Cs 10 ⁻⁶ M)	- " -	- " -
- " -				0.0362 (init. Cs 10 ⁻⁷ M)	- " -	- " -
- " -				0.060 (init. Cs 10 ⁻⁸ M)	- " -	- " -
Avonlea Na-bentonite, SGW, L/S 0.385 m ³ /kg	ρ _d 500			1.04 0.92	sorption on compacted bentonite	Oscarson et al., 1994
Avonlea Na-bentonite, SGW, L/S 0.274 m ³ /kg	ρ _d 700			0.79 0.81	- " -	- " -
Avonlea Na-bentonite, SGW, L/S 0.0822 m ³ /kg	ρ _d 900			0.52 0.52 0.52	- " -	- " -
Avonlea Na-bentonite, SGW, L/S 0.0595 m ³ /kg	ρ _d 1250			0.49 0.49 0.44	- " -	- " -
Avonlea Na-bentonite, SGW, L/S 0.0495 m ³ /kg	ρ _d 1500			0.45 0.42 0.43	- " -	- " -

Table 5. Experimentally determined diffusion coefficient and sorption data for Cs from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Avonlea Na-bentonite, SGW, L/S 0.405 m ³ /kg				1.88 1.8	batch sorption	Oscarson et al., 1994
Avonlea Na-bentonite, SGW, L/S 0.289 m ³ /kg				1.71 1.69	-- --	-- --
Avonlea Na-bentonite, SGW, L/S 0.0891 m ³ /kg				1.38 1.41 1.38	-- --	-- --
Avonlea Na-bentonite, SGW, L/S 0.0600 m ³ /kg				1.28 1.26 1.32	-- --	-- --
Avonlea Na-bentonite, SGW, L/S 0.0537 m ³ /kg				1.19 1.2 1.22	-- --	-- --
Na-bentonite DW	ρ _d 200	1.6·10 ⁻¹¹			IIa	Sato et al., 1992
-- --	ρ _d 400	1.2·10 ⁻¹¹			-- --	-- --
-- --	ρ _d 600	8.1·10 ⁻¹²			-- --	-- --
-- --	ρ _d 800	6.6·10 ⁻¹²			-- --	-- --
-- --	ρ _d 1000	4.8·10 ⁻¹²			-- --	-- --
-- --	ρ _d 1200	3.2·10 ⁻¹²			-- --	-- --
-- --	ρ _d 1400	2.2·10 ⁻¹²			-- --	-- --
-- --	ρ _d 1600	1.3·10 ⁻¹²			-- --	-- --
-- --	ρ _d 1800	7.9·10 ⁻¹³			-- --	-- --
-- --	ρ _d 2000	4.0·10 ⁻¹³			-- --	-- --
Na-bentonite, SGW 1700 ppm salt	2000 ~ 10 % water	2.4·10 ⁻¹²		1.4	transient diff. batch sorption	Torstenfelt, 1986a

Table 6. Experimentally determined diffusion coefficient data for chlorine from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Ca-bentonite	ρ _d 1570	9.5·10 ⁻¹¹			IIa	Choi et al., 1992
Na-bentonite	ρ _b 2100	6·10 ⁻¹²			transient diff.	Eriksen and Jacobsson, 1981
– " –	– " –		3.1·10 ⁻¹³		III	– " –
– " –	– " –	6·10 ⁻¹²			IIb	Eriksen, 1982
– " –	– " –	9.2·10 ⁻¹¹			III time-lag	– " –
– " –	– " –		3·10 ⁻¹³		III	– " –
Ca-bentonite 25 °C	– " –	1·10 ⁻¹²			transient diff.	Eriksen et al., 1981
bentonite 2 g/l cation	400	1·10 ⁻⁹			IIb	Gillham et al., 1984
Wyoming Na-bentonite, DW	ρ _d 1160	2.09·10 ⁻¹⁰ 1.82·10 ⁻¹⁰			IIa	Kim et al., 1993
– " –	ρ _d 1360	1.38·10 ⁻¹⁰ 1.29·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1580	7.57·10 ⁻¹¹ 7.58·10 ⁻¹¹			– " –	– " –
– " –	ρ _d 1820	6.21·10 ⁻¹² 5.71·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1900	5.68·10 ⁻¹² 4.95·10 ⁻¹²			– " –	– " –
Na-bentonite 0.01 M NaCl	1200	7·10 ⁻¹¹ (1.4·10 ⁻¹⁰ time-lag)	7.7·10 ⁻¹²		III	Muurinen et al., 1989
Na-bentonite 0.1 M NaCl	– " –	2.8·10 ⁻¹⁰ (2·10 ⁻¹⁰ time-lag)	2.9·10 ⁻¹¹		– " –	– " –
Na-bentonite 1 M NaCl	– " –	5.1·10 ⁻¹⁰ (3.2·10 ⁻¹⁰ time-lag)	1.2·10 ⁻¹⁰		– " –	– " –
Na-bentonite 0.01 M NaCl	1800	2·10 ⁻¹¹ (5·10 ⁻¹¹ time-lag)	3.3·10 ⁻¹³		– " –	– " –

Table 6. Experimentally determined diffusion coefficient data for chlorine from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite 0.1 M NaCl	1800	3.1·10 ⁻¹¹ (5.4·10 ⁻¹¹ time-lag)	4.8·10 ⁻¹³		III	Muurinen et al., 1989
Na-bentonite 1 M NaCl	– " –	5.2·10 ⁻¹¹ (7.7·10 ⁻¹¹ time-lag)	4·10 ⁻¹²		– " –	– " –
Na-bentonite SGW 1056 ppm salt 600 ppm HCO ₃ ⁻	ρ _d 1200	2·10 ⁻¹⁰ (1.5·10 ⁻¹⁰ time-lag)	1.1·10 ⁻¹¹		– " –	– " –
– " –	ρ _d 1500	9.4·10 ⁻¹¹ (7·10 ⁻¹¹ time-lag)	2.8·10 ⁻¹²		– " –	– " –
– " –	ρ _d 1800	5.1·10 ⁻¹¹ (5.2·10 ⁻¹¹ time-lag)	2·10 ⁻¹³		– " –	– " –
– " –	ρ _d 2100	3·10 ⁻¹¹	8.5·10 ⁻¹⁴		– " –	– " –
0.75/0.25 basalt/bentoniteSG W	ρ _b 1640	3.22·10 ⁻¹⁰			IIb	Relyea et al., 1986
– " –	ρ _b 1590	3.25·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1680	2.86·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1810	2.36·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1630	3.32·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1810	1.14·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1860	3.70·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1630	5.14·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1630	5.99·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1630	5.99·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1710	5.14·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1750	3.95·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1810	2.89·10 ⁻¹⁰			– " –	– " –

Table 6. Experimentally determined diffusion coefficient data for chlorine from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
0.75/0.25 basalt/bentoniteSG W	ρ _b 1570	8.80·10 ⁻¹⁰			IIb	Relyea et al., 1986
– " –	ρ _b 1570	8.17·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1590	9.01·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1770	3.71·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1810	7.35·10 ⁻¹⁰			– " –	– " –
– " –	ρ _b 1900	3.07·10 ⁻¹⁰			– " –	– " –
90 wt% tonalite + 10 wt% Na-bentonite GW		1.4·10 ⁻¹⁰ 1.8·10 ⁻¹⁰	6.9·10 ⁻¹³ 3.7·10 ⁻¹³		III	Uusheimo et al., 1993
60 wt% granite + 40 wt% Na-bentonite GW		1.6·10 ⁻¹⁰ 2.6·10 ⁻¹⁰	1.6·10 ⁻¹¹ 1.3·10 ⁻¹¹		– " –	– " –

Iodine

Iodine has the two valence states -1 and +5 in groundwater, with the corresponding chemical forms I^- and IO_3^- (Hakanen and Hölttä, 1992). In the pH range of 1-12 and Eh range of -0.3 V to 0.3 V, iodine is expected to be mostly in the form of the highly soluble anionic iodide I^- in the solution (Assemi and Erten, 1994).

Iodine is not strongly sorbed onto clay barrier materials or the minerals that constitute the rock mass that will surround a disposal vault (Oscarson et al., 1992). Its relatively rapid transport, coupled with its long half-life, means that ^{129}I , of all the radionuclides present in nuclear fuel waste, has the greatest potential of reaching the biosphere before decaying to insignificant levels (Oscarson et al., 1992).

Oscarson et al. (1992) found that there would be little advantage, regarding the travel time of ^{129}I within the barrier, to increase the density of the barrier material, given the very long half-life of ^{129}I . It should be noted here, however, that the flux will be decreased markedly by increasing the clay density.

Eriksen and Jansson (1996) measured the effective and apparent diffusivity of iodide in compacted clay at different ionic strengths and iodide concentrations. They used the through diffusion method. In contrast to many other investigations they found clear evidence of ion exclusion effects on the rate of transport. In summary the apparent diffusion coefficient D_a in low ionic strength water was $3.5 \cdot 10^{-11} \text{ m}^2/\text{s}$ and $9.2 \cdot 10^{-11} \text{ m}^2/\text{s}$ in saline water respectively. The effective diffusivities D_e were $2.1 \cdot 10^{-13} \text{ m}^2/\text{s}$ and $7.0 \cdot 10^{-12} \text{ m}^2/\text{s}$ in saline water respectively. The effective porosity was 0.006 and 0.06-0.11 for low and high ionic strength respectively.

Experimentally determined sorption and diffusion coefficient data of iodine are given in Table 7.

A realistic K_d -value of $0 \text{ m}^3/\text{kg}$ is proposed

On the basis of the experimentally obtained diffusion data, a D_a -value of $5 \cdot 10^{-11} \text{ m}^2/\text{s}$ is proposed for I^- in compacted bentonite. Considering the uncertainties in available data an effective porosity of 0.05 is proposed for low as well as high ionic strength waters.

Table 7. Experimentally determined diffusion coefficient and sorption data for iodine from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
bentonite SGW				0.001- 0.002	batch sorption	Assemi and Erten, 1994
Avonlea Na-bentonite	1250	2·10 ⁻¹³			I	Cheung et al., 1986
– " –	1500	2·10 ⁻¹³			– " –	– " –
– " –	1750	3·10 ⁻¹³			– " –	– " –
Avonlea Na-bentonite low-salinity water	ρ _d 1750	9·10 ⁻¹²			III	Cheung, 1989
Na-bentonite 0.1 M NaCl	ρ _d 1300	4.8·10 ⁻¹¹ time-lag	7.5·10 ⁻¹²	0.0034	III batch sorption	Choi and Oscarson, 1996
Ca-bentonite 0.05 M CaCl ₂	– " –	1.6·10 ⁻¹¹ time-lag	2.9·10 ⁻¹¹	0.0074	– " –	– " –
Na-bentonite pH 13.3	ρ _d 1100	1.8·10 ⁻¹⁰			IIa	Christiansen and Torstenfelt, 1988
Na-bentonite	ρ _b 2100	4·10 ⁻¹²			transient diff.	Eriksen and Jacobsson, 1981
– " –	– " –		2.1·10 ⁻¹³		III	– " –
– " –	– " –	4·10 ⁻¹²			IIb	Eriksen, 1982
– " –	– " –	3.5·10 ⁻¹¹			III time-lag	– " –
– " –	– " –		2·10 ⁻¹³		III	– " –
Ca-bentonite 25 °C	– " –		1.3·10 ⁻¹³		– " –	Eriksen et al., 1981
Na-bentonite MX-80 Ionic Str. 0.018	1800	3.5·10 ⁻¹¹	2.1·10 ⁻¹³		III	Eriksen and Jansson 1996
Na-bentonite MX-80 Ionic Str. 0.018	– " –	9.2·10 ⁻¹¹	7·10 ⁻¹²		– " –	– " –
Avonlea Na-bentonite, SGW	ρ _d 920	3.5·10 ⁻¹⁰			IIb	Oscarson et al., 1992
– " –	ρ _d 880	3.6·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 890	4.2·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1130	3.0·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1120	3.2·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1100	3.1·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1300	2.1·10 ⁻¹⁰			– " –	– " –

Table 7. Experimentally determined diffusion coefficient and sorption data for iodine from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Avonlea Na-bentonite, SGW	ρ _d 1300	1.8·10 ⁻¹⁰			IIb	Oscarson et al., 1992
– " –	ρ _d 1320	2.1·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1500	1.0·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1500	8.7·10 ⁻¹¹			– " –	– " –
– " –	ρ _d 1510	9.5·10 ⁻¹¹			– " –	– " –
– " –	ρ _d 920	1.05·10 ⁻⁹	3.3·10 ⁻¹¹		III	– " –
– " –	ρ _d 900	7.6·10 ⁻¹⁰	9.4·10 ⁻¹¹		– " –	– " –
– " –	ρ _d 1080	6.4·10 ⁻¹⁰	4.6·10 ⁻¹¹		– " –	– " –
– " –	ρ _d 1110	2.5·10 ⁻¹⁰	2.7·10 ⁻¹¹		– " –	– " –
– " –	ρ _d 1090	2.3·10 ⁻¹⁰	1.8·10 ⁻¹¹		– " –	– " –
– " –	ρ _d 1290	3.8·10 ⁻¹⁰	3.2·10 ⁻¹¹		– " –	– " –
– " –	ρ _d 1390	2.2·10 ⁻¹⁰	2.1·10 ⁻¹¹		– " –	– " –
– " –	ρ _d 1340	2.9·10 ⁻¹⁰	2.9·10 ⁻¹¹		– " –	– " –
– " –	ρ _d 1450	4.2·10 ⁻¹¹	1.8·10 ⁻¹²		– " –	– " –
– " –	ρ _d 1460	1.4·10 ⁻¹⁰	1.8·10 ⁻¹²		– " –	– " –
– " –	ρ _d 1460	2.6·10 ⁻¹¹	1.2·10 ⁻¹²		– " –	– " –
– " –	ρ _d 1610	1.6·10 ⁻¹⁰	1.0·10 ⁻¹²		– " –	– " –
– " –	ρ _d 1640	7.4·10 ⁻¹¹	1.2·10 ⁻¹²		– " –	– " –
– " –	ρ _d 1630	2.1·10 ⁻¹⁰	6.8·10 ⁻¹²		– " –	– " –
Na-bentonite DW	ρ _d 200	8.4·10 ⁻¹⁰			IIa	Sato et al., 1992
– " –	ρ _d 800	2.6·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1400	8.5·10 ⁻¹¹			– " –	– " –
– " –	ρ _d 2000	2.4·10 ⁻¹¹			– " –	– " –

Table 7. Experimentally determined diffusion coefficient and sorption data for iodine from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite SGW 1700 ppm salt	2000 ~ 10 % water	1.1·10 ⁻¹³ - 2.6·10 ⁻¹²		0.001	transient diff. batch sorption	Torstenfelt, 1986a
Na-bentonite + 0.5% Fe, SGW 1700 ppm salt	- " -	1.7·10 ⁻¹³ 1.9·10 ⁻¹²		0.001	- " -	- " -
Na-bentonite + 1% Fe, SGW 1700 ppm salt	- " -	5.3·10 ⁻¹⁴ - 1.2·10 ⁻¹²		0.001	- " -	- " -
Na-bentonite + 1% KMnO ₄ , SGW 1700 ppm salt	- " -	7.5·10 ⁻¹⁴ - 1.4·10 ⁻¹²		0.001	- " -	- " -

Lead

Lead will probably form carbonate and chloride complexes in groundwater, which may be anionic (Brandberg and Skagius, 1991).

A K_d -value for lead and Na-bentonite was found in the compilation made by Oscarson et al. (1984). From a measurement in a NH_4NO_3 -solution at pH 5, a K_d of $1 \text{ m}^3/\text{kg}$ was reported.

Goodwin et al. (1995) conducted diffusion experiments for lead in a 1-to-1 compacted mixture of silica sand/sodium bentonite. A single apparent diffusion coefficient could not satisfactorily fit the experimental concentration profile. It appeared that more than one lead species, each with a different diffusion coefficient, was involved in the transport process. If the diffusion coefficients are within the range of 10^{-15} to $10^{-11} \text{ m}^2/\text{s}$, the experimental results could be explained satisfactorily.

Experimentally determined diffusion coefficient data of Pb are given in Table 8.

Table 8. Experimentally determined diffusion coefficient data for Pb from the literature.

Material and condition	Density (kg/m ³)	D_a (m ² /s)	D_e (m ² /s)	K_d (m ³ /kg)	Measuring method	Reference
50/50 silica sand/ Na-bentonite, SGW	ρ_d 1500	10^{-15} - 10^{-11}			I	Goodwin et al., 1995

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A realistic K_d -value of $0.5 \text{ m}^3/\text{kg}$ is proposed and a conservative value of 0 is proposed, as proposed in SKB TR 91-16.

On the basis of the experimentally obtained diffusion data, a D_a -value of $1 \cdot 10^{-12} \text{ m}^2/\text{s}$ is proposed for Pb in compacted bentonite.

Neptunium

Neptunium is redox-sensitive in groundwater. Under oxic conditions Np(V) will dominate entirely and the most probable diffusing species are $\text{NpO}_2(\text{CO}_3)_2^{3-}$, $\text{NpO}_2\text{CO}_3^-$, $\text{NpO}_2(\text{OH})$ and possibly $\text{NpO}_2(\text{OH})_2^-$ (Eriksen and Jacobsson, 1984). Under reducing conditions the uncharged Np(IV) hydroxide $\text{Np}(\text{OH})_4$ is probably formed. At low pH, Np(V) species may have higher concentration in the solution than Np(IV) because of high solubility of NpO_2^+ (Pratopo et al., 1993). Under reducing conditions, formation of colloidal species was observed. The colloidal species could be $\text{Np}(\text{OH})_4$ and may affect the apparent K_d values (Pratopo et al., 1993).

Sorption of Np on the Na-bentonite Kunipia F at pH between 2 and 8 was studied by Kozai et al. (1993). The highest K_d ($>0.1 \text{ m}^3/\text{kg}$) appeared at pH 2, and then the K_d decreased with increasing pH until about pH 5, above which it maintained a constant value of about $0.02 \text{ m}^3/\text{kg}$ up to pH 8. The neptunium would not have access to the interlayer of bentonite immersed in near neutral solutions, because the interlayer spacing may be smaller than the radius of the hydrated neptunium ion. Consequently, most neptunium would be sorbed at the outer surface of bentonite. On the other hand, the hydrated neptunium ion would have access to a H^+ ion-expanded interlayer whose formation is favoured by low pH. In these circumstances, neptunium can be sorbed on both the outer surface and the interlayer of bentonite (Kozai et al., 1993). Neptunium sorbed under neutral pH was readily desorbed by 1 M KCl solution, while most Np sorbed by bentonite at low pH could not be desorbed by 1 M KCl solution but desorbed by 1 M HCl solution.

Bertetti et al. (1996) conducted batch sorption experiments for Np(V) on Namontmorillonite. The sorption begins to increase at a solution pH of about 7. For solution in equilibrium with atmospheric CO_2 , the magnitude of sorption peaks near pH 8-8.5 and decreases as the pH increases, forming a high pH sorption edge. Solutions that were capped and remained undersaturated with respect to atmosphere CO_2 show a continuous increase in sorption with increasing pH. There is a net uptake ($K_d = 20\text{-}30 \text{ ml/g}$) of Np(V) from pH~4.5-7, regardless of CO_2 conditions.

Experimentally determined sorption and diffusion coefficient data of Np are given in Table 9.

For oxidizing conditions, a realistic K_d -value of $0.02 \text{ m}^3/\text{kg}$ is proposed and a conservative value of $0.005 \text{ m}^3/\text{kg}$ is proposed. For reducing conditions, a realistic K_d -

value of 3 m³/kg is proposed and a conservative value of 0.1 m³/kg is proposed, as proposed in SKB TR 91-16.

On the basis of the experimentally obtained diffusion data, a D_a -value of $1 \cdot 10^{-12}$ m²/s and $2 \cdot 10^{-13}$ m²/s for oxidizing and reducing conditions respectively is proposed for Np in compacted bentonite at reducing conditions.

Table 9. Experimentally determined diffusion coefficient and sorption data for Np from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite + 1% Fe	ρ _d 2000	9.2·10 ⁻¹⁶			IIa	Albinsson et al., 1990b
Na-bentonite + 1% FeO	– " –	1.8·10 ⁻¹⁴ 3.5·10 ⁻¹⁴			– " –	– " –
Na-bentonite + 1% Cu	– " –	1.7·10 ⁻¹³			– " –	– " –
Na-bentonite + 1% Cu ₂ O	– " –	2·10 ⁻¹³			– " –	– " –
Na-bentonite + 1% vivianite	– " –	1.4·10 ⁻¹³			– " –	– " –
Na-bentonite + 1% magnetite	– " –	2.9·10 ⁻¹³			– " –	– " –
Wyoming MX-80 Na-bentonite, SGW				0.05 0.12	batch sorption	Allard et al., 1982
Na-bentonite, 5<pH<8				0.02	batch sorption	Kozai et al., 1993
Ca-bentonite, 0.1 M carbonate reducing				10 ^{1.01} as Np(OH) ₂ ²⁺ (CO ₃) ₂ ²⁻	batch sorption	Pratopo et al., 1993
– " –				10 ^{0.93} as Np(OH) ₄ ⁴⁺ (CO ₃) ₂ ⁴⁻	– " –	– " –
– " –				10 ^{-1.9} as NpO ₂ ⁺	– " –	– " –
– " –				10 ^{2.82} as NpO ₂ (OH) ₂ ⁻	– " –	– " –
Na-bentonite DW	ρ _d 200	1.2·10 ⁻¹¹			IIa	Sato et al., 1992
– " –	ρ _d 600	5.2·10 ⁻¹²			– " –	– " –
– " –	ρ _d 800	5.0·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1000	2.0·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1200	1.0·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1400	3.0·10 ⁻¹³			– " –	– " –

Table 9. Experimentally determined diffusion coefficient and sorption data for Np from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite DW	ρ _d 1600	2.0·10 ⁻¹³			IIa	Sato et al., 1992
– " –	ρ _d 1800	1.0·10 ⁻¹³			– " –	– " –
– " –	ρ _d 2000	3.0·10 ⁻¹⁴			– " –	– " –
Wyoming MX-80 Na-bentonite, SGW	ρ _d 1900 - 2000	2.2·10 ⁻¹³ 3.7·10 ⁻¹³			transient diff.	Torstenfelt et al., 1982
Wyoming MX-80 Na-bentonite + 0.5% Fe, SGW	ρ _d 1900 - 2300	4.5·10 ⁻¹⁵ 1.6·10 ⁻¹⁴			– " –	Torstenfelt et al., 1983
Wyoming MX-80 Na-bentonite, SGW	ρ _d 2000	8.2·10 ⁻¹³			– " –	Torstenfelt and Allard, 1986
Wyoming MX-80 Na-bentonite + 0.5% Fe, SGW	– " –	4.2·10 ⁻¹⁴			– " –	– " –
Wyoming MX-80 Na-bentonite + 1% Fe ₃ (PO ₄) ₂ , SGW	– " –	2.1·10 ⁻¹⁴			– " –	– " –
Kunigel V1 Na-bentonite, 40°C	ρ _b 1000	2·10 ⁻¹²			I	Tsukamoto et al., 1994
Kunigel V1 Na-bentonite, 90°C	– " –	5·10 ⁻¹³			– " –	– " –

Nickel

The solution chemistry of nickel is much the same as that of cobalt and iron(II) (Hakanen and Hölltä, 1992). In water, nickel is in the form of partly hydrated Ni^{2+} ion ($\text{Ni}(\text{H}_2\text{O})_6^{2+}$). In groundwater and bentonite water, the most important complexes are cationic (carbonate, hydroxide, sulphate), or neutral (hydroxide, sulphate). Sorption of nickel in groundwater on mineral surfaces of clays occurs as ion exchange. Nickel coprecipitates with insoluble hydroxides.

Some K_d -values for Ni in Na-montmorillonite were reported by Oscarson et al. (1984) for pH ranging from 6.1 to 6.8 at various ionic strengths of the solution. The values were from different experimental investigations. At an ionic strength of 0.002 M, K_d -values of 0.098 and 0.13 m^3/kg were given. From another investigation, K_d -values of 0.014 and 0.0013 m^3/kg were given for an ionic strength in the solution of 0.3 and 1.0 M, respectively. These results indicated a dependence of the sorption on the ionic strength at pH below 6.8.

Experimentally determined diffusion coefficient data of Ni are given in Table 10.

Table 10. Experimentally determined diffusion coefficient data for Ni from the literature.

Material and condition	Density (kg/m^3)	D_a (m^2/s)	D_e (m^2/s)	K_d (m^3/kg)	Measuring method	Reference
Na-bentonite pH 13.3	ρ_d 1100	$7.7 \cdot 10^{-13}$ partly as $\text{Ni}(\text{OH})_2$ or NiOH^+			IIa	Christiansen and Torstenfelt, 1988
– " –	– " –	$9.6 \cdot 10^{-12}$ partly as Ni^{2+}			– " –	– " –

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A realistic K_d -value of 0.1 m^3/kg is proposed and a conservative value of 0.02 m^3/kg is proposed.

On the basis of the experimentally obtained diffusion data, a D_a -value of $5 \cdot 10^{-12}$ m^2/s is proposed for Ni in compacted bentonite.

Niobium

In the groundwater conditions the oxidation state of niobium is Nb(V). The main features of the groundwater chemistry of niobium are its hydrolysis even under acidic conditions, the strong tendency to form colloids, and the sorption on hydrous silica to form carriercolloids (Hakanen and Hölttä, 1992).

In a compilation of sorption data for bentonite made by Oscarson et al. (1984), K_d -values were given for Nb in bentonite saturated with a synthetic groundwater with an ionic strength of 0.0055 M and a pH of 8.3-8.7. The reported values for a solution to solid ratio of 10 ml/g ranged from 0.18 to 0.51 m³/kg, and for a solution to solid ratio of 20 ml/g, K_d -values from 0.51 to 1.7 m³/kg were reported.

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A realistic K_d -value of 0.2 m³/kg is proposed and a conservative value of 0 is proposed, as proposed in SKB TR 91-16.

A D_a -value of $1 \cdot 10^{-12}$ m²/s is proposed for Nb in compacted bentonite.

Palladium

Pd forms oxyhydroxides in the valence states II and IV, of which only PdO·H₂O is stable. The dissolved chemical form is the palladous ion, [Pd(H₂O)]²⁺, or the palladite ion, PdO₂²⁻, both resulting from hydrolysis in basic conditions (Hakanen and Hölttä, 1992).

No experimental data on sorption of palladium have been found. A realistic K_d -value of 0.01 m³/kg is proposed and a conservative value of 0 is proposed, as proposed in SKB TR 91-16 (Brandberg and Skagius, 1991).

A D_a -value of $5 \cdot 10^{-12}$ m²/s is proposed for Pd in compacted bentonite.

Plutonium

Plutonium has four relatively stable oxidation states, III, IV, V, and VI. Under oxic conditions both Pu(V) and Pu(IV) could exist and probable diffusing species are $\text{Pu}(\text{OH})_4$ and possibly $\text{PuO}_2(\text{CO}_3)_2^{3-}$, depending on the carbonate concentration in the bentonite (Eriksen and Jacobsson, 1984). Under reducing conditions the major form is cationic Pu(III) complexes or, in low carbonate water, Pu(IV) hydroxide or Pu^{3+} and Pu(IV) sulphate (Hakanen and Hölttä, 1992).

The sorption mechanism of Pu under aerobic and especially under reducing conditions is probably ion exchange (Hakanen and Hölttä, 1992).

Idemitsu et al. (1990) used Method IIa to measure the diffusion coefficients of Pu in raw bentonite from Kuroishi mine, and raw bentonite, Na-bentonite and H-bentonite from Tsukinuno mine. Both mines are situated in Japan. The diffusion of Pu in Tsukinuno raw- and Na-bentonite was too slow to obtain a distinct concentration profile, and the apparent diffusion coefficient in these two types of bentonite was estimated to be less than $10^{-14} \text{ m}^2/\text{s}$. In H-bentonite, plutonium diffusion was observed, and the apparent diffusion coefficient was $3 \cdot 10^{-13} \text{ m}^2/\text{s}$ as the best fit. In Kuroishi raw bentonite, the apparent diffusion coefficient was $6 \cdot 10^{-13} \text{ m}^2/\text{s}$ as the best fit. Diffusion coefficients in H-bentonite showed a tendency to decrease with increasing density. Quartz content up to 50% or hematite up to 1% did not influence plutonium diffusion coefficient significantly in H-bentonite.

Experimentally determined sorption and diffusion coefficient data of Pu are given in Table 11.

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A realistic K_d -value of $3 \text{ m}^3/\text{kg}$ is proposed and a conservative value of $1 \text{ m}^3/\text{kg}$ is proposed, as proposed in SKB TR 91-16.

On the basis of the experimentally obtained diffusion data, a D_a -value of $5 \cdot 10^{-14} \text{ m}^2/\text{s}$ is proposed under reducing and oxidizing conditions for Pu in compacted bentonite.

Table 11. Experimentally determined diffusion coefficient and sorption data for Pu from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite + 1% FeO	ρ _d 2000	<3·10 ⁻¹⁶			IIa	Albinsson et al., 1991
Wyoming MX-80 Na-bentonite, SGW				3.5	batch sorption	Allard et al., 1982
Na-bentonite DW	200	7·10 ⁻¹⁴			IIa	Ashida et al., 1994
H-bentonite DW	1200	7·10 ⁻¹³			IIa	Idemitsu et al., 1990
- " -	1400	3·10 ⁻¹²			- " -	- " -
- " -	1600	3·10 ⁻¹³			- " -	- " -
- " -	1800	1·10 ⁻¹³			- " -	- " -
Koroishi raw-bentonite, DW	1600	6·10 ⁻¹³			- " -	- " -
85/15 H-bentonite/quartz, DW	1600	5·10 ⁻¹³			- " -	- " -
70/30 H-bentonite/quartz, DW	1600	3·10 ⁻¹³			- " -	- " -
50/50 H-bentonite/quartz, DW	1600	3·10 ⁻¹³			- " -	- " -
99.5/0.05 H-bentonite/hematite, DW	1600	3·10 ⁻¹³			- " -	- " -
99/1 H-bentonite/hematite, DW	1600	3·10 ⁻¹³			- " -	- " -
Wyoming MX-80 Na-bentonite, SGW	ρ _d 1900 - 2000	6.9·10 ⁻¹⁵ 3.0·10 ⁻¹⁴			transient diff.	Torstenfelt et al., 1982
Wyoming MX-80 Na-bentonite, SGW	ρ _d 1900 - 2300	3.2·10 ⁻¹⁵ 8.6·10 ⁻¹⁵			- " -	Torstenfelt et al., 1983
Wyoming MX-80 Na-bentonite, SGW	ρ _d 2000	1.9·10 ⁻¹⁵ (3.2·10 ⁻¹² a small fraction of Pu)			- " -	Torstenfelt and Allard, 1986

Protactinium

Protactinium has two stable oxidation states, Pa(IV) and Pa(V), under the redox conditions of natural waters. Under aerobic conditions, Pa(V) is expected to be dominating due to easy oxidation of Pa(IV). Pa(IV) is produced in the inert atmosphere only with strong reductants (Hakanen and Hölttä, 1992). The dominating species in groundwater is expected to be $\text{PaO}_2(\text{OH})_2^-$ (Eriksen and Jacobsson, 1984). Experimentally determined sorption and diffusion coefficient data of Pa are given in Table 12.

Table 12. Experimentally determined diffusion coefficient and sorption data for Pa from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Wyoming MX-80 Na-bentonite, SGW				5.0	batch sorption	Allard et al., 1982
Wyoming MX-80 Na-bentonite, SGW	ρ _d 1900 - 2000	6.0·10 ⁻¹³			– " –	Torstenfelt et al., 1982
Wyoming MX-80 Na-bentonite, SGW	ρ _d 2000	6.2·10 ⁻¹³			– " –	Torstenfelt and Allard, 1986

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). For fresh water a realistic K_d-value of 3 m³/kg is proposed and a conservative value of 0.1 m³/kg is proposed, as proposed in SKB TR 91-16. For saline water ten times lower values are proposed.

On the basis of the experimentally obtained diffusion data, a D_a-value of 1·10⁻¹² m²/s is proposed for Pa in compacted bentonite.

Radium

Like other alkaline earth elements, radium exists in solution as unhydrated Ra^{2+} ion. The chemistry of Ra is much the same as that of strontium. The main difference in saline groundwaters is the lower solubility of RaSO_4 (Hakanen and Hölttä, 1992).

Ames et al. (1983b) measured sorption of radium on montmorillonite in 0.01 M solutions. For a liquid to solid ratio of 20 ml/g and an initial radium concentration of about 10^{-9} to 10^{-7} M, a K_d -value of 3.1 to 3.7 m^3/kg was obtained at 25°C.

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A realistic K_d -value of 0.5 m^3/kg is proposed and a conservative value of 0.1 m^3/kg is proposed for sorption in fresh groundwater; and a realistic K_d -value of 0.01 m^3/kg is proposed and a conservative value of 0.001 m^3/kg is proposed for sorption in saline water, as proposed in SKB TR 91-16.

In analogy to Sr, a D_a -value of $2 \cdot 10^{-11}$ m^2/s is proposed for Ra in compacted bentonite.

Radon

No data on Rn have been found. Radon is a noble gas and is not expected to sorb (Brandberg and Skagius, 1991).

A D_a -value of $1 \cdot 10^{-10}$ m^2/s is proposed for Rn in compacted bentonite.

Samarium

No data on Sm have been found. As samarium is chemically similar to cerium, the same K_d -value as for cerium was proposed in SKB TR 91-16 (Brandberg and Skagius, 1991). A realistic K_d -value of 1 m^3/kg is proposed and a conservative value of 0.2 m^3/kg is proposed, as proposed in SKB TR 91-16.

A D_a -value of $1 \cdot 10^{-13}$ m^2/s is proposed for Sm in compacted bentonite.

Selenium

Selenium has the stable oxidation states -II, 0, +IV, and +VI under natural water conditions (Bruno and Sellin, 1992). Under oxic conditions selenium exists in the form of anionic selenate or selenite ions. In the oxic waters (pH over 7.5) the selenite SeO_3^{2-} dominates. Under reducing conditions, the thermodynamically dominant form is HSe^- . The reduction of selenate to Se(IV) is improbable under groundwater conditions (Hakanen and Hölttä, 1992).

Sato et al. (1995) measured the apparent diffusion coefficient of Se in Kunigel bentonite with degrees of compaction ranging from 400 to 1800 kg/m³. The diffusivities decreased from 4.3 to $0.68 \cdot 10^{-11}$ m²/s. Sorption values of 0.001- 0.002 m³/kg at pH between 8.1 and 9.1. Sorption of Se on Ca-bentonite were also given in a compilation of sorption data made by Oscarson et al. (1984). At a pH of 7, ionic strength of about 0.09 M and Eh of -160 mV, a K_d -value of 0.003 m³/kg was reported.

No further experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A realistic K_d -value of 0.003 m³/kg is proposed and a conservative value of 0 is proposed, as proposed in SKB TR 91-16.

A D_a -value of $1 \cdot 10^{-11}$ m²/s and a K_d -value of 0.003 m³/kg is proposed for Se in compacted bentonite.

Strontium

Strontium will prevail as Sr^{2+} in the bentonite porewater (Brandberg and Skagius, 1991).

The dominant sorption mechanism of Sr on bentonite was observed by Liang et al. (1993) to be an ion-exchange reaction, while physical adsorption functioned in a minor role.

Liang et al. (1993) measured the sorption of Sr on Wyoming MX-90 bentonite. A modified Freundlich isotherm was used to describe the sorption equilibrium. It was observed that the exponent in the Freundlich isotherm was near unity when the initial radionuclide concentration was <0.01 M, suggesting a linear isotherm.

Muurinen et al. (1985; 1987) measured the sorption and diffusion of strontium in sodium bentonite contacted with solutions with different salt concentrations. At high salt concentrations they obtained a much lower K_d -value than at low salt concentrations. This

was explained by that ion exchange was the predominant mechanism of sorption. Higher salt concentration in the solution means more cations competing about the available ion-exchange sites and consequently a lower K_d -value for strontium.

Recently Eriksen and Jansson (1996) measured the apparent diffusivity and sorption of cesium in compacted clay at different ionic strengths and cesium concentrations. They used the through diffusion method. Their experimental technique permitted them to separately evaluate the surface diffusion coefficient and the pore diffusion coefficient. They found that the lower the ionic strength was, the higher was the sorption and also the transport in the sorbed phase. In summary the apparent diffusion coefficient D_a in low ionic strength water was $1.2 \cdot 10^{-11}$ m²/s and $1.0 \cdot 10^{-11}$ m²/s in saline water respectively. The effective diffusivities D_e were $6.4 \cdot 10^{-10}$ m²/s and $3.5 \cdot 10^{-10}$ m²/s in saline water respectively.

Experimentally determined sorption and diffusion coefficient data of Sr are given in Table 13.

Although some new sorption data for strontium have been found since SKB TR 91-16 (Brandberg and Skagius, 1991), the K_d -values proposed in SKB TR 91-16 seem still pertinent. A realistic K_d -value of 0.5 m³/kg is proposed and a conservative value of 0.1 m³/kg is proposed for sorption in fresh groundwater; and a realistic K_d -value of 0.01 m³/kg is proposed and a conservative value of 0.001 m³/kg is proposed for sorption in saline water, as proposed in SKB TR 91-16.

On the basis of the experimentally obtained diffusion data, a D_a -value of $2 \cdot 10^{-11}$ m²/s is proposed for Sr in compacted bentonite.

Table 13. Experimentally determined diffusion coefficient and sorption data for Sr from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Ca-bentonite	ρ _d 1590	1.1·10 ⁻¹¹		2.139	IIa batch sorption	Choi et al., 1992
Na-bentonite 0.1 M NaCl	ρ _d 1300	8.5·10 ⁻¹² time-lag	2.4·10 ⁻¹¹	0.076	III batch sorption	Choi and Oscarson, 1996
Ca-bentonite 0.05 M CaCl ₂	– " –	1.8·10 ⁻¹¹ time-lag	1.5·10 ⁻¹⁰	0.012	– " –	– " –
Na-bentonite pH 13.3	ρ _d 1100	5.8·10 ⁻¹² partly as Sr ²⁺			IIa	Christiansen and Torstenfelt, 1988
– " –	– " –	2.3·10 ⁻¹³ partly as SrCO ₃			– " –	– " –
Na-bentonite 25 °C	ρ _b 2100	2.3·10 ⁻¹¹		0.6	transient diff. sorption on compacted bentonite	Eriksen et al., 1981
Na-bentonite 70 °C	– " –	4.1·10 ⁻¹¹			I	– " –
Ca-bentonite 25 °C	– " –	4.8·10 ⁻¹¹			– " –	– " –
Ca-bentonite 70 °C	– " –	2.6·10 ⁻¹¹			– " –	– " –
bentonite 25 °C, L/S 0.25 - 1 ml/g				0.6 - 1.3	batch sorption	– " –
Na-bentonite	– " –	1.4·10 ⁻¹² - 1.5·10 ⁻¹²			IIb	Eriksen and Jacobsson, 1981
– " –	– " –	8·10 ⁻¹² - 1·10 ⁻¹¹			IIa	– " –
Ca-bentonite	ρ _b 1600	7·10 ⁻¹² - 3.2·10 ⁻¹¹			IIb	– " –
– " –	ρ _b 2100	1.3·10 ⁻¹¹			– " –	– " –
– " –	ρ _b 1600	1.5·10 ⁻¹¹ - 1.6·10 ⁻¹¹			IIa	– " –
Na-bentonite MX-80 Ionic Str. 0.018	ρ _d 1800	1.2·10 ⁻¹¹	6.7·10 ⁻⁹	-	III	Eriksen & Jansson 1996
Na-bentonite MX-80 Ionic Str. 0.218	ρ _d 1800	1.0·10 ⁻¹¹	3.5·10 ⁻¹⁰	0.006	– " –	– " –
montmorillonite GW 1 g/l salt				1.5	batch sorption	Erten et al., 1988

Table 13. Experimentally determined diffusion coefficient and sorption data for Sr from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
bentonite 2 g/l cation	400	7.3·10 ⁻¹¹		0.0209	IIb batch sorption	Gillham et al., 1983
bentonite, water 0.001M Na 0.002 M Ca 0.001 M Mg				0.05- 0.25 (med. 0.11)	batch sorption	Horyna and Dlouhy, 1988
Wyoming Na- bentonite, DW				0.555	batch sorption	Kim et al., 1993
- " -	ρ _d 1040	1.93·10 ⁻¹¹ 1.82·10 ⁻¹¹			IIa	- " -
- " -	ρ _d 1240	1.63·10 ⁻¹¹ 1.52·10 ⁻¹¹			- " -	- " -
- " -	ρ _d 1570	1.03·10 ⁻¹¹ 1.14·10 ⁻¹¹			- " -	- " -
- " -	ρ _d 1730	6.41·10 ⁻¹² 6.62·10 ⁻¹²			- " -	- " -
- " -	ρ _d 1900	3.4·10 ⁻¹² 3.13·10 ⁻¹²			- " -	- " -
bentonite, DW pH 2.7				0.06	batch sorption	Konishi et al., 1988
bentonite, DW pH 9.4				6.3	- " -	- " -
bentonite, SGW pH 2.7				0.04	- " -	- " -
bentonite, SGW pH 9.4				4.02	- " -	- " -
Na-bentonite 0.6 M NaCl	ρ _d 1740 diff.exp	6.9·10 ⁻¹²		0.007	transient diff. batch sorption	Muurinen et al., 1985
Na-bentonite 0.001 M NaCl	- " -	9.1·10 ⁻¹²		1.5	- " -	- " -
Na-bentonite GW 10 g/l salt	ρ _d 2000 diff.exp	8.6·10 ⁻¹²		0.05	- " -	- " -
Na-bentonite 0.001 M NaCl				1.465	batch sorption	Muurinen et al., 1987
Na-bentonite 0.01 M NaCl				1.465	- " -	- " -

Table 13. Experimentally determined diffusion coefficient and sorption data for Sr from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite 0.1 M NaCl				0.087	batch sorption	Muurinen et al., 1987
Na-bentonite 0.5 M NaCl				0.01	– " –	– " –
Na-bentonite 1 M NaCl				0.003	– " –	– " –
Na-bentonite 0.01 M NaCl	ρ _d 1750			1.204	sorption on compacted bentonite	– " –
Na-bentonite 0.1 M NaCl	– " –	8.7·10 ⁻¹² (3.1·10 ⁻¹² time-lag)	2.9·10 ⁻⁹	0.191	III sorption on compacted bentonite	– " –
Na-bentonite 0.5 M NaCl	– " –	6·10 ⁻¹² (4.5·10 ⁻¹² time-lag)	2.9·10 ⁻⁹	0.008- 0.027	– " –	– " –
bentonite, SGW, L/S 0.5 ml/g				2.7	batch sorption	Neretnieks and Skagius, 1978
bentonite, SGW	ρ _b 2350	3.3·10 ⁻¹¹				– " –
Wyoming MX-80, SGW	ρ _d 1600	1.7·10 ⁻¹¹			IIa	Ramebäck et al., 1994
– " –	ρ _d 2100	6.3·10 ⁻¹²			– " –	– " –
Na-bentonite DW	ρ _d 200	5.1·10 ⁻¹¹			IIa	Sato et al., 1992
– " –	ρ _d 400	3.4·10 ⁻¹¹			– " –	– " –
– " –	ρ _d 600	2.5·10 ⁻¹¹			– " –	– " –
– " –	ρ _d 700	1.7·10 ⁻¹¹			– " –	– " –
– " –	ρ _d 1000	7.6·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1200	7.6·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1500	5.4·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1700	4.6·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1900	5.2·10 ⁻¹²			– " –	– " –

Table 13. Experimentally determined diffusion coefficient and sorption data for Sr from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite DW	ρ _d 2000	4.3·10 ⁻¹²			IIa	Sato et al., 1992
Na-bentonite SGW 1700 ppm salt	2000 ~ 10 % water	2·10 ⁻¹¹		2.9	transient diff. batch sorption	Torstenfelt, 1986a

Tchnetium

Tchnetium is a redox sensitive element. It can exist in the VII, IV, and 0 oxidation states (Sawatsky and Oscarson, 1991b). Tc is an anion under oxidizing conditions but a cation under reducing conditions (Albinsson et al., 1991).

In oxidizing environments, Tc exists as the negatively charged pertechnetate species (TcO₄⁻), which is very poorly sorbed, if at all, on bentonite (Sawatsky and Oscarson, 1991a). TcO₄⁻ is highly soluble. Under reducing conditions technetium is removed from water by the formation of poorly soluble solids and sorption of the neutral oxyhydroxide. The chemical form of Tc(IV) is TcO(OH)₂ and the stable solid phase probably TcO₂ (Hakanen and Hölttä, 1992).

Sawatsky and Oscarson (1991b) measured the apparent diffusion coefficients for technetium in the Avonlea bentonite under oxidizing and reducing conditions by Method IIb. In the reducing environment, the apparent diffusion coefficient values were one to two orders of magnitude lower than those obtained under oxidizing conditions. The concentration profiles in the bentonite specimen obtained in the reducing experiments showed a sharp drop at the interface. These phenomena suggested that, in addition to the diffusion process, Tc was being transformed to a less mobile form, probably due to the reduction of Tc(VII) to Tc(IV) and the subsequent formation of a Tc(IV)-carbonate complex, perhaps Tc(OH)(CO₃)₂⁻. This complex is larger and has a different stereochemistry than the TcO₄⁻ species present in the oxidizing environments (Sawatsky and Oscarson, 1991b). At 80°C, the apparent diffusion coefficient values were two to three times greater than those obtained at 25°C at all densities (Sawatsky and Oscarson, 1991b).

The slow diffusion rates were also reported in the study of Albinsson et al. clearly indicating reduction from Tc(VII) (TcO₄⁻) to Tc(IV) (Albinsson et al., 1991).

Experimentally determined sorption and diffusion coefficient data of Tc are given in Table 14.

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A K_d -value of 0 is proposed for sorption under oxidizing conditions; and for sorption under reducing conditions, a realistic K_d -value of $0.1 \text{ m}^3/\text{kg}$ is proposed and a conservative value of $0.01 \text{ m}^3/\text{kg}$ is proposed, as proposed in SKB TR 91-16.

On the basis of the experimentally obtained diffusion data, a D_a -value of $2 \cdot 10^{-12} \text{ m}^2/\text{s}$ is proposed for both oxidizing and reducing conditions for Tc in compacted bentonite.

Table 14. Experimentally determined diffusion coefficient and sorption data for Tc from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite aerobic	ρ _d 2000	2·10 ⁻¹²			IIa	Albinsson et al., 1990a
Na-bentonite + 1% Fe	ρ _d 2000	1.1·10 ⁻¹⁴			IIa	Albinsson et al., 1991
Na-bentonite + 1% FeO	– " –	5.4·10 ⁻¹³			– " –	– " –
Na-bentonite + 1% Cu	– " –	4.4·10 ⁻¹²			– " –	– " –
Na-bentonite + 1% Cu ₂ O	– " –	1.5·10 ⁻¹¹			– " –	– " –
bentonite, oxidizing				0.0002 as TcO ₄ ⁻	batch sorption	Ito and Kanno, 1988
bentonite, reducing				0.0472	– " –	– " –
Na-bentonite DW	ρ _d 200	6.9·10 ⁻¹⁰			IIa	Sato et al., 1992
– " –	ρ _d 600	3.1·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1000	1.3·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1400	3.2·10 ⁻¹¹			– " –	– " –
– " –	ρ _d 1800	1.2·10 ⁻¹¹			– " –	– " –
– " –	ρ _d 2000	1.0·10 ⁻¹¹			– " –	– " –
Avonlea Na-bentonite, SGW, oxidizing	ρ _d 900	8·10 ⁻¹¹			III	Sawatsky and Oscarson, 1991a
– " –	ρ _d 1300	4·10 ⁻¹²			– " –	– " –
Avonlea Na-bentonite, SGW, oxidizing, 25°C	ρ _d 920	2.3·10 ⁻¹⁰			IIb	Sawatsky and Oscarson, 1991b
– " –	ρ _d 910	2.3·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 930	2.3·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1130	1.8·10 ⁻¹⁰			– " –	– " –
– " –	ρ _d 1120	1.9·10 ⁻¹⁰			– " –	– " –

Table 14. Experimentally determined diffusion coefficient and sorption data for Tc from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Avonlea Na-bentonite, SGW, oxidizing, 25°C	ρ _d 1130	1.7·10 ⁻¹⁰			IIb	Sawatsky and Oscarson, 1991b
— " —	ρ _d 1340	7.3·10 ⁻¹¹			— " —	— " —
— " —	ρ _d 1340	1.0·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 1360	9.4·10 ⁻¹¹			— " —	— " —
Avonlea Na-bentonite, SGW, oxidizing, 80°C	ρ _d 940	5.2·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 940	6.2·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 940	4.9·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 1150	3.2·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 1180	3.1·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 1180	3.5·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 1350	2.1·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 1370	1.7·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 1360	1.9·10 ⁻¹⁰			— " —	— " —
Avonlea Na-bentonite, SGW, reducing, 25°C	ρ _d 1360	6.3·10 ⁻¹²			— " —	— " —
— " —	ρ _d 1240	8.9·10 ⁻¹²			— " —	— " —
— " —	ρ _d 1300	7.0·10 ⁻¹²			— " —	— " —
— " —	ρ _d 1290	8.2·10 ⁻¹²			— " —	— " —
— " —	ρ _d 1280	2.7·10 ⁻¹²			— " —	— " —
— " —	ρ _d 1280	4.7·10 ⁻¹²			— " —	— " —

Table 14. Experimentally determined diffusion coefficient and sorption data for Tc from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite SGW 1700 ppm salt	2000 ~ 10 % water	1.2·10 ⁻¹² as Tc(VII)		0	transient diff. batch sorption	Torstenfelt, 1986a
– " –	– " –	6.7·10 ⁻¹³ as Tc(VII)			– " –	– " –
Na-bentonite + 0.5% Fe, SGW 1700 ppm salt	– " –	1.5·10 ⁻¹³ as Tc(IV)		0.05	– " –	– " –
– " –	– " –	8.4·10 ⁻¹⁴ as Tc(IV)		0.05	– " –	– " –

Thorium

Thorium has the oxidation state Th(IV) in water. The Th⁴⁺ ion has a strong tendency to hydrolyse and to form hydroxy complexes that can polymerise to colloids. In groundwater, thorium probably exists in hydrolysed form and sorption of thorium on minerals is probably by adsorption and not pH dependent. In non-complexing (other than hydroxide) conditions, sorption of thorium on bentonite is high. The diffusion of thorium in bentonite is reported to be the same as that of Pu(IV) and americium (Hakanen and Hölttä, 1992).

Experimentally determined sorption and diffusion coefficient data of Th are given in Table 15.

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A realistic K_d-value of 3 m³/kg is proposed and a conservative value of 0.1 m³/kg is proposed, as proposed in SKB TR 91-16.

On the basis of the experimentally obtained diffusion data, a D_a-value of 1·10⁻¹⁴ m²/s is proposed for Th in compacted bentonite.

Table 15. Experimentally determined diffusion coefficient and sorption data for Th from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Wyoming MX-80 Na-bentonite, SGW				>6.3	batch sorption	Allard et al., 1982
Wyoming MX-80 Na-bentonite, SGW	ρ _d 1900 - 2000	4.6·10 ⁻¹⁵ 8.5·10 ⁻¹⁵			- " -	Torstenfelt et al., 1982
Wyoming MX-80 Na-bentonite, SGW	ρ _d 2000	7.7·10 ⁻¹⁵			- " -	Torstenfelt and Allard, 1986

Tin

In the nature, tin exists mainly in the form of Sn(IV). In solution, Sn(II) is readily oxidised to Sn(IV) by dissolved oxygen, especially at elevated temperatures. Under reducing conditions, tin is still most probably in Sn(IV) form. Both Sn(IV) and Sn(II) are strongly hydrolysed and precipitated in pH range from 4 to 9, forming sparingly soluble components like SnOCl₂ and Sn(OH)₂Cl₂. In basic solutions, Sn(OH)₄ partially dissolves due to the formation of [Sn(OH)₆]²⁻ and Sn(OH)₂ is dissolved as HSnO₂⁻. Speciation calculations indicate that the dominating form of tin in solution is the tetrahydroxide Sn(OH)₄ (Hakanen and Hölttä, 1992).

No sorption data on Sn have been found. A realistic K_d-value of 3 m³/kg was proposed in SKB TR 91-16 in analogy with thorium, and a conservative value of 0.01 m³/kg was proposed in SKB TR 91-16.

A D_a-value of 1·10⁻¹⁴ m²/s is proposed for Sn in compacted bentonite in analogy with thorium.

Uranium

In natural waters, uranium has three thermodynamically stable oxidation states, IV, V, and VI. The chemistry of uranium is known to be very sensitive to the surrounding conditions (Muurinen and Lehtikoinen, 1992). The valence of uranium depends on redox conditions. The change of valence affects the speciation and solubility. Uranium also forms carbonate and hydroxyl complexes, which also affect solubility. In aerated solutions uranium is in the U(VI) oxidation state and hydroxides and carbonate complexes $\text{UO}_2(\text{CO}_3)_3^{4-}$, $\text{UO}_2(\text{CO}_3)_2^{2-}$, and $\text{UO}_2(\text{OH})_3^-$ are probably the dominating species (Eriksen and Jacobsson, 1984). On addition of reducing agents to the clay the uncharged U(IV) hydroxide $\text{U}(\text{OH})_4$ will be present in increasing concentration.

Ames et al. (1983a) measured the sorption of uranium onto montmorillonite in 0.01 M NaCl solution and in 0.01 M NaHCO_3 solution under oxidizing conditions. Their results showed dependence of K_d -values on the concentration of uranium and other salts. The K_d -values obtained in the 0.01 M NaCl solution were 0.36 and 0.53 m^3/kg for the initial uranium concentration of 10^{-4} and $3.5 \cdot 10^{-7}$ M, respectively. The K_d -values obtained in the 0.01 M NaHCO_3 solution were 0.086 and 0.068 m^3/kg for the initial uranium concentration of 10^{-4} and $3.5 \cdot 10^{-7}$ M, respectively.

Idemitsu et al. (1996) measured the diffusion coefficients of uranium in Na-bentonite containing corrosion products of carbon steel which creates a reducing environment. The presence of carbon steel decreased the apparent diffusion coefficient by one order of magnitude. Idemitsu et al. also observed that the apparent diffusion coefficient decreased with increasing density, and the apparent diffusion coefficient was independent of the sorption distribution coefficient.

Sorption and diffusion measurements of uranium in sodium bentonite MX-80 was reported (Muurinen et al., 1989; Muurinen, 1990). A general conclusion was that there was usually no sorption. In some experiments with high uranyl nitrate concentrations, sorption or possibly precipitation was, however, evident. This was caused by the low pH-value of the mixture when high tracer concentration was used (Muurinen, 1990). The diffusion coefficients of uranium were strongly dependent on the compaction of bentonite so that in the highly compacted samples the diffusion was very restricted (Muurinen, 1990). On the basis of the sorption experiments there seemed to be ion exclusion and no sorption. The high dependence of the diffusion coefficients on the compaction also pointed to the ion exclusion phenomenon.

In the experiments by Torstenfelt and Allard (1986), a fast moving fraction was identified with apparent diffusion coefficient about one order of magnitude greater than the rest of the uranium.

Pusch et al. (1989) observed a rapid retardation of the advance of the uranium front in bentonite. They stated that a probable explanation could be that uranium was precipitated in the form of $\text{Na}_2\text{U}_2\text{O}_7 \cdot (\text{H}_2\text{O})_3$ and/or $\text{CaU}_6\text{O}_{19} \cdot (\text{H}_2\text{O})_{10}$, by interaction of released sodium or calcium and uranium ions in the presence of hydroxyls provided by the smectite and porewater. This effect is particularly strong at high clay density and low temperature.

Muurinen and Lehtikoinen (1992) studied experimentally the diffusion of uranium in compacted sodium bentonite (Wyoming MX-80) where the clay density, salt content of the solution and redox conditions varied. In the studies with non-saline water (TDS \approx 300 ppm), uranium occurred in aerobic experiments as anionic carbonate complexes and followed the anionic diffusion mechanism. Uranium dissolved in aerobic conditions stayed mostly on valence U(VI) when the solution was taken into the anaerobic box for anaerobic experiments and the Eh-value decreased from 0.4 V to 0.1 V. In the studies with saline water (TDS \approx 35000 ppm), uranium occurred in aerobic experiments probably as cationic hydroxide complexes and followed the cationic diffusion mechanism. Uranium dissolved in aerobic conditions precipitated in the anaerobic diffusion experiments. Uranium in those experiments probably occurred as $\text{U}(\text{OH})_4$ and followed the diffusion mechanism of neutral species.

Tests were made to find out the difference in solubility of uranium in bentonite porewater at room temperature and at 90 °C (Pusch et al., 1989). At room temperature the solubility of uranium was found to be 25-75 ppm, while at 90 °C it was found to be considerably lower. It is expected, however, that pH in the clay voids is affected by the uptake of uranium, which thus controls the actual solubility. Uranium was found to migrate much more rapidly at 90 °C than at room temperature (Pusch et al., 1989).

Experimentally determined sorption and diffusion coefficient data of U are given in Table 16.

Table 16. Experimentally determined diffusion coefficient and sorption data for U from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite + 1% Fe	ρ _d 2000	1.2·10 ⁻¹⁴			IIa	Albinsson et al., 1991
Na-bentonite + 1% FeO	– " –	2.8·10 ⁻¹⁴			– " –	– " –
Na-bentonite + 1% Cu	– " –	1.4·10 ⁻¹³			– " –	– " –
Wyoming MX-80 Na-bentonite, SGW				0.093	batch sorption	Allard et al., 1982
Kunigel V1 Na-bentonite	ρ _d 1000	2.2·10 ⁻¹²			I	Idemitsu et al., 1996
– " –	ρ _d 1200	1.2·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1400	1.3·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1600	8.6·10 ⁻¹³			– " –	– " –
– " –	ρ _d 1800	4.2·10 ⁻¹³			– " –	– " –
– " –	ρ _d 2000	4.5·10 ⁻¹³			– " –	– " –
Kunigel V1 Na-bentonite + carbon steel	ρ _d 1000	4.4·10 ⁻¹³			– " –	– " –
– " –	ρ _d 1200	2.5·10 ⁻¹³			– " –	– " –
– " –	ρ _d 1400	2.1·10 ⁻¹³			– " –	– " –
– " –	ρ _d 1600	7.3·10 ⁻¹⁴			– " –	– " –
– " –	ρ _d 1800	4.8·10 ⁻¹⁴			– " –	– " –
– " –	ρ _d 2000	3.4·10 ⁻¹⁴			– " –	– " –
Kunipia F Na-bentonite	ρ _d 800	3.7·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1000	6.9·10 ⁻¹³			– " –	– " –
– " –	ρ _d 1200	3.3·10 ⁻¹³			– " –	– " –
– " –	ρ _d 1400	8.7·10 ⁻¹⁴			– " –	– " –
– " –	ρ _d 1600	4.4·10 ⁻¹⁴			– " –	– " –

Table 16. Experimentally determined diffusion coefficient and sorption data for U from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Kunipia F Na-bentonite	ρ _d 1800	3.1·10 ⁻¹⁴			I	Idemitsu et al., 1996
Kunipia F Na-bentonite + carbon steel	ρ _d 800	2.2·10 ⁻¹²			– " –	– " –
– " –	ρ _d 1000	1.2·10 ⁻¹³			– " –	– " –
– " –	ρ _d 1200	8.8·10 ⁻¹⁴			– " –	– " –
– " –	ρ _d 1400	6.5·10 ⁻¹⁴			– " –	– " –
– " –	ρ _d 1600	4.5·10 ⁻¹⁴			– " –	– " –
– " –	ρ _d 1800	4.3·10 ⁻¹⁴			– " –	– " –
bentonite L/S 50 aerobic				0.06	batch sorption	Morgan et al., 1988
bentonite L/S 100 aerobic				0.14	– " –	– " –
bentonite L/S 200 aerobic				0.33	– " –	– " –
Na-bentonite SGW 1056 ppm salt 600 ppm HCO ₃ ⁻ aerobic	ρ _d 600 - 2100			0.0005 - 0.002	sorption on compacted bentonite	Muurinen et al., 1989
– " –	ρ _d 600	2.2·10 ⁻¹¹ (4.8·10 ⁻¹² time-lag) as UO ₂ (NO ₃) ₂	8.9·10 ⁻¹²		III	– " –
– " –	ρ _d 900	3.8·10 ⁻¹² (8.4·10 ⁻¹³ time-lag) as UO ₂ (NO ₃) ₂	2.1·10 ⁻¹²		– " –	– " –
– " –	ρ _d 1100	1.6·10 ⁻¹² as UO ₂ (NO ₃) ₂	4.9·10 ⁻¹³		– " –	– " –
– " –	ρ _d 600	2.6·10 ⁻¹¹ (3·10 ⁻¹² time-lag) as UO ₂ Cl ₂	7.7·10 ⁻¹²		– " –	– " –

Table 16. Experimentally determined diffusion coefficient and sorption data for U from the literature (continued).

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Na-bentonite SGW 1056 ppm salt 600 ppm HCO ₃ ⁻ aerobic	ρ _d 1200	1.2·10 ⁻¹³ as UO ₂ Cl ₂	6·10 ⁻¹⁴		III	Muurinen et al., 1989
-- " --	ρ _d 600	5.3·10 ⁻¹² (2.6·10 ⁻¹² time-lag) U dissolved from UO ₂	8.3·10 ⁻¹²		-- " --	-- " --
Na-bentonite 35000 ppm salt, anaerobic	ρ _d 1710			> 0.846	sorption on compacted bentonite	Muurinen and Lehikoinen, 1992
Na-bentonite 300 ppm salt, aerobic	ρ _d 700	6.3·10 ⁻¹³	1.9·10 ⁻¹²	0.0034	III sorption on compacted bentonite	-- " --
Na-bentonite 300 ppm salt, anaerobic	ρ _d 760	1.3·10 ⁻¹³ - 1.7·10 ⁻¹³	1.9·10 ⁻¹²	0.0132	-- " --	-- " --
Na-bentonite 35000 ppm salt, aerobic	ρ _d 1150	6.2·10 ⁻¹³	5.8·10 ⁻¹²	0.0104	-- " --	-- " --
Na-bentonite 35000 ppm salt, anaerobic	ρ _d 1150	7.5·10 ⁻¹⁴	5.7·10 ⁻¹¹	0.63	-- " --	-- " --
Na-bentonite SGW salt 1700 ppm	2000 ~ 10 % water	2.7·10 ⁻¹³ - 3.4·10 ⁻¹²			transient diff.	Torstenfelt, 1986b
Na-bentonite + 0.5% Fe, SGW 1700 ppm salt	-- " --	1.6·10 ⁻¹³ 3.3·10 ⁻¹¹			-- " --	-- " --
Na-bentonite + 1% Fe ₃ (PO ₄) ₂ SGW 1700 ppm salt	-- " --	1·10 ⁻¹³ 3.1·10 ⁻¹³ 8.2·10 ⁻¹²			-- " --	-- " --
Na-bentonite SGW 1700 ppm salt, 0.6 g/l NaHCO ₃	-- " --	8.3·10 ⁻¹⁴ 2.3·10 ⁻¹³ 8.5·10 ⁻¹²			-- " --	-- " --
Na-bentonite SGW 1700 ppm salt, 10 mg/l humic acid	-- " --	2.3·10 ⁻¹³ 6.8·10 ⁻¹³ 6.7·10 ⁻¹²			-- " --	-- " --

For oxidizing conditions, a realistic K_d -value of $0.05 \text{ m}^3/\text{kg}$ is proposed and a conservative value of 0.005 is proposed, as proposed in SKB TR 91-16 (Brandberg and Skagius, 1991). For reducing conditions, a realistic K_d -value of $1 \text{ m}^3/\text{kg}$ is proposed and a conservative value of $0.01 \text{ m}^3/\text{kg}$ is proposed.

On the basis of the experimentally obtained diffusion data, a D_a -value of $1 \cdot 10^{-12} \text{ m}^2/\text{s}$ and $2 \cdot 10^{-13} \text{ m}^2/\text{s}$ is proposed for oxidizing and reducing conditions respectively for U in compacted bentonite.

Zirconium

Zirconium has the oxidation state IV in groundwater. The chemical form in water is the Zr^{4+} ion or its hydrolysis products (Hakanen and Hölttä, 1992). In neutral solutions $\text{Zr}^{4+}(\text{aq})$ tends to directly convert to tetranuclear $[\text{Zr}_4(\text{OH})_3(\text{H}_2\text{O})_{16}]^{8+}$ ion. In groundwater, Zr ion forms anionic complexes only with sulphate and fluoride ions. Cationic hydroxyl complexes have a strong tendency to polymerise and to form colloids. At high pH, the anionic hydroxyl complex (metazirconate ion) is the major soluble species.

Migration of non-colloidal zirconium by diffusion is expected to be slow (Hakanen and Hölttä, 1992). Sato et al. (1995) measured the apparent diffusion coefficient of Zr in Kunigel bentonite with degrees of compaction ranging from 400 to $1800 \text{ kg}/\text{m}^3$. The diffusivities decreased from 5 to $1 \cdot 10^{-14} \text{ m}^2/\text{s}$ for compactations between 400 and $1400 \text{ kg}/\text{m}^3$. No sorption data for compacted bentonite have been found. Allard et al. (1977) have, however, reported K_d -values for sorption of Zr on a mixture of bentonite (10%) and quartz (90%) to be in the range 0.05 to $1 \text{ m}^3/\text{kg}$.

No new experimental sorption data have been found since SKB TR 91-16 (Brandberg and Skagius, 1991). A realistic K_d -value of $2 \text{ m}^3/\text{kg}$ is proposed and a conservative value of $0.2 \text{ m}^3/\text{kg}$ is proposed, as proposed in SKB TR 91-16.

A D_a -value of $1 \cdot 10^{-14} \text{ m}^2/\text{s}$ is proposed for Zr in compacted bentonite.

Tritiated water

We also include data on tritiated water because we will use these data later in the discussion on diffusion mechanisms. Table 17 below is a compilation of available data.

Table 17. Experimentally determined diffusion coefficient and sorption data for HTO from the literature.

Material and condition	Density (kg/m ³)	D _a (m ² /s)	D _e (m ² /s)	K _d (m ³ /kg)	Measuring method	Reference
Wyoming MX-80 Na-bentonite, DW	ρ _d 2100		1.8·10 ⁻¹¹		III	Neretnieks, 1982
Na-bentonite, SGW	ρ _d 2100		3.6·10 ⁻¹²		III	Eriksen, 1982
— " —	ρ _d 1800		6.5·10 ⁻¹¹		— " —	— " —
bentonite	400	1.2·10 ⁻⁹			transient diff.	Gillham et al., 1984
Kunipia F Na-bentonite	200	1.4·10 ⁻⁹		0	batch sorption IIa	Miyahara et al., 1991
— " —	400	8.2·10 ⁻¹⁰			IIa	— " —
— " —	800	3.8·10 ⁻¹⁰			— " —	— " —
— " —	1200	1.9·10 ⁻¹⁰			— " —	— " —
— " —	1600	9.4·10 ⁻¹¹			— " —	— " —
Kunipia F Na-bentonite, DW	ρ _d 200	1.1·10 ⁻⁹			IIa	Sato et al., 1992
— " —	ρ _d 400	6.9·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 800	4.8·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 1200	1.8·10 ⁻¹⁰			— " —	— " —
— " —	ρ _d 1600	9.5·10 ⁻¹¹			— " —	— " —
Na-bentonite, 0.1 M NaCl	ρ _d 1300	8.0·10 ⁻¹¹	7.8·10 ⁻¹¹		III	Choi and Oscarson, 1996
Ca-bentonite, 0.05 M CaCl ₂	— " —	1.1·10 ⁻¹⁰	1.5·10 ⁻¹⁰		— " —	— " —
0.5/0.5 basalt/bentonite SGW	ρ _b 1540	1.89·10 ⁻¹¹			IIb	Relyea et al., 1986
— " —	ρ _b 1580	1.82·10 ⁻¹¹			— " —	— " —

An effective diffusivity of 2·10⁻¹¹ m²/s is proposed for compacted bentonite.

5 DISCUSSION AND CONCLUSIONS

In this report, recent studies on sorption and diffusion of radionuclides in compacted bentonite have been reviewed. The sorption distribution coefficient and diffusion coefficient data obtained from experiments in the literature have been compiled. Based on these experimental data and the report SKB TR 91-16 (Brandberg and Skagius, 1991), this report proposes a set of sorption distribution coefficient and diffusion coefficient values for modelling purpose for safety analysis of nuclear waste repositories. It applies for a dry density in the range 1600-2000 kg/m³. This set of data is summarized in Table 18.

Sorption mechanisms in compacted bentonite are discussed in the report. The main content of bentonite is the smectite clay mineral montmorillonite, which consists of stacks, containing elementary flakes, or platelets. The water in the interlayer space, or interlamellar space, is referred to as internal water, while the water in the voids between the stacks is termed external water. In highly compacted bentonite, the water content is mainly the internal water, which has a thickness of only a few angstroms, comparable to the thickness of electric double layer. It should be difficult to distinguish a solution phase and a sorbed phase in such a narrow space. Consequently, sorption and hence its distribution coefficient is not well-defined.

Based on the above argument, a pore diffusion coefficient or a surface diffusion coefficient is not well-defined either. Therefore, an apparent diffusion coefficient and a total concentration gradient should be more relevant in describing the diffusion process in compacted bentonite. This may provide another explanation to the discrepancy between the measured apparent diffusion coefficient and the value calculated based on the distribution coefficient.

Different methods for diffusion coefficient measurements are discussed in the report. Filters used in Method I and Method III could introduce errors to the measured parameters. In Method IIa, improper placing angle and shape of the tracer source plate could cause a positive bias to the measured concentration, resulting an error. When Method IIb is used, the source clay plug should be well mixed with the tracer to ensure a uniform initial concentration.

Sometimes it would be necessary to incorporate reactions in the radionuclide migration model, e.g., precipitation at high pH. In the diffusion experiments for Ni by Christiansen and Torstenfelt (1988), only part of the Ni was mobile, while the rest was immobile due to precipitation of the hydroxide, Ni(OH)₂. This kind of phenomena cannot be simply

represented by a distribution coefficient, because the precipitation has a discontinuity in the equilibrium relation. Solution phase complexation may change the speciation and thereby influence the sorption equilibrium.

Table 18. Proposed distribution coefficient and diffusion coefficient data for a dry compaction density range 1600-2000 kg/m³

Element	K _d (m ³ /kg)		D _a (m ² /s)	(D _e)(m ² /s)	Comments
	Realistic	Conservative	Realistic	D _a (ε+k _d ρ)	
Am	3	1	1·10 ⁻¹⁴	7·10 ⁻¹¹	
C*	0	0	5·10 ⁻¹²	1·10 ⁻¹²	
Ce	1	0.2	1·10 ⁻¹³	2·10 ⁻¹⁰	Similar to Am ε=0.05, ion exclusion
Cl*	0	0	2·10 ⁻¹¹	1·10 ⁻¹²	
Cs	0.5 (fresh)	0.2 (fresh)	5·10 ⁻¹²	6·10 ⁻⁹	ε=0.05, ion exclusion
	0.05 (saline)	0.005 (saline)	5·10 ⁻¹²	6·10 ⁻¹⁰	
I*	0	0	5·10 ⁻¹¹	3·10 ⁻¹²	
Nb	0.2	0	1·10 ⁻¹²	5·10 ⁻¹⁰	
Ni	0.1	0.02	5·10 ⁻¹²	1·10 ⁻⁹	
Np	0.02 (ox)	0.005 (ox)	1·10 ⁻¹²	5·10 ⁻¹¹	
	3 (red)	0.1 (red)	2·10 ⁻¹³	1·10 ⁻⁹	
Pa	3 (fresh)	0.1 (fresh)	1·10 ⁻¹²	7·10 ⁻⁹	
	0.3 (saline)	0.01 (saline)		7·10 ⁻¹⁰	
Pb	0.5	0	1·10 ⁻¹²	1·10 ⁻⁹	Extremely uncertain value
Pd	0.01	0	5·10 ⁻¹²	1·10 ⁻¹⁰	
Pu	3	1	5·10 ⁻¹⁴	3·10 ⁻¹⁰	
Ra	0.5 (fresh)	0.1 (fresh)	2·10 ⁻¹¹	2·10 ⁻⁸	Similar to Sr
	0.01 (saline)	0.001 (saline)	2·10 ⁻¹¹	5·10 ⁻¹⁰	
Rn	0	0	1·10 ⁻¹⁰	3·10 ⁻¹¹	
Se	0.003	0	1·10 ⁻¹¹	7·10 ⁻¹¹	
Sm	1	0.2	1·10 ⁻¹³	2·10 ⁻¹⁰	Similar to Am
Sn	3	0.01	1·10 ⁻¹⁴	7·10 ⁻¹¹	
Sr	0.5 (fresh)	0.1 (fresh)	2·10 ⁻¹¹	2·10 ⁻⁸	
	0.01 (saline)	0.001 (saline)	2·10 ⁻¹¹	5·10 ⁻¹⁰	
Tc*	0 (ox)	0 (ox)	2·10 ⁻¹²	1·10 ⁻¹³	ε=0.05, ion exclusion (ox)
	0.1 (red)	0.01 (red)	2·10 ⁻¹²	5·10 ⁻¹⁰	
Th	3	0.1	1·10 ⁻¹⁴	7·10 ⁻¹¹	
U	0.05 (ox)	0.005 (ox)	1·10 ⁻¹²	1·10 ⁻¹⁰	
	1 (red)	0.01 (red)	2·10 ⁻¹³	5·10 ⁻¹⁰	
Zr	2	0.2	1·10 ⁻¹⁴	5·10 ⁻¹¹	

* The D_e and ε values are realistic for a saline water. They can be expected to be even lower for a fresh water. To little experimental data is available to recommend the use of lower values.

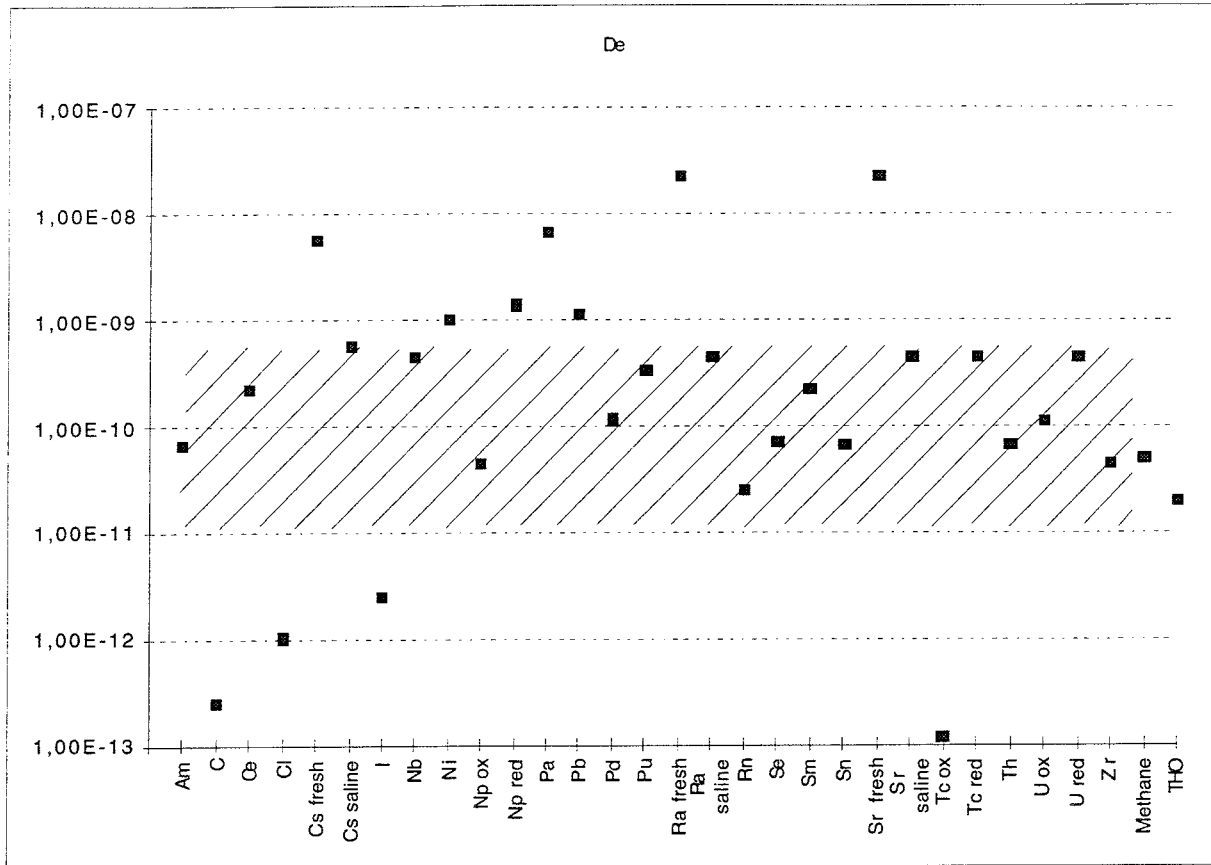


Figure 9. Effective diffusivities. Dotted lines at 10^{-9} and 10^{-11} indicate span where neither ion exclusion effects nor surface diffusion effects are important.

Figure 9 shows the effective diffusivity values D_e obtained from the D_a , K_D and ε values by equation (7).

If there were no ion exclusion or surface diffusion effects the D_e value could be estimated from the diffusivity of a species in bulk water, the tortuosity, constrictivity of the pores and porosity of the compacted bentonite and given by $D_e = D_w \varepsilon \frac{\delta}{\tau^2}$. The entity $\varepsilon \frac{\delta}{\tau^2}$ is on the order of $1/20$ for compacted bentonite with a dry density of 2000 kg/m^3 . As the diffusivity in bulk water D_w is on the order of $2 \cdot 10^{-9} \text{ m}^2/\text{s}$ for all the nuclides of interest D_e will be of order $10^{-10} \text{ m}^2/\text{s}$. Measurements with dissolved methane (Neretnieks and Skagius 1978) gave a D_e -value of $4 \cdot 10^{-11} \text{ m}^2/\text{s}$ for $50 \text{ }^\circ\text{C}$ and a dry density of 2150 kg/m^3 . Measurements with tritiated water gives values of around $2 \cdot 10^{-11} \text{ m}^2/\text{s}$, see Table (17). Both methane and tritiated water are uncharged molecules. These measured values and the estimate above from diffusivity in bulk water indicate that a D_e value near $10^{-10} \text{ m}^2/\text{s}$ is reasonable.

If there are significant ion exclusion effects it will be lower and if there are significant surface diffusion effects it will be higher.

The variability and uncertainty of the diffusivity data span somewhat more than an order of magnitude up and down. In Figure (9) this uncertainty band is shown by the dotted lines at D_e 10^{-9} m²/s and 10^{-11} m²/s. Values below or above this band indicate surface diffusion and ion exclusion effects respectively.

Ion exclusion effects are observed for C, Cl, I and for Tc in oxidizing waters. This is expected because these species exist as anions in the water compositions of interest.

Surface diffusion effects are found for Cs, Pa and Sr in low ionic strength waters. The surface diffusion effect should decrease in saline waters which is seen for Cs and Sr where there are data available. It is also deemed that Ra will have this effect because of its similarity with Sr. Because of insufficient data it cannot be ruled out that Ni and Pb also can migrate by surface diffusion.

For performance assessment, the following simplified approach could be used to obtain diffusion data in compacted bentonite with a dry density of 1600- 2000 kg/m³.

Realistic or best estimate values

In high ionic strength waters all cationic species are given a D_e of $1 \cdot 10^{-10}$ m²/s. In low ionic strength waters neutral and cationic species that sorb by other mechanisms than ion exchange have an effective diffusivity D_e of $1 \cdot 10^{-10}$ m²/s. Anions have 50 times *smaller* diffusivities due to ion exclusion. For anions use a bentonite porosity of 0.05 to obtain the apparent diffusion coefficient D_a from Equation (7). The cations Cs, Pa Ra, Sr have a 100 times *larger* diffusivities. Apparent diffusivities are obtained from Equation (7) with "realistic" sorption coefficients from Table 18. The uncertainty in the diffusion data are of the order of one order of magnitude up and down and this hardly warrants the use of more "accurate" numbers.

Conservative values

Increase D_e to $1 \cdot 10^{-9}$ m²/s. Add Ni and Pb to the cations Cs, Pa Ra, Sr and let them all have 10 times *larger* diffusivities. Apparent diffusivities are obtained from Equation (7) with "conservative" sorption coefficients from Table 18.

NOTATION

C	Total concentration in clay	mol/m^3
C_o	Boundary concentration	mol/m^3
C_p	Concentration in pore water in clay	mol/m^3
C_w	Concentration in water	mol/m^3
D_a	Apparent diffusion coefficient	m^2/s
D_e	Effective diffusion coefficient	m^2/s
D_p	Pore diffusion coefficient	m^2/s
D_s	Surface diffusion coefficient	m^2/s
D_w	Diffusion coefficient in bulk water	m^2/s
J	Flux of diffusing species	$\text{mol/m}^2\cdot\text{s}$
K_d	Sorption distribution coefficient	m^3/kg
L_f	Thickness of filter	m
M	Total amount of tracer per unit area	mol/m^2
q	Concentration in sorbed phase	mol/kg
t	Time	s
x	Distance	m
ε	Porosity	-
ε_f	Porosity of filter	-
δ	Constrictivity	-
ρ	Bulk clay density	kg/m^3
τ^2	Tortuosity	-

REFERENCES

- Abou-Jamous J Kh, 1992, Radioactive waste treatment using natural Syrian bentonite, *Journal of Radioanalytical and Nuclear Chemistry*, 162(2), 325-338.
- Albinsson Y, Andersson K, Börjesson S and Allard B, 1993, Diffusion of radionuclides in concrete/bentonite systems, Swedish Nuclear Fuel and Waste Management Co, SKB TR 93-29.
- Albinsson Y, Andersson K, Börjesson S and Allard B, 1996, Diffusion of radionuclides in concrete and concrete-bentonite systems, *Journal of Contaminant Hydrology*, 21, 189-200.
- Albinsson Y, Christiansen-Sätmark B, Engkvist I and Johansson W, 1991, Transport of actinides and Tc through a bentonite backfill containing small quantities of iron or copper, *Radiochimica Acta*, 52/53, 283-286.
- Albinsson Y and Engkvist I, 1989, Diffusion of Am, Pu, U, Np, Cs, I and Tc in compacted sand-bentonite mixture, Swedish Nuclear Fuel and Waste Management Co, SKB TR 89-22.
- Albinsson Y, Forsyth R, Skarnemark G, Skålberg M, Torstenfelt B and Werme L, 1990a, Leaching/migration of UO₂-fuel in compacted bentonite, In: Overby V M and Brown P W (eds.) *Materials Research Society Symp. Proc.*, Vol.176, Pittsburgh, Pennsylvania, 559-565.
- Albinsson Y, Sätmark B, Engkvist I and Johansson W, 1990b, Transport of actinides and Tc through a bentonite backfill containing small quantities of iron, copper or minerals in inert atmosphere, Swedish Nuclear Fuel and Waste Management Co, SKB TR 90-06.
- Allard B, Kipatsi H and Rydberg J, 1977, Sorption av långlivade radionuklider i lera och berg, Swedish Nuclear Fuel and Waste Management Co, KBS TR 55 (in Swedish).
- Allard B, Olofsson U, Torstenfelt B, Kipatsi H and Andersson K, 1982, Sorption of actinides in well-defined oxidation states on geological media, In: Lutze W (ed.), *Scientific Basis for Nuclear Waste Management*, Elsevier Science Publishers B V, Amsterdam.
- Allard B, Torstenfelt B and Andersson K, 1981, Sorption studies of HCO₃⁻ on some geologic media and concrete, In: More J G (ed.), *Scientific Basis for Nuclear Waste Management*, Plenum Publication Corporation, New York.
- Ames L L, McGarrah J E and Walker B A, 1983a, Sorption of trace constituents from aqueous solutions onto secondary minerals, I. Uranium, *Clays and Clay minerals*, 31, 321-334.
- Ames L L, McGarrah J E and Walker B A, 1983b, Sorption of trace constituents from aqueous solutions onto secondary minerals, II. Radium, *Clays and Clay minerals*, 31, 335-342.
- Ashida T, Kohara Y and Yui M, 1994, Migration behaviour of Pu released from Pu-doped glass in compacted bentonite, *Radiochimica Acta*, 66/67, 359-362.
- Assemi S and Erten H N, 1994, Sorption of radioiodine on organic rich soil, clay minerals and alumina, *Journal of Radioanalytical and Nuclear Chemistry*, 178(1), 193-204.

- Bertetti F P, Pabalan R T, Turner D R and Almendarez M G, 1996, Neptunium(V) sorption behavior on clinoptilolite, quartz and montmorillonite, In: Murphy W M and Knecht D A (eds.) Scientific Basis for Nuclear Waste Management XIX, Materials Research Society Symp. Proc. vol. 412, Pittsburgh, Pennsylvania, 631-638.
- Brandberg F and Skagius K, 1991, Porosity, sorption and diffusivity data compiled for the SKB 91 study, Swedish Nuclear Fuel and Waste Management Co, SKB TR 91-16.
- Bruno J and Sellin P, 1992, Radionuclide solubilities to be used in SKB 91, Swedish Nuclear Fuel and Waste Management Co, SKB TR 92-13.
- Carslaw H S and Jaeger J C, 1959, Conduction of Heat in Solids, Oxford University Press, Oxford, 70-71.
- Cheung S C H, Oscarson D W and Gray M N, 1986, Diffusion of Cs⁺ and I⁻ in compacted bentonite-based materials, Proceedings of the Canadian Nuclear Society 2nd International Conference on Radioactive Waste Management, Winnipeg, Man., 217-220.
- Cheung S C H, 1989, Methods to measure apparent diffusion coefficients in compacted bentonite clays and data interpretation, Can. J. Civ. Eng., 16, 434-443.
- Cheung S C H, 1990, A new interpretation of measured ionic diffusion coefficient in compacted bentonite-based materials, Engineering Geology, 28, 369-378.
- Cho W J, Oscarson D W, Gray M N and Cheung S C H, 1993, Influence of diffusant concentration on diffusion coefficients in clay, Radiochimica Acta, 60, 159-163.
- Choi J W, Jung C H, Chun C H, Park H S, Whang J H and Lee B H, 1992, Diffusion of radionuclides in compacted bentonite, Proceedings of Conference on High Level Radioactive Waste Management, La Grande Park, IL, American Nuclear Society, 2278-2283.
- Choi J-W and Oscarson D W, 1996, Diffusive transport through compacted Na- and Ca-bentonite, Journal of Contaminant Hydrology, 22, 189-202.
- Christiansen B and Torstenfelt B, 1988, Diffusion of nickel, strontium, iodine, cesium and americium in loosely compacted bentonite at high pH, Radiochimica Acta, 44/45, 219-223.
- Crank J, 1975, The mathematics of Diffusion, Oxford University Press, London.
- Eriksen T E, 1982, Diffusion of hydrogen, hydrogen sulfide and large molecular weight anions in bentonite, Swedish Nuclear Fuel Supply Co/Division KBS, SKBF/KBS TR 82-17.
- Eriksen T E and Jacobsson A, 1981, Ion diffusion in compacted sodium and calcium bentonites, Swedish Nuclear Fuel Supply Co/Division KBS, SKBF/KBS TR 81-12.
- Eriksen T E and Jacobsson A, 1984, Diffusion in clay - experimental techniques and theoretical models, Swedish Nuclear Fuel Supply Co/Division KBS, SKBF/KBS TR 84-05.
- Eriksen T E, Jacobsson A and Pusch R, 1981, Ion diffusion through highly compacted bentonite, Swedish Nuclear Fuel Supply Co/Division KBS, SKBF/KBS TR 81-06.
- Eriksen T E, Jansson M, 1996, Diffusion of I⁻, Cs⁺ and Sr²⁺ in compacted bentonite. Anion exclusion and surface diffusion. SKB TR 96-16, 1996.

Erten H N, Aksoyoglu S, Hatipoglu S and Göktürk H, 1988, Sorption of cesium and strontium on montmorillonite and kaolinite, *Radiochimica Acta*, 44/45, 147-151.

Gillham R W, Robin M J L and Dytyynyshyn D J, 1983, Diffusive transport of strontium-85 in sand-bentonite mixtures, Atomic Energy of Canada Limited, AECL-6838.

Gillham R W, Robin M J L, Dytyynyshyn D J and Johnson H, 1984, Diffusion of nonreactive and reactive solutes through fine-grained barrier materials, *Can. Geotech. J.*, 21, 541-550.

Goodwin F E, Mathew P M and Serne R J, 1995, North American studies on lead mobility in simulated nuclear waste repository environments, *Radioactive Waste Management and Environmental Restoration*, 19, 303-320.

Güngör N and Tulun T, 1994, Studies on the zeta potential of bentonite clay in aqueous solution, 39(2), 177-182.

Güven N, 1990, Longevity of bentonite as buffer material in a nuclear-waste repository, *Engineering Geology*, 28, 233-247.

Hakanen M and Hölttä P, 1992, Review of sorption and diffusion parameters for TVO-92, Nuclear Waste Commission of Finnish Power Companies, Report YJT-92-14.

Helfferich F, 1962, *Ion Exchange*, McGraw-Hill, New York.

Horyna J and Dlouhy Z, 1988, A study on the sorption properties of selected clays, *Jaderná energie*, 34, 300-302.

Hsu C-N and Chang K-P, 1994, Sorption and desorption behavior of cesium on soil components, *Applied Radiation Isotopes*, 45(4), 433-437.

Idemitsu K, Ishiguro K, Yusa Y, Sasaki N and Tsunoda N, 1990, Plutonium diffusivity in compacted bentonite, *Engineering Geology*, 28, 455-462.

Idemitsu K, Tachi Y, Furuya H, Inagaki Y and Arima T, 1996, Diffusion of uranium in compacted bentonites in the reducing condition with corrosion products of iron, In: Murphy W M and Knecht D A (eds.) *Scientific Basis for Nuclear Waste Management XIX*, Materials Research Society Symp. Proc. vol. 412, Pittsburgh, Pennsylvania, 683-690.

Ito K and Kanno T, 1988, Sorption behavior of carrier-free technetium-95m on minerals, rocks and backfill materials under both oxidizing and reducing conditions, *Journal of Nuclear Science and Technology*, 25(6), 534-539.

Kato H, Muroi M, Yamada N, Ishida H and Sato H, 1995, Estimation of effective diffusivity in compacted bentonite, Paper presented in XVIII International Symposium on the Scientific Basis for Nuclear Waste Management held in Kyoto, Japan 1994. Proceedings p 277-284

Kawaguchi M, Kaminoyama M, Takase H and Suzuki K, 1989, Study on cation adsorption model on sodium bentonite, In: Slate S C, Kohout R and Suzuki A (eds.) *Proceedings of the Joint Int. Waste Management Conference*, Vol. 2, Kyoto, Japan, American Society of Mechanical Engineers, New York, 313-317.

Khan, S A, Riaz-ur-Rehman and Khan M A, 1994, Sorption of cesium on bentonite, *Waste Management*, 14(7), 629-642.

Kim H-T, Suk T-W and Park S-H, 1993, Diffusivities for ions through compacted Na-bentonite with varying dry bulk density, *Waste Management*, 13, 303-308.

Konishi M, Yamamoto K, Yanagi T and Okajima Y, 1988, Sorption behavior of cesium, strontium and americium ions on clay materials, *Journal of Nuclear Science and Technology*, 25(12), 929-933.

Kozai N, Ohnuki T and Muraoka S, 1993, Sorption characteristics of neptunium by sodium-smectite, *Journal of Nuclear Science and Technology*, 30, 1153-1159.

Lehikoinen J, Carlsson T, Muurinen A, Olin M and Salonen P, 1996, Evaluation of factors affecting diffusion in compacted bentonite, In: Murphy W M and Knecht D A (eds.) *Scientific Basis for Nuclear Waste Management XIX*, Materials Research Society Symp. Proc. vol. 412, Pittsburgh, Pennsylvania, 675-682.

Liang T-J, Hsu C-N and Liou D-C, 1993, Modified Freundlich sorption of cesium and strontium on Wyoming bentonite, *Applied Radiation Isotopes*, 44(9), 1205-1208.

Miyahara K, Ashida T, Kohara Y, Yusa Y and Sasaki N, 1991, Effect of bulk density on diffusion for cesium in compacted sodium bentonite, *Radiochimica Acta*, 52/53, 293-297.

Morgan R D, Pryke D C and Rees J H, 1988, Data for the sorption of actinides on candidate materials for use in repository, Harwell Lab., UK, DOE-RW-87.094, AERA-R-12370, ILWRP-86-P11A.

Muurinen A, 1990, Diffusion of uranium in compacted sodium bentonite, *Engineering Geology*, 28, 359-367.

Muurinen A, 1992, Transport processes in the buffer and backfill of spent fuel disposal data for TVO-92 safety analysis, Nuclear Waste Commission of Finnish Power Companies, Report YJT-92-09.

Muurinen A, 1994, Diffusion of anions and cations in compacted sodium bentonite, Technical Research Centre of Finland, VTT Publications 168.

Muurinen A and Lehikoinen J, 1992, Diffusion of uranium in compacted sodium bentonite, Nuclear Waste Commission of Finnish Power Companies, Report YJT-92-13.

Muurinen A and Lehikoinen J, 1995, Evaluation of phenomena affecting diffusion of cations in compacted bentonite, Nuclear Waste Commission of Finnish Power Companies, Report YJT-95-05.

Muurinen A, Lehikoinen J and Pusch R, 1994, Literature study on the microstructure of bentonite and its effects on diffusion, Nuclear Waste Commission of Finnish Power Companies, Report YJT-94-22.

Muurinen A, Penttilä-Hiltunen P and Rantanen J, 1987, Diffusion mechanisms of strontium and cesium in compacted sodium bentonite, In: Bates J K and Seefeldt W B (eds.) *Scientific Basis for Nuclear Waste Management X*, Symp. Proc. vol. 84, Materials Research Society, Pittsburgh, Pennsylvania, 803-812.

Muurinen A, Penttilä-Hiltunen P and Uusheimo K, 1989, Diffusion of chloride and uranium in compacted sodium bentonite, In: Lutze W and Ewing R C (eds.) *Scientific Basis for Nuclear Waste Management XII*, Symp. Proc. vol. 127, Materials Research Society, Pittsburgh, Pennsylvania, 743-748.

Muurinen A, Rantanen J and Penttilä-Hiltunen P, 1985, Diffusion mechanisms of strontium, cesium and cobalt in compacted sodium bentonite, In: Werme L O (ed.)

Scientific Basis for Nuclear Waste Management X, Symp. Proc. vol. 50, Materials Research Society, Pittsburgh, Pennsylvania, 617-624.

Nagasaki S, Tanaka S and Suzuki A, 1994, Colloid formation and sorption of americium in the water/bentonite system, *Radiochimica Acta*, 66/67, 207-212.

Neretnieks I, 1982, Diffusivities of some dissolved constituents in compacted wet bentonite clay MX-80 and the impact on radionuclide migration in the buffer, Swedish Nuclear Fuel Supply Co/Division KBS, SKBF/KBS TR 82-27.

Neretnieks I and Skagius K, 1978, Diffusivitetmätningar av metan i våt lera, Swedish Nuclear Fuel and Waste Management Co, KBS TR 86 (in Swedish).

Nikiforov A S, Savushkina M K and Kosareva I M, 1991, Adsorption of caesium on bentonite under the conditions of thermal and radiation-thermal stresses, *Russian Journal of Physical Chemistry*, 65(8), 1171-1173.

Oscarson D W, 1994, Surface diffusion: Is it an important transport mechanism in compacted clays, *Clays and Clay Minerals*, 42, 534-543.

Oscarson D W, Hume H B and King F, 1994, Sorption of cesium on compacted bentonite, *Clays and Clay Minerals*, 42(6), 731-736.

Oscarson D W and Hume H B, 1994, Diffusion of ^{14}C in dense saturated bentonite under steady-state conditions, *Transport in Porous Media*, 14, 73-84.

Oscarson D W, Hume H B, Sawatsky N G and Cheung S C H, 1992, Diffusion of iodide in compacted bentonite, *Soil Sci. Soc. Am. J.*, 56, 1400-1406.

Oscarson D W, Watson R L and LeNeveu D M, 1984, Compilation of distribution coefficients for radioactive and other toxic contaminants with bentonite for use in SYVAC, AECL TR-288, Whiteshell Nuclear Research Establishment, Pinawa, Manitoba, Canada.

Oscarson D W, Watson R L and Miller H G, 1987, The interaction of trace levels of cesium with montmorillonitic and illitic clays, *Applied Clay Science*, 2, 29-39.

Papp T, Kjellbert N, Eng T, Nilsson S, Sellin P and Ström A, 1990, Arbetsbeskrivning för SKB 91, Swedish Nuclear Fuel and Waste Management Co, SKB AR 90-26.

Pratopo M I, Yamaguchi T, Moriyama H and Higashi K, 1993, Sorption and colloidal behavior of Np(IV) in a bentonite-carbonate solution system, *Journal of Nuclear Science and Technology*, 30(6), 560-566.

Puigdomènech I and Bergström U, 1994, Calculated distribution of radionuclides in soils and sediments, Swedish Nuclear Fuel and Waste Management Co, SKB TR 94-32.

Pusch R and Hökmark H, 1990, Basic model of water- and gas-flow through smectite clay buffers, *Engineering Geology*, 28, 379-389.

Pusch R, 1992, Use of bentonite for isolation of radioactive waste products, *Clay Minerals*, 27, 353-361.

Pusch R, Karnland O and Hökmark H, 1990, GMM - a general microstructural model for qualitative and quantitative studies of smectite clays, Swedish Nuclear Fuel and Waste Management Co, SKB TR 90-43.

Pusch R, Karnland O and Muurinen A, 1989, Transport and microstructural phenomena in bentonite clay with respect to the behavior and influence of Na, Cu, and U, Swedish Nuclear Fuel and Waste Management Co, SKB TR 89-34.

Ramebäck H, Albinsson Y, Skålberg M and Werme L, 1994, Release and diffusion of ^{90}Sr from spent UO_2 fuel in bentonite clay, *Radiochimica Acta*, 66/67, 405-408.

Rasmuson A and Neretnieks I, 1983, Surface migration in sorption processes, Swedish Nuclear Fuel Supply Co/Division KBS, SKBF/KBS TR 83-37.

Relyea J F, Trott D P, McIntyre C V and Rieger C G, 1986, Diffusion of tritiated water and chloride in basalt-bentonite mixtures, *Nuclear Technology*, 74, 317-323.

Robin M J L, Gillham R W and Oscarson D W, 1987, The diffusion of Sr and Cl in compacted clay-based materials, *Soil Science of America Journal*, 5, 1102-1107.

Sato H, Ashida T, Kohara Y, Yui M and Sasaki N, 1992, Effect of dry density on diffusion of some radionuclides in compacted sodium bentonite, *Journal of Nuclear Science and Technology*, 29(9), 873-882.

Sato H, Yui M, Hideki Y, 1995, Diffusion behavior for Se and Zr in Sodium-bentonite, Paper presented in XVIII International Symposium on the Scientific Basis for Nuclear Waste Management held in Kyoto, Japan 1994. Proceedings p 269-276

Sawatsky N G and Oscarson D W, 1991a, Diffusion of technetium in dense bentonite, *Water, Air, and Soil Pollution*, 57-58, 449-456.

Sawatsky N G and Oscarson D W, 1991b, Diffusion of technetium in dense bentonite under oxidizing and reducing conditions, *Soil Sci. Soc. Am. J.*, 55, 1261-1267.

Torstenfelt B, 1986a, Migration of the fission products strontium, technetium, iodine and cesium in clay, *Radiochimica Acta*, 39, 97-104.

Torstenfelt B, 1986b, Migration of the actinides thorium, protactinium, uranium, neptunium and americium in clay, *Radiochimica Acta*, 39, 105-112.

Torstenfelt B and Allard B, 1986, Migration of fission products and actinides in compacted bentonite, Swedish Nuclear Fuel and Waste Management Co, SKB TR 86-14.

Torstenfelt B, Allard B, Andersson K, Kipatsi H, Eliasson L, Olofsson U and Persson H, 1983, Radionuclide diffusion and mobilities in compacted bentonite, Swedish Nuclear Fuel and Waste Management Co, KBS TR 83-34.

Torstenfelt B, Kipatsi H, Andersson K, Allard B and Olofsson U, 1982, Transport of actinides through a bentonite backfill, In: Lutze W (ed.), *Scientific Basis for Nuclear Waste Management*, Elsevier Science Publishers B V, Amsterdam.

Tsukamoto M, Ohe T and Fujita T, 1994, Diffusion of neptunium(V) in loosely compacted sodium bentonite, *Radiochimica Acta*, 66/67, 397-403.

Uusheimo K, Muurinen A, Lehtikainen J and Olin M, 1993, Diffusion in concrete, crushed rock and mixtures of crushed rock and bentonite, Nuclear Waste Commission of Finnish Power Companies, Report YJT-93-26.

Wanner H, Albinsson Y and Wieland E, 1994, An ion exchange model for the prediction of distribution coefficients of cesium in bentonite, Swedish Nuclear Fuel and Waste Management Co, SKB TR 94-10.

Wanner H, Wersin P and Sierro N, 1992, Thermodynamic modelling of bentonite-groundwater interaction and implications for near field chemistry in a repository for spent fuel, Swedish Nuclear Fuel and Waste Management Co, SKB TR 92-37.

Wiborgh M, 1995, Prestudy of final disposal of long-lived low and intermediate level waste, Swedish Nuclear Fuel and Waste Management Co, SKB TR 95-03.

List of SKB reports

Annual Reports

1977-78

TR 121

KBS Technical Reports 1 – 120

Summaries

Stockholm, May 1979

1979

TR 79-28

The KBS Annual Report 1979

KBS Technical Reports 79-01 – 79-27

Summaries

Stockholm, March 1980

1980

TR 80-26

The KBS Annual Report 1980

KBS Technical Reports 80-01 – 80-25

Summaries

Stockholm, March 1981

1981

TR 81-17

The KBS Annual Report 1981

KBS Technical Reports 81-01 – 81-16

Summaries

Stockholm, April 1982

1982

TR 82-28

The KBS Annual Report 1982

KBS Technical Reports 82-01 – 82-27

Summaries

Stockholm, July 1983

1983

TR 83-77

The KBS Annual Report 1983

KBS Technical Reports 83-01 – 83-76

Summaries

Stockholm, June 1984

1984

TR 85-01

Annual Research and Development Report 1984

Including Summaries of Technical Reports Issued during 1984. (Technical Reports 84-01 – 84-19)

Stockholm, June 1985

1985

TR 85-20

Annual Research and Development Report 1985

Including Summaries of Technical Reports Issued during 1985. (Technical Reports 85-01 – 85-19)

Stockholm, May 1986

1986

TR 86-31

SKB Annual Report 1986

Including Summaries of Technical Reports Issued during 1986

Stockholm, May 1987

1987

TR 87-33

SKB Annual Report 1987

Including Summaries of Technical Reports Issued during 1987

Stockholm, May 1988

1988

TR 88-32

SKB Annual Report 1988

Including Summaries of Technical Reports Issued during 1988

Stockholm, May 1989

1989

TR 89-40

SKB Annual Report 1989

Including Summaries of Technical Reports Issued during 1989

Stockholm, May 1990

1990

TR 90-46

SKB Annual Report 1990

Including Summaries of Technical Reports Issued during 1990

Stockholm, May 1991

1991

TR 91-64

SKB Annual Report 1991

Including Summaries of Technical Reports Issued during 1991

Stockholm, April 1992

1992

TR 92-46

SKB Annual Report 1992

Including Summaries of Technical Reports Issued during 1992

Stockholm, May 1993

1993

TR 93-34

SKB Annual Report 1993

Including Summaries of Technical Reports Issued during 1993

Stockholm, May 1994

1994

TR 94-33

SKB Annual Report 1994

Including Summaries of Technical Reports Issued during 1994

Stockholm, May 1995

1995

TR 95-37

SKB Annual Report 1995

Including Summaries of Technical Reports Issued during 1995

Stockholm, May 1996

1996

TR 96-25

SKB Annual Report 1996

Including Summaries of Technical Reports Issued during 1996

Stockholm, May 1997

List of SKB Technical Reports 1997

TR 97-01

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Mark Elert

Kemakta Konsult AB

February 1997

TR 97-02

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Roy Stanfors¹, Mikael Erlström²,

Ingemar Markström³

¹ RS Consulting, Lund

² SGU, Lund

³ Sydkraft Konsult, Malmö

March 1997

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Ingvar Rhén (ed.)¹, Göran Bäckblom (ed.)², Gunnar Gustafson³, Roy Stanfors⁴, Peter Wikberg²

¹ VBB Viak, Göteborg

² SKB, Stockholm

³ VBB Viak/CTH, Göteborg

⁴ RS Consulting, Lund

April 1997

TR 97-04

Äspö HRL – Geoscientific evaluation 1997/3. Results from pre-investigations and detailed site characterization. Comparison of predictions and observations. Geology and mechanical stability

Roy Stanfors¹, Pär Olsson², Håkan Stille³

¹ RS Consulting, Lund

² Skanska, Stockholm

³ KTH, Stockholm

May 1997

TR 97-05

Äspö HRL – Geoscientific evaluation 1997/4. Results from pre-investigations and detailed site characterization. Comparison of predictions and observations. Geohydrology, groundwater chemistry and transport of solutes

Ingvar Rhén¹, Gunnar Gustafson², Peter Wikberg³

¹ VBB Viak, Göteborg

² VBB Viak/CTH, Göteborg

³ SKB, Stockholm

April 1997

TR 97-06

Äspö HRL – Geoscientific evaluation 1997/5. Models based on site characterization 1986–1995

Ingvar Rhén (ed.)¹, Gunnar Gustafson²,

Roy Stanfors⁴, Peter Wikberg⁴

¹ VBB Viak, Göteborg

² VBB Viak/CTH, Göteborg

³ RS Consulting, Lund

⁴ SKB, Stockholm

May 1997

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Golder Associates Inc.

March 1997

TR 97-08

Äspö Hard Rock Laboratory Annual Report 1996

SKB

April 1997

TR 97-09

A regional analysis of groundwater flow and salinity distribution in the Äspö area

Urban Svensson

Computer-aided Fluid Engineering AB

May 1997

TR 97-10

On the flow of groundwater in closed tunnels. Generic hydrogeological modelling of nuclear waste repository, SFL 3-5

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Uppsala University/Golder Associates AB

June 1997

TR 97-11

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Forsyth Consulting

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