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Migration of the Fission Products Strontium, Technetium, Iodine and Cesium in Clay

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Bentonite clay / Diffusion / Fission products / Laboratory measurements / Migration

Abstract

The migration in compacted bentonite i. e., the diffusivity, of the fission products strontium, technetium, iodine and cesium have been studied in laboratory experiments.

The clay used in the experiments was a sodium bentonite, Wyoming Bentonite MX-80, compacted to a density of 2000 kg/m³. The aqueous phase was synthetic groundwater representative of Swedish deep granitic groundwaters, and was preequilibrated with the clay. The influence of complex-forming and redox controlling agents was studied by mixing the clay with small amounts of the chemical reagents PbO, $KMnO_4$ or powdered iron, or the minerals chalcopyrite / pyrite or cinnabar.

The diffusivity of strontium was on the order of 10^{-11} m²/s. For cesium and technetium (as pertechnetate) under oxidizing conditions the diffusivities are on the order of 10^{-12} m²/s; for technetium under reducing conditions the diffusivity is about one order of magnitude lower. Iodine appears to diffuse with two mechanisms, resulting in two diffusion rates – one close to 10^{-12} m²/s and the other one-tenth slower, 10^{-13} m²/s, contributing to the observed overall apparent diffusivity.

1. Introduction

Because clay has been proposed as a suitable host material and / or backfill material for the storage of radioactive waste, there has been an increasing need to examine transport rates of different hazardous waste elements in clay. As a consequence of this, a number of papers and reports have recently been published concerning both experimental measurements and theoretical evaluations of diffusion properties in clay [1–11].

For a thorough understanding of the behaviour of solutes in clay, the chemical composition of the pore water in the clay and the speciation of the diffusing species need to be known. This paper presents direct measurements of the diffusion of strontium, technetium, iodine and cesium in a bentonite, compacted to a density of 2000 kg/m³ and saturated with a synthetic groundwater which was preequilibrated with the clay. The paper is largely based on previously published conference proceedings and a technical report [12–15], but the diffusion coefficients have been reexamined.

2. Experimental

2.1. Radionuclides

Among the fission products in spent nuclear fuel, nuclides of interest to study are strontium, technetium, iodine and cesium.

Technetium and iodine both have long-lived isotopes – technetium-99 (2.1×10^5 y) and iodine-129 (1.57×10^7 y) – and both are anionic under oxidizing condition; thus, they are expected to have only minor interactions with the solid phase and a mobility in the water-saturated clay close to the self-diffusion of ions in dilute water solutions. The isotopes used in the experiments were ^{99}Tc and ^{125}I with a total amount of approximately 10^{-6} and 10^{-13} mol added, respectively.

Strontium and cesium are the major contributors to the radiation hazards of spent nuclear fuel during the first 300 years after discharge from the reactor. They are also suitable model elements in studying the behavior of uncomplexed di- and monovalent cations. Approximately 10^{-12} mol of ^{85}Sr and ^{134}Cs were used in all experiments.

2.2. Solids and the aqueous phase

A complete list of the studied systems is given in Table 1.

Sodium bentonite (MX-80, Wyoming bentonite), compacted to a density of 2000 kg/m³ and preequilibrated with simulated groundwater, was used. The preequilibration was accomplished by first mixing the clay with the water and then separating the two phases by high-speed centrifugation (~ 27000 g) after a contact time of more

Table 1. Studied systems (aqueous phase: artificial groundwater preequilibrated with the clay, cf., Table 2)

Diffusing species	Solid phase
^{85}Sr	Bentonite [a]
^{99}Tc	Bentonite
^{99}Tc	Bentonite + 0.5% Fe(s)
^{99}Tc	Bentonite / Bentonite + 0.5% Fe(s) [b]
^{125}I	Bentonite
^{125}I	Bentonite + 0.5% Fe(s)
^{125}I	Bentonite + 1% Fe(s)
^{125}I	Bentonite + 1% $KMnO_4$
^{125}I	Bentonite + 0.5% Fe(s) + 1% Chalcopyrite / pyrite
^{125}I	Bentonite + 0.5% Fe(s) + 1% Cinnabar
^{125}I	Bentonite + 0.5% Fe(s) + 1% PbO
^{125}I	Bentonite + 1% Fe(s) / Bentonite + 1% $KMnO_4$ [b]
^{125}I	Bentonite / Bentonite + 1% $KMnO_4$ [b]
^{134}Cs	Bentonite

[a] Wyoming bentonite, MX-80; density: 2000 kg/m³.
[b] One type in each half-cell.

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than one week. The clay was then dried in an oven (~ 105 °C), finely ground in an agate mortar, then used for the investigations.

The possibility of controlling the ion mobility in the clay was examined by mixing different additives, i. e., "getters", with the preequilibrated clay. Chemical interactors (complex-forming agents) were added in the form of finely ground minerals (chalcopyrite/pyrite, cinnabar) or chemical reagents (PbO), and in some experiments reducing conditions or oxidizing conditions were achieved by adding iron powder or potassium permanganate, respectively.

The aqueous phase was originally an artificial groundwater representative of Swedish deep granitic groundwaters [16] but was preequilibrated with the clay (see above, this section). This caused the resulting pore water, consisting mainly of the cations sodium and potassium and the anions sulphate, chloride and nitrate to have a slightly higher ionic strength (cf., Table 2).

Table 2. Composition of the aqueous phase (pH 8.8 – 9.0)

Cation	ppm	Anion	ppm
K^+	11	Cl^-	132
Na^+	670	NO_3^-	6.8
Ca^{2+}	6.6	SO_4^{2-}	870
Mg^{2+}	1.7		

2.3. Diffusion measurements

The technique adopted for studying the diffusion was developed at the Department of Nuclear Chemistry, Chalmers University of Technology, Göteborg, and is described in detail in reference [17]. The main principle of the experimental technique is that the studied species is introduced into a diffusion cell as a thin layer placed radially in the middle of a cylindrical clay sample (Fig. 1). The entire diffusion cell is submerged in water and the species in the thin layer diffuses axially into the clay; thus, with time, a concentration profile moves slowly from the center out towards both ends. After a suitable diffusion time (i. e., yielding measurable concentration profiles) the clay is sliced in thin sections and the radioactivity of the diffusing species analysed.

Prior to the introduction of the diffusing species, the clay is wetted and homogenized with the preequilibrated water. The time allowed for the homogenization is about one month in order to minimize chemical concentration profiles in the water and density variations in the solid.

The radioactive species is introduced into the clay by first making a clay slurry, then taking drops of this slurry and drying them on a glass plate. When the drops are completely dry, the clay residue makes a thin (~ 0.2 – 0.5 mm) hard plate which is fairly easy to remove from the glass. On this plate the solution containing the radioactive species is dropped and evaporated to dryness under an IR-lamp. The radioactive sample is then placed on a wet, compacted, preequilibrated clay cylinder in the diffusion cell and another wet compacted clay cylinder is pressed over it (cf., Fig. 1).

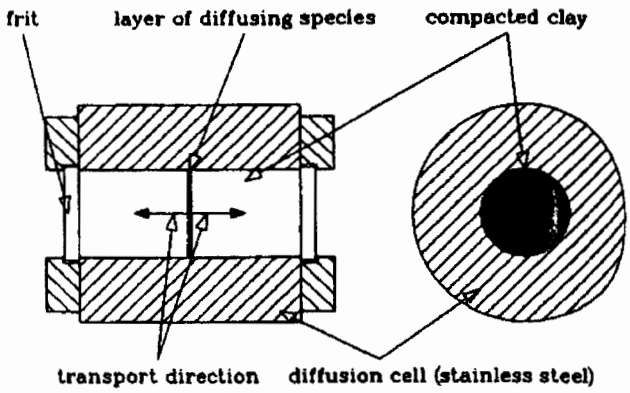

Fig. 1. Schematic view of the diffusion cell (cf., ref. 17)

After the necessary diffusion time, the diffusion cell is opened and the mantle surface of the sample removed to avoid measuring any surface diffusion along the cell wall. While the sample is still wet, it is sliced with a knife, and the concentration profile is analysed with an appropriate radioactive counter.

3. Diffusion theory

Flow of water through the clay is not possible; thus, the transport rate of a species through the wet clay is dependent only on diffusion. Diffusion through a porous solid is dependent on molecular diffusion in the aqueous phase, on sorption phenomena and possibly surface transport on the solid, and also on the pore constrictivity and tortuosity. Therefore the measured diffusivity is an apparent diffusivity, D_a, and not a pure molecular diffusivity, D.

The apparent diffusivity for one-dimensional diffusion is given by the equation [18, 19]:

$$\frac{dC}{dt} = \frac{\partial}{\partial x}\left(D_a \frac{dC}{dx}\right). \tag{1}$$

For the one-dimensional diffusion of a plane source consisting of a limited amount of substance in a cylinder of infinite length and assuming diffusion independent of concentration, the solution of Eqn. (1) is:

$$\frac{C}{M} = \frac{0.5}{\sqrt{\pi D_a t}} \times e^{\frac{-x^2}{4D_a t}} \tag{2}$$

where C = concentration (moles/m^3), M = total amount of diffusing substance added per unit area (moles/m^2), x = distance from source (m), D_a = apparent diffusivity (m^2/s), and t = time (s).

If the sorption on the solid is reversible, with the sorption defined by the distribution coefficient, K_d (moles per kg solid/moles per m^3 liquid, m^3/kg), the relation

Table 3. Distribution coefficients [20–22] and measured diffusivities in bentonite (density of the clay = 2000 kg/m³)

Element	$\log C_i$[a]	Time (d)	K_d[b] (m³/kg)	D_a (m²/s)	D[c] (m²/s)
Sr	−12.3	50	2.9	2.0×10^{-11}	–
Tc	− 5.8	69	0.0	1.2×10^{-12}	–
Tc[d]	− 5.8	70	0.05	1.5×10^{-13}	3.5×10^{-11}
Tc[e]	− 5.8	218	0.05	8.4×10^{-14} (Fe) 6.7×10^{-13} (no Fe)	2.0×10^{-11}
Cs	−11.4	53	1.4	2.4×10^{-12}	–

a C_i = Number of moles initially added.
b For a total nuclide concentration of 10^{-9} M.
c Evaluated from Eq. (3).
d 0.5% Fe(s) in the clay.
e 0.5% Fe(s) in part of the clay.

between the apparent diffusivity (D_a) and the diffusivity not affected by sorption (D) is:

$$D = D_a(1 + K_d \rho(1-\epsilon)/\epsilon) \qquad (3)$$

where ρ = density of the solid (kg/m³) and ϵ = porosity of the solid (m³ void fraction/m³ total volume).

If the sorption is concentration-dependent, both the distribution coefficient and the apparent diffusivity will vary with the concentration and Eqn. (2) will not be valid. In this case the sorption isotherm must be known and Eqn. (1) solved by numerical methods.

4. Results and discussion

Equation (2) can be rewritten as $C = 0.5M(\pi D_a t)^{-0.5} \times e^{\frac{-x^2}{4D_a t}}$. Taking its logarithm yields $\log C = \text{const.} - (1/(4D_a t))\,x^2$, which, plotted as $\log C$ versus x^2, is a straight line with the slope giving the diffusivity. The measured concentration profiles, given as $\log C$ versus x^2, for strontium, technetium, cesium and iodine are shown in Figures 2–13[a]. Experimental parameters and calculated diffusivities are given in Tables 3 and 4.

4.1. Strontium

The apparent diffusivity of strontium (as Sr^{2+}) was measured to be 2.0×10^{-11} m²/s (Fig. 2, Table 3). Considering the high sorption of strontium (cf., Table 3), this indicates an unexpectedly high mobility in the clay. In fact, the diffusivity calculated from equation (3) is two orders of magnitude higher than self-diffusion in pure water which is, of course, not possible. Thus, either the measured batch-K_d is not applicable in the diffusion experiments or equation (3) is not valid for cation exchange sorption processes where the sorption/desorption rate is much faster than the diffusion process. The ionic strength of the pore water in the diffusion experiments is slightly higher than in the batch experiments,

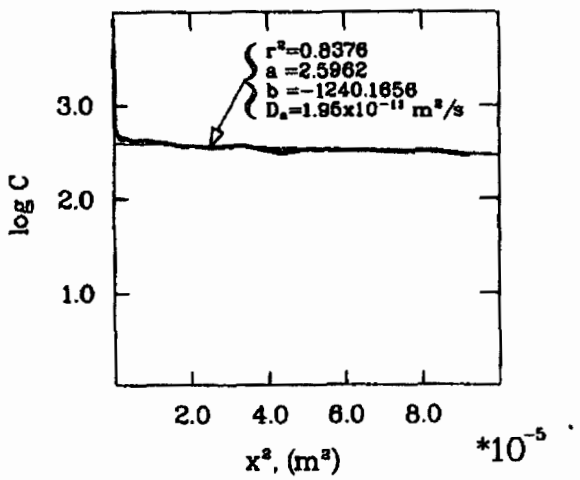

Fig. 2. Diffusion of strontium in compacted bentonite. Diffusion time = 50 days

thus, giving a lower K_d. It is probably not possible, however, to use Eq. (3) for species sorbing by a cation exchange mechanism.

4.2. Technetium

Technetium was measured under both oxidizing (Fig. 3a) and reducing (Fig. 3b) conditions and also when only part of the cell was filled with clay mixed with powdered iron (producing both reducing and oxidizing regions (Fig. 4). Under oxidizing conditions technetium is heptavalent and forms the anion pertechnetate, TcO_4^-, which is non-sorbing in bentonite. Without sorption, the apparent diffusivity (1.2×10^{-12} m²/s) is dependent only on the physical transport in the aqueous phase. Under reducing conditions, the apparent diffusivity was almost one order of magnitude lower (1.5×10^{-13} m²/s). An even lower apparent diffusivity (8.4×10^{-14} m²/s) was observed in the ex-

a The values for r^2, a and b from the least square fit of $\log C = a + b \times x^2$ and the apparent diffusivity calculated from the b-value are given.

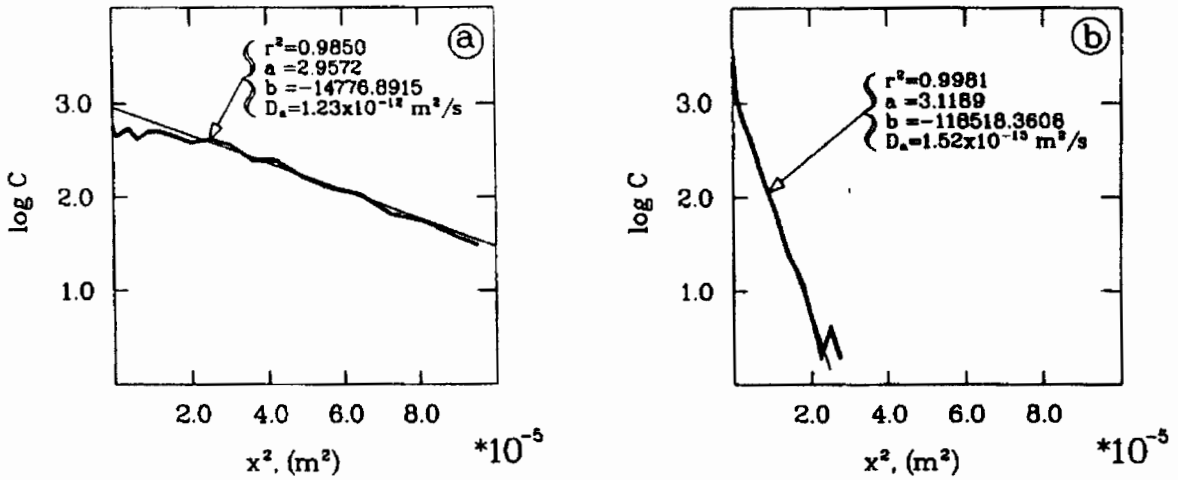

Fig. 3. Diffusion of technetium in compacted bentonite. a) Ordinary bentonite, diffusion time = 69 days. b) Bentonite mixed with 0.5% iron powder, diffusion time = 70 days

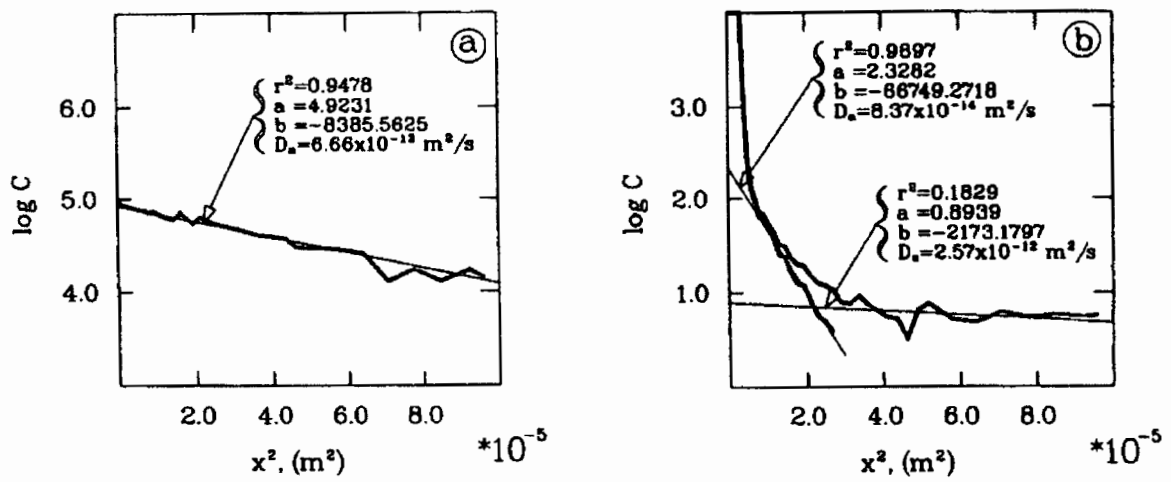

Fig. 4. Diffusion of technetium in compacted bentonite with one half of the cell filled with ordinary clay a), and the other half of the cell filled with clay mixed with 0.5% iron powder b). Diffusion time = 218 days

periment with different conditions in the two half-cells. Obviously, the non-sorbing pertechnetate must be reduced by the iron to the tetravalent state, which forms sorbing species, the hydroxide ($Tc(OH)_4$) or the oxide (TcO_2) (cf., Table 3) [20]. The fact that the diffusivity is lower in the mixed half-cells is probably due to the much longer diffusion time, which provides sufficient time for the reduction (of greater amounts) of pertechnetate.

The sorption mechanism for technetium – physical adsorption of the oxide/hydroxide – is different from that of strontium. The diffusivity of technetium, evaluated from equation (3), is 2×10^{-11} m^2/s which indicates that equation (3) is more feasible to use for technetium than for strontium. With a K_d of 0.01, a reasonable value considering the ionic strength of the pore water, the diffusivity is 3.9×10^{-12} m^2/s which is close to the 1.2×10^{-12} m^2/s measured for the pertechnetate.

In Figure 4b it can be seen that a fraction of the technetium has diffused with the same rate as the pertechnetate. Because almost the same diffusivity is found for iodine and some of the actinides [24]; this value appears to be the diffusivity achieved for non-interactive transport through the clay.

4.3. Cesium

Cesium, an uncomplexed monovalent cation, sorbs by cation exchange and has an apparent diffusivity of 2.4×10^{-12} m^2/s (Fig. 5 and Table 3). This is the same diffusivity as those of the non-sorbing elements. Thus, as for strontium, the high distribution coefficient does not seem to have any retarding influence on the transport rate; in fact, it seems to be almost the contrary, at least for strontium.

Table 4. Distribution coefficients [23] and measured diffusivities for iodine in bentonite (init. amount of iodine = 1.8×10^{-13} moles; density = 2000 kg/m³).

Additive	Time (d)	K_d^a (m³/kg)	$(D_a)_{fast}$ (m²/s)	$(D_a)_{slow}$ (m²/s)	D^b (m²/s)
–	157	0.001	2.6×10^{-12}	2.4×10^{-13}	1.4×10^{-12}
–	218	0.001	1.2×10^{-12}	1.1×10^{-13}	6.2×10^{-13}
0.5% Fe(s)	194	0.001	1.9×10^{-12}	1.7×10^{-13}	7.4×10^{-13}
1% Fe(s)	213	0.001	1.2×10^{-12}	7.2×10^{-14}	4.1×10^{-13}
1% $KMnO_4$	210	0.001	1.3×10^{-12}	1.3×10^{-13}	7.4×10^{-13}
1% $KMnO_4$ [c]	213		1.1×10^{-12}	7.5×10^{-14} ($KMnO_4$)	
			1.5×10^{-12}	1.5×10^{-13} (no $KMnO_4$)	
1% $KMnO_4$/1% Fe[d]	225		1.4×10^{-12}	8.0×10^{-14} ($KMnO_4$)	
			1.1×10^{-12}	5.3×10^{-14} (Fe(s))	
0.5% Fe + 1% Chalcopyrite	225	0.001	1.0×10^{-12}	9.6×10^{-14}	5.4×10^{-12}
0.5% Fe + 1% Cinnabar	226	0.8	1.4×10^{-12}	2.6×10^{-14}	1.5×10^{-12}
0.5% Fe + 1% PbO	224	0.6	1.3×10^{-12}	4.4×10^{-14}	2.5×10^{-12}

[a] For a total nuclide concentration of 10^{-9} *M*.
[b] Evaluated from Eq. (3) using $(D_a)_{slow}$.
[c] In half of the clay.
[d] Each in separate halfs of the clay.

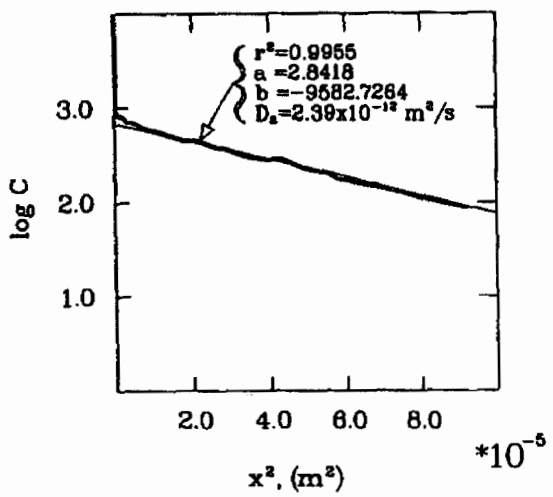

Fig. 5. Diffusion of cesium in compacted bentonite. Diffusion time = 53 days

4.4. Iodine

The diffusion of iodine is dependent on two (or more) different mechanisms or on two diffusing species (cf., Fig. 6). Figure 6 shows that by subtracting the least square-fitted line of the "tail" of the curve from the measured curve, a new least square fit can be performed on the remaining curve. This indicates that the measured diffusion consists of two (or maybe more [a]) mechanisms, each satisfying equation (2) and each with a distinct diffusivity. The apparent diffusivities measured for iodine, as iodide (I^-), are 1.2 to 2.6×10^{-12} m²/s and 1.1 to

[a] It is often possible to fit a third line to the data close to the starting point; however, this is probably just a consequence of the measuring technique. Because a small difference in the radial density of the clay close to the added radioactive layer could flip the layer axially, an artificial concentration profile could result at the starting point.

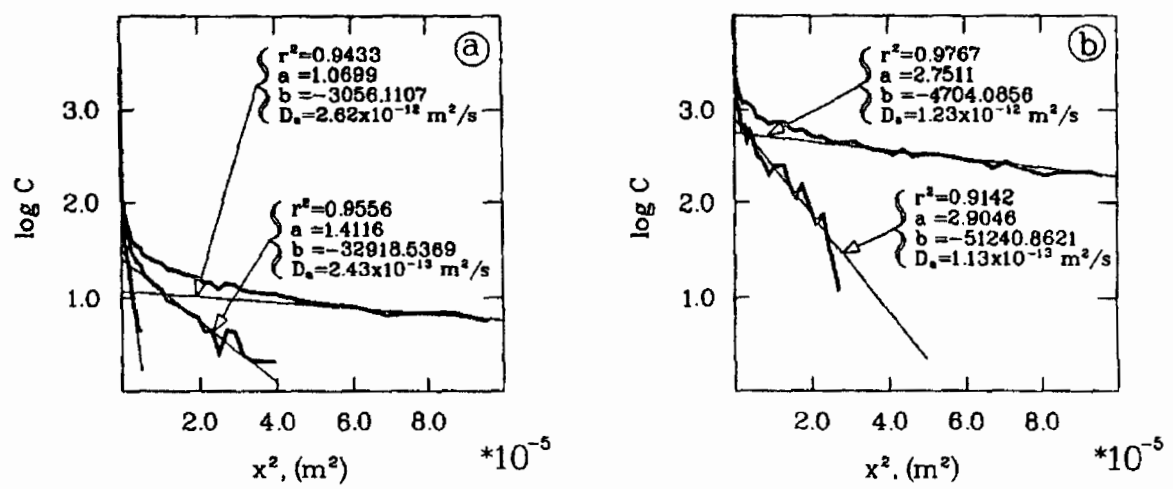

Fig. 6. Diffusion of iodine in compacted bentonite. a) Diffusion time = 157 days. b) Diffusion time = 218 days

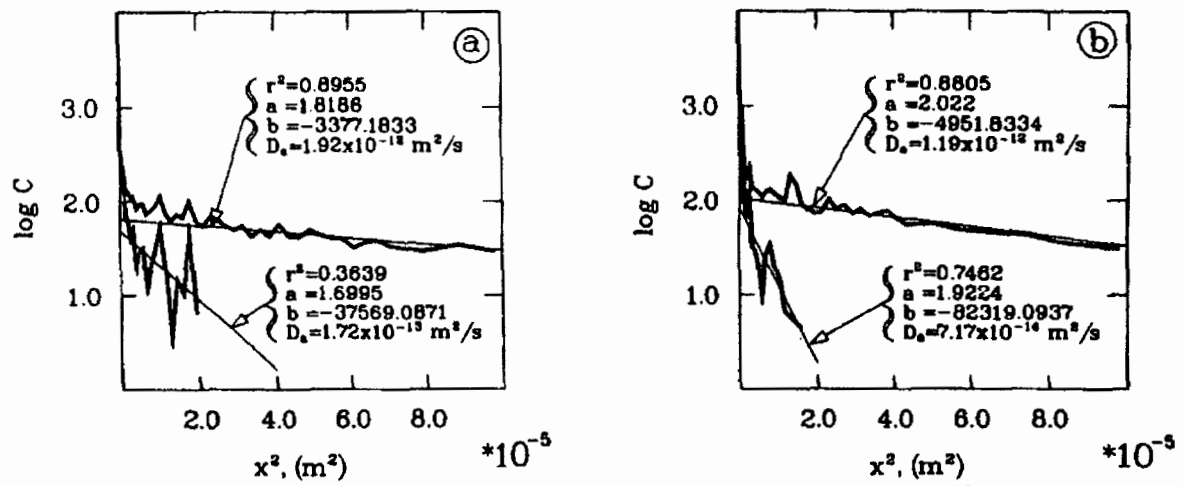

Fig. 7. Diffusion of iodine in compacted bentonite. a) Bentonite mixed with 0.5% iron powder, diffusion time = 194 days. b) Bentonite mixed with 1% iron powder, diffusion time = 213 days

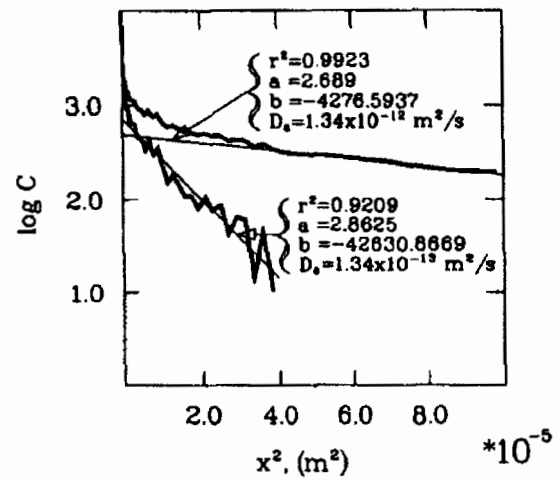

Fig. 8. Diffusion of iodine in compacted bentonite mixed with 1% $KMnO_4$. Diffusion time = 210 days

2.4×10^{-13} m^2/s. The two overlapping diffusivities are expected to be caused by either steric effects and dead end pores or by two different forms of iodine–iodide (I^-) and possibly hypoiodous acid (HIO, which is possibly stable under the chemical conditions in the clay [25]). The presence of two overlapping diffusivities is not an uncommon phenomenon. For the actinides especially, which can commonly exist in more than one chemical form simultaneously, two separate diffusivities contributing to the overall apparent diffusivity is often observed [24].

By thinking of the clay as a two channel system – one large channel representing interconnected pores through which the main fraction of the iodide is transported and one small channel representing the dead-end pores and pores where the constrictivity severely hinders the diffusion – the idea that steric effects could lead to two observed diffusion mechanisms can be explained. This, too, would result in the slow transport of a more or less minor fraction of the iodine [26].

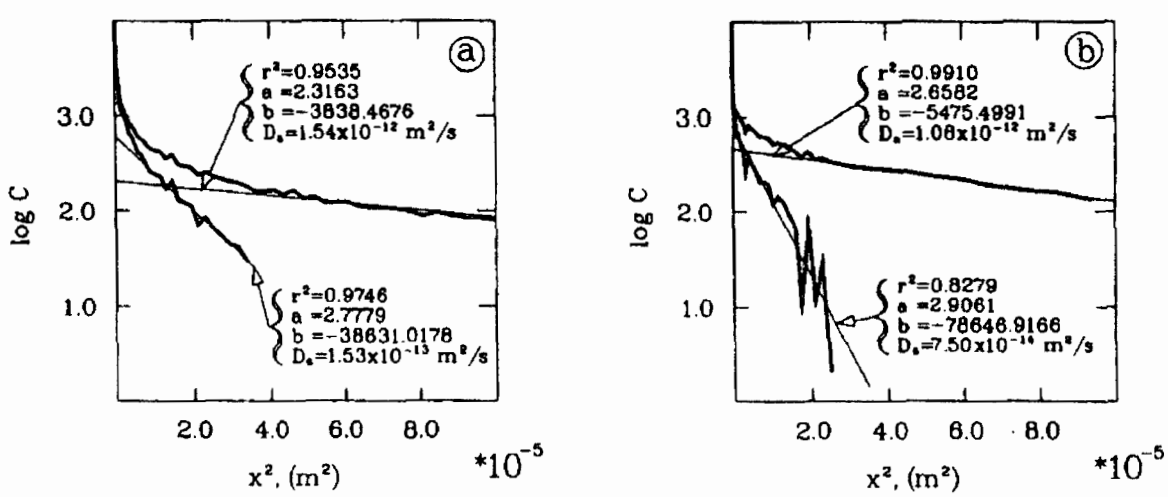

Fig. 9. Diffusion of iodine in compacted bentonite with half of the cell filled with ordinary clay a), and the other half of the cell filled with clay mixed with 1% $KMnO_4$ b). Diffusion time = 213 days

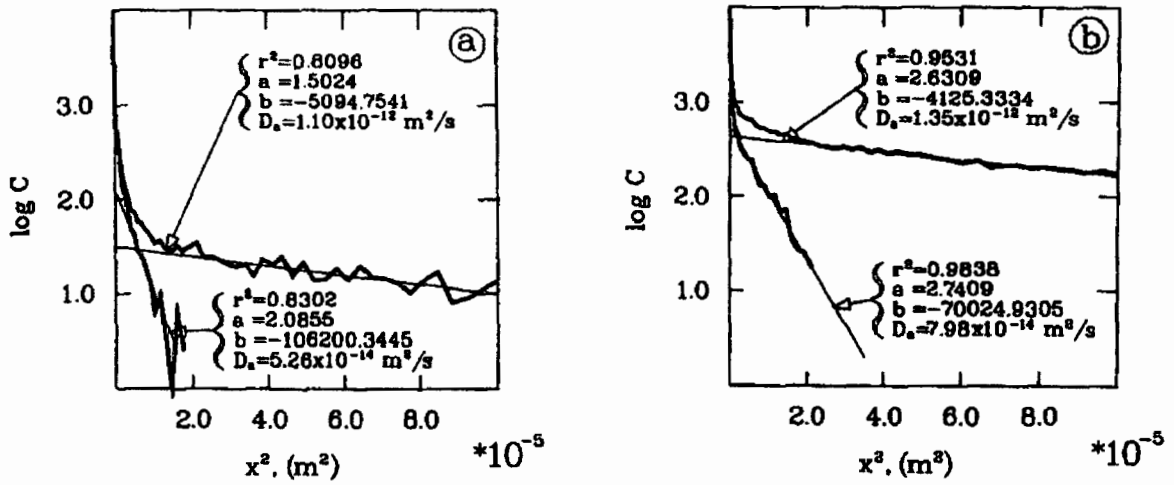

Fig. 10. Diffusion of iodine in compacted bentonite with half of the cell filled with clay mixed with 1% iron powder a), and the other half of the cell filled with clay mixed with 1% $KMnO_4$ b). Diffusion time = 225 days

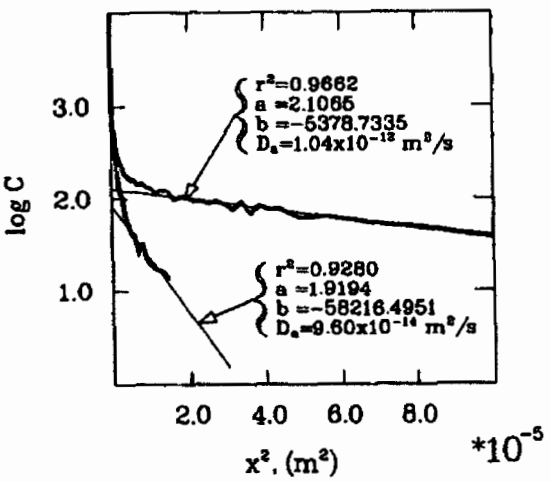

Fig. 11. Diffusion of iodine in compacted bentonite mixed with 0.5% iron powder and 1% chalcopyrite/pyrite. Diffusion time = 225 days

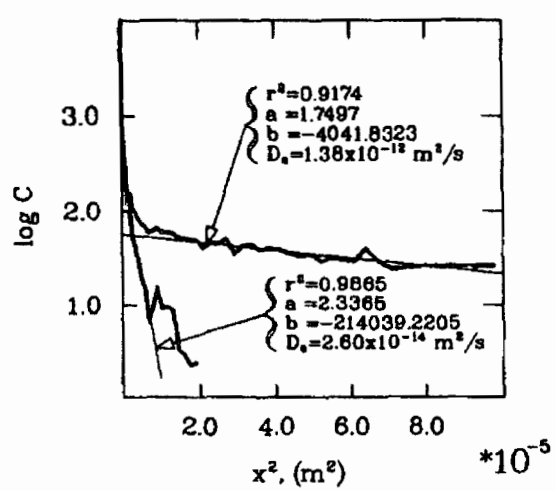

Fig. 12. Diffusion of iodine in compacted bentonite mixed with 0.5% iron powder and 1% cinnabar. Diffusion time = 226 days

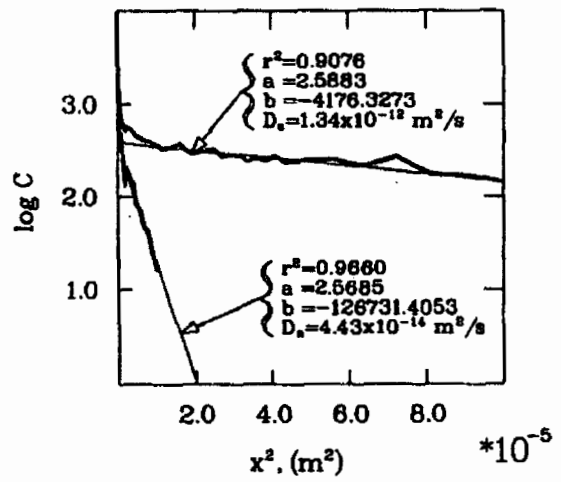

Fig. 13. Diffusion of iodine in compacted bentonite mixed with 0.5% iron powder and 1% PbO. Diffusion time = 224 days

The experiments in which attempts were made to control and preserve the chemical speciation of the iodine (as I^- by adding metallic iron and as IO_3^- by adding $KMnO_4$) did not produce any significant differences in the diffusivities compared with the experiments in normal clay (Figures 7–10). The same phenomen of two distinct mechanisms is observed, with virtually the same diffusivities as in normal clay (Table 4).

Also, when the minerals containing iodide complex-forming agents and the iodide complex-forming chemical reagent were added to the clay, no significant change from the diffusion rate in the ordinary clay could be observed (Figs. 11–13).

5. Conclusions

By writing the diffusion equation as the linear equation $\log C = \text{const.} - (1/(4D_a t))x^2$ and plotting $\log C$ versus

x^2, the apparent diffusivity, D_a, is obtained from the slope. If consecutive least square fits can be done on the plotted curve, it can be concluded that the transport consists of more than one mechanism and that it is possible to distinguish between the different transport mechanisms for the studied element, each mechanism having a distinct diffusivity.

Equation (3), $D = D_a(1 + K_d\rho(1-\epsilon)/\epsilon)$, is not applicable for species sorbing by cation exchange processes when the diffusion is slow, but seems to be useful for species sorbing by chemisorption or physical adsorption.

The apparent diffusivity of strontium is 2.0×10^{-11} m^2/s and is 2.4×10^{-12} m^2/s for cesium. Both elements sorb by cation exchange; thus, it is not possible to calculate the diffusivity, D, from D_a and K_d according to equation (3). They exist in only one oxidation state, +2 for strontium and +1 for cesium, under the conditions expected in the clay, and have only one diffusion mechanism as evaluated from the figures.

Technetium, as pertechnetate, has an apparent diffusivity of 1.2×10^{-12} m^2/s. Reduced to the tetravalent state, the apparent diffusivity of technetium is 1×10^{-13} m^2/s, although a small fraction still exhibited the same mobility as the pertechnetate.

Iodine diffuses by two mechanisms, possibly due to different species of iodine in the form of I^- and HIO or IO_3^-, or perhaps due to pore-size effects in the clay. The two overlapping mechanisms observed result in apparent diffusivities an order of magnitude apart, 1.2×10^{-12} m^2/s and 1.1×10^{-13} m^2/s for long diffusion time (218 days). The same two diffusivities were observed in reducing media, oxidizing media, and when iodide complex-forming agents were added to the clay.

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