

Diffusion of I⁻, Cs⁺ and Sr²⁺ in compacted bentonite**- Anion exclusion and surface diffusion**

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**Diffusion of Γ , Cs^+ and Sr^{2+} in Compacted
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Anion exclusion and surface diffusion.

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ABSTRACT

The diffusion of I^- , Cs^+ and Sr^{2+} in bentonite compacted to a dry density of 1.8 g cm^{-3} and saturated with two ground waters of different ionic strength have been studied experimentally using the through diffusion technique.

The I^- diffusivity and diffusion porosity were found to be concentration independent in the concentration range 10^{-8} to $10^{-2} \text{ mol dm}^{-3}$. The diffusion porosity, being only a fraction of the water porosity for normal ground waters, is strongly ionic strength dependent due to anion exclusion.

The dependence of the diffusion of Cs^+ and Sr^{2+} on the sorption intensity is accommodated by a model encompassing diffusion of the sorbed cations within the electrical double layer next to the mineral surface in addition to diffusion in the pore water.

SAMMANFATTNING

Rapporten beskriver diffusionsförsök i bentonit kompakterad till tätheten 1.8 g cm^{-3} och jämviktad med NaCl och simulerade grundvattenlösningar.

Diffusionskonstanter och fördelningskoefficienter (K_d) har beräknats genom att simulera genombrottskurvor och koncentrationsprofiler i bentoniten.

Diffusionens beroende av lösningarnas jonstyrka kan för katjonerna Cs^+ , Sr^{2+} bäst förklaras av en modell som omfattar två diffusionsmekanismer, diffusion i porvattnet och diffusion av sorberade katjoner.

Jodid-diffusionens jonstyrkeberoende, ökande diffusionsporositet med ökande jonstyrka, förklaras av anjonförträngning från små porer i den vattenmättade bentoniten.

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SUMMARY AND CONCLUSIONS

The diffusion of I^- , Cs^+ and Sr^{2+} in Wyoming MX-80 Na-bentonite, compacted to 1.8 g cm^{-3} dry density and saturated with NaCl solutions or synthetic groundwater solutions with different ionic strength, was studied experimentally by a through diffusion technique.

The apparent and effective diffusivity of I^- were found to be concentration independent in the concentration range 10^{-8} to $10^{-2} \text{ mol dm}^{-3}$. The diffusivities are increasing with increasing ionic strength of the ground water, due to anion exclusion which is ionic strength dependent.

The apparent and effective diffusion constant in bentonite equilibrated with saline ground water (NASK-solution) were found to be $D_a = 9.2(\pm 1.3) \cdot 10^{-7}$ and $D_e = 7.0(\pm 1.7) \cdot 10^{-8} \text{ cm}^2 \text{ s}^{-1}$, respectively.

The corresponding diffusivities in bentonite equilibrated with granitic ground water (Allard-solution) are $D_a = 3.5 \cdot 10^{-7}$, $D_e = 2.1 \cdot 10^{-9} \text{ cm}^2 \text{ s}^{-1}$.

The distribution coefficients $K_d (\text{cm}^3 \text{ g}^{-1})$ for Cs^+ and Sr^{2+} , calculated by simulation of the break through curves and concentration profiles from the diffusion experiments, fall well within the range of distribution coefficients observed in batch sorption experiments with bentonite suspensions and compacted bentonite and can be calculated for groundwaters with different ionic strength $I (\text{mol dm}^{-3})$ using the equations.

$$\begin{aligned}\log K_d(Sr^{2+}) &= 0.6(\pm 0.3) - 1.28(\pm 0.06) \log I \\ \log K_d(Cs^+) &= 1.54(\pm 0.3) - 0.58(\pm 0.09) \log I.\end{aligned}$$

The observed dependence of the diffusivity of Cs^+ and Sr^{2+} on the distribution coefficient is best accommodated by a model based on the assumption of diffusion within the electrical double layer next to the mineral surface in addition to pore water diffusion i.e. the diffusion constants are given by the equations

$$\begin{aligned}D_a &= \varepsilon D_p / (\varepsilon + K_d \rho) + K_d \rho D_s / (\varepsilon + K_d \rho) \\ D_e &= D_a (\varepsilon + K_d \rho).\end{aligned}$$

By using the water porosity, 0.32 for bentonite compacted to the dry density $\rho = 1.8 \text{ g cm}^{-3}$, the pore water and surface diffusivities were found to be $D_p = 8 \cdot 10^{-6}$, $D_s = 6 \cdot 10^{-9}$ and $D_p = 3 \cdot 10^{-6}$, $D_s = 1 \cdot 10^{-7} \text{ cm}^2 \text{ s}^{-1}$ for Cs^+ and Sr^{2+} respectively.

The Sr^{2+} and Cs^+ diffusivities in bentonite compacted to dry density 1.8 cm^{-3} and equilibrated with granite (Allard) and saline ground water are accordingly

Water	Ionic strength	Sr^{2+}		Cs^+	
		D_a cm^2s^{-1}	D_e cm^2s^{-1}	D_a cm^2s^{-1}	D_e cm^2s^{-1}
Allard	0.018	$1.2 \cdot 10^{-7}$	$6.7 \cdot 10^{-5}$	$1.0 \cdot 10^{-8}$	$6.4 \cdot 10^{-6}$
NASK	0.218	$1.0 \cdot 10^{-7}$	$6.0 \cdot 10^{-6}$	$2.3 \cdot 10^{-8}$	$3.5 \cdot 10^{-6}$

1. INTRODUCTION

Characteristic features of the clay to be used as buffer material in repositories for spent nuclear fuel are low permeability, high sorptivity and long term stability. The low permeability of compacted bentonite prevents material transport by hydraulic flow and makes diffusion the principal mechanism for transport of corrosive solutes to the surface of the waste canister and migration of radionuclides released on failure of a waste canister.

The diffusive transport of solutes through compacted bentonite is governed by the geometrical microstructure of the pore network i.e. the parameters porosity, tortuosity, constrictivity, and the interaction between the three components diffusing solute, water and clay. Despite extensive studies several of the processes involved are not, due to the complicated microstructure of the bentonite, fully understood. The diffusion of some sorbing solutes e.g. the cations Na^+ , Cs^+ and Sr^{2+} has repeatedly been reported to be faster than predicted from a model based on diffusion in the aqueous phase and immobilisation by sorption on the solid phase. Diffusion within the electrical double layer next to the mineral surface, referred to surface diffusion, or less sorption, i.e. a lower K_d value in the compacted than in freely expanded clay, have been offered to explain the experimental results. Van Schaik et al (1966), Rasmuson and Neretnieks (1983), Torstenfelt (1983), Eriksen and Jacobsson (1981,1984), Soudek et al (1984), Jahnke and Radke (1987), Muurinen et al (1985,1987, 1990,1994), Cheung (1989), Cook (1989), Choi et al (1992), Berry and Bond (1992), Kim et al (1993).

To shed some light on surface diffusion, if it exists, in bentonite clay and its relative importance to the function of bentonite as a barrier to radionuclide diffusion, we have studied the diffusive transport of Sr^{2+} and Cs^+ in compacted bentonite using a through diffusion technique. Apparent diffusivity and transport K_d are, in each experiment, obtained by computer simulation of the experimentally measured cumulative flux through a plug of compacted bentonite and the concentration profile within the bentonite. Due to the particular interest in Cs^+ and I^- which can possibly be released from a damaged spent fuel canister we have also studied the diffusion of these ions in the concentration range 10^{-2} to 10^{-8} mol dm^{-3} .

2. EXPERIMENTAL

2.1 Materials

The bentonite used in this investigation is the American Colloid Co. type MX-80 (Wyoming Na-bentonite). The bentonite (MX-80) has a clay content ($< 2\mu\text{m}$) of approximately 85% and a montmorillonite content of 80-90 wt% of this fraction. The remaining silt fraction contains quartz, feldspar and some micas, sulphides and oxides (Pusch and Karnland 1986). Two different synthetic groundwater solutions (Allard and NASK) were used in the diffusion experiments, see Table 1.

Table 1

Composition of ground water solutions used in the sorption and diffusion experiments.

NASK		Allard	
NaCl	140 mM	NaHCO ₃	2 mM
KHCO ₃	2 mM	CaCl ₂	0.45 mM
CaCl ₂	20 mM	KCl	0.1mM
MgSO ₄	4 mM	NaSiO ₄	0.2 mM
pH	7.7	MgSO ₄	0.1mM
Ionic strength	0.218	MgCl ₂	0.78 mM
		NaCl	10 mM
		pH	8 - 8.2
		Ionic strength	0.0182

Simulated ground waters and NaCl solutions with different concentration were used to vary the ionic strength in the sorption experiments. The solutions were prepared from analytical grade chemicals and Millipore deionized, triple distilled water. The radionuclides used, ¹³⁴Cs (Amersham) and ⁸⁵Sr, ¹²⁵I (DuPont Scandinavia) were purchased in aqueous solution. Tracer solutions were prepared by adding small aliquots of the stock solution to the solutions used in the experiments. The overall diffusant concentration was obtained by adding small volumes of standardized inactive solutions.

2.2 Diffusion experiments

The diffusion cell, made of PEEK, is shown schematically in Figure 1. Bentonite was statically compacted in the diffusion cylinder (internal diameter 10 mm and 5 mm long) to a dry density of 1.8 kg dm⁻³. The diffusion cylinder and endplates containing in and outlet channels fitted with metallic filters (0.82 mm thick) were assembled and the clay equilibrated with the aqueous solution for at least three weeks by pumping solution from reservoirs through the end plate channels, see Figure 2. After water saturation a small volume of diffusant solution, containing the tracer and inactive carrier required to give the chosen diffusant concentration, was added to the reservoir at the inlet side of the diffusion cell. The activity of the solutions in inlet and outlet reservoirs were monitored by γ -spectrometry on small sample volumes using a germanium detector and multichannel analyzer. The volume of the inlet reservoir was sufficient to keep the concentration nearly constant (within a few percent) throughout the experiments. At the end of the Cs⁺ and Sr²⁺ experiments the diffusion cell was dismantled and the bentonite sliced into thin sections. Each section was weighed and the activity measured by γ -counting.

The diffusion properties of the filters were measured in separate through diffusion experiments, using a package of five filters, and the filter porosity was measured by weighing dry and water saturated filters.

2.3 Sorption

Sorption was measured in batch experiments with 0.1 g bentonite and 20 cm³ spiked diffusant solutions.

3. DATA EVALUATION

3.1 Iodide diffusion

The filters, being thin (0.82 mm) and with porosity (0.26) comparable to the water porosity of the compacted bentonite (0.32), were judged to have but a slight effect on the diffusion of iodide through the diffusion cell. The effective and apparent diffusion constants were therefore evaluated using equations (1) and (2) respectively (Eriksen and Jacobsson 1984).

$$D_e = J L / A[C_o - C_i] \quad (1)$$

$$D_a = L^2 / 6 t_e \quad (2)$$

- D_e effective diffusion constant (cm² s⁻¹)
 D_a apparent diffusion constant (cm² s⁻¹)
 J flux through the diffusion cell (Bq s⁻¹)
 L thickness of the bentonite plug (cm)
 A surface area perpendicular to the diffusion direction (cm²)
 C_o concentration in the inlet reservoir (const) (Bq cm⁻³)
 C_i concentration in the outlet reservoir (Bq cm⁻³)
 t_e time lag, the point where the asymptote of the break through curve intercepts the time axis (See Figure 3)

3.2 Cesium and strontium diffusion.

For sorbing diffusants, e.g. Cs⁺ and Sr²⁺, evaluation of diffusion parameters using Equations 1 and 2 will be strongly influenced by the presence of filters in the diffusion cell. In evaluating the experimental diffusion data for Cs⁺ and Sr²⁺ an analysis of the complete diffusion system, as represented schematically in Figure 4 is required.

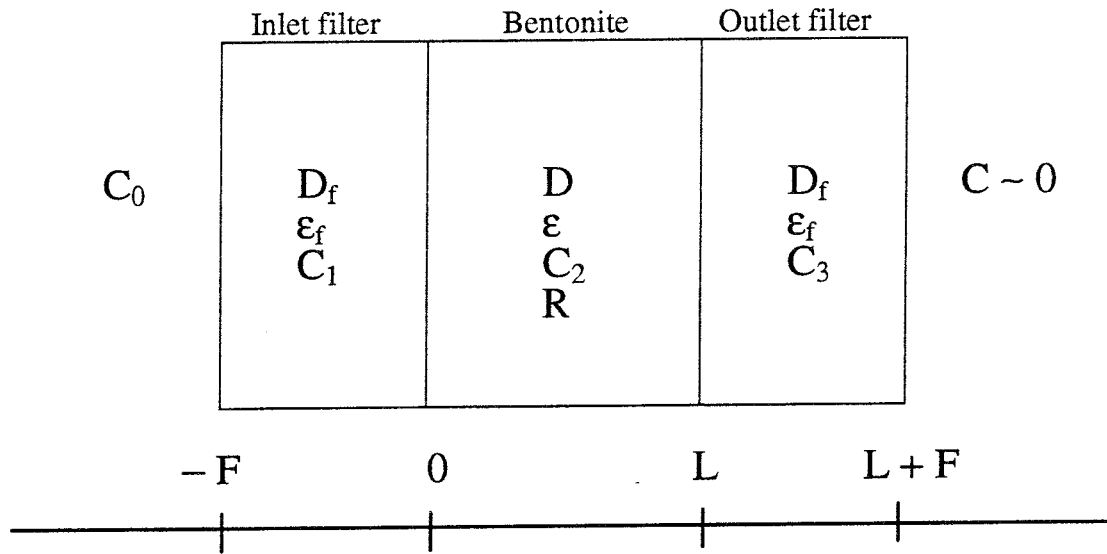


Figure 4

Schematic representation of the barriers in the diffusion cell.

The diffusive transport through the inlet filter ($-F < x < 0$) is given by equation (3)

$$J = -A \varepsilon_f D_f (\partial C_1 / \partial x) \quad (3)$$

and the boundary conditions

$$C(-F, t) = C_0$$

ε_f filter porosity

D_f apparent diffusion constant in filter ($\text{cm}^2 \text{s}^{-1}$)

F filter thickness (cm)

C_1 concentration in filter (Bq cm^{-3})

C_0 concentration in inlet solution (Bq cm^{-3})

The diffusive transport through the compacted bentonite ($0 < x < L$) is given by equation (4)

$$J = -A \varepsilon D_a R (\partial C_2 / \partial x) \quad (4)$$

R capacity (retardation) factor defined as the ratio of the overall concentration of diffusant in the compacted bentonite to the concentration in the solution accessible for diffusion

$$(R = \varepsilon + \rho K_d / \varepsilon)$$

C_2 concentration in pore solution (Bq cm^{-3}).

At the boundary between the inlet filter and the compacted bentonite we have the following conditions

$$C_1(0,t) = C_2(0,t)$$

$$\varepsilon_f D_f (\partial C_1 / \partial x)_{x=0} = \varepsilon D_a R (\partial C_2 / \partial t)_{x=0} \quad (\text{no storage in the boundary})$$

The corresponding transport equation and boundary conditions for the outlet filter are given by

$$J = -A \varepsilon_f D_f (\partial C_3 / \partial x) \quad (5)$$

$$C_2(L,t) = C_3(L,t)$$

$$\varepsilon D_a R (\partial C_2 / \partial x)_{x=L} = \varepsilon_f D_f (\partial C_3 / \partial x)_{x=L}$$

$$C_3(L+F, t) = 0$$

The accumulated flow of diffusant through the outlet filter $Q(t)$ is given by the flux through the boundary $x = L + F$ integrated from time zero to t

$$Q(t) = A \varepsilon_f D_f \int_0^t (\partial C_3(x,t) / \partial x)_{x=L+F} dt \quad (6)$$

An analytical solution to equation (6), defining the break through curve, can be obtained by the Laplace transform method (Put 1991). However, to make use of all the experimental data i.e. the accumulated flow and the concentration profile within the compacted bentonite, we have chosen to use a finite difference based simulation code (ANADIFF). The code ANADIFF calculates the apparent diffusion coefficient D_a and the sorption coefficient for transport K_d . In the simulations A , ε_f , ε , D_f , C_0 , F , L and ρ are kept constant while D_a and K_d are varied.

4. RESULTS AND DISCUSSION

4.1 Iodide

Measured apparent and effective diffusion constants for I^- in compacted ($\rho_{\text{dry}} = 1.8 \text{ g cm}^{-3}$) NASK-water equilibrated bentonite are given in Table 2 together with corresponding diffusion constants measured in an earlier study with Allard-water saturated bentonite. As can be seen, there is no concentration dependence within the concentration range 10^{-2} to $10^{-6} \text{ mol dm}^{-3}$ for the NASK-water equilibrated bentonite. In Allard-water saturated bentonite the apparent and effective diffusion constants are approximately 2-3 and 30 times respectively smaller than the corresponding diffusion constants in Allard-water saturated bentonite.

Neglecting possible slight sorption effects the I^- transport can be assumed to take place by pore water diffusion only. The relation between the apparent and effective diffusion constant is thus given by $D_e = D_a \varepsilon_p$. The calculated diffusion porosities (ε_p) are given in Table 2.

Table 2.

Diffusion constants and porosities for I⁻ in compacted bentonite ($\rho_{\text{dry}} = 1.8 \text{ g cm}^{-3}$) equilibrated with NASK and Allard ground water solutions.

Solution	I ⁻ -conc mol dm ⁻³	D _a cm ² s ⁻¹	D _e cm ² s ⁻¹	ε _d
NASK	10 ⁻²	8.6·10 ⁻⁷	5.3·10 ⁻⁸	0.06
	10 ⁻³	1.1·10 ⁻⁶	6.5·10 ⁻⁸	0.06
	10 ⁻⁴	8.7·10 ⁻⁷	9.4·10 ⁻⁸	0.11
	10 ⁻⁶	8.6·10 ⁻⁷	6.8·10 ⁻⁸	0.08
Allard*	10 ⁻⁸	3.5·10 ⁻⁷	2.1·10 ⁻⁹	0.006

* Eriksen and Jacobsson (1984)

The variations in diffusivity and diffusion porosity can be explained by the difference in ionic strength of the ground water solutions used (Allard 0.018 and NASK 0.218 mol dm⁻³) and anion exclusion from the small pores. Increasing the iodide concentration in Allard water to 10⁻² mol dm⁻³ may thus be expected to give a small increase in the apparent and effective diffusion constants as well as diffusion porosity given in Table 2 due to the increase in ionic strength on addition of iodide to the solution.

4.2. Cesium and strontium

Typical experimental and simulated accumulated flux and concentration profile plots are shown in Figures 5 a, b and 6 a, b. Apparent diffusion constants and diffusion K_d values obtained by simulation calculations are given in Table 3.

Table 3

Apparent diffusivities and distribution coefficients for Cs⁺ and Sr²⁺ in compacted bentonite ($\rho_{\text{dry}} = 1.8 \text{ g cm}^{-3}$) equilibrated with Allard and NASK groundwater solutions.

Solution	Ion	Conc. mol dm ⁻³	D _a cm ² s ⁻¹	K _d cm ³ g ⁻¹
NASK	Sr ²⁺	10 ⁻⁴	1.05 · 10 ⁻⁷	11
		10 ⁻⁶	1.4 · 10 ⁻⁷	8.5
	Cs ⁺	10 ⁻²	5.0 · 10 ⁻⁸	40.5
		10 ⁻⁴	2.6 · 10 ⁻⁸	46.6
		10 ⁻⁶	3.2 · 10 ⁻⁸	46.8
Allard	Cs ⁺	10 ⁻⁸	2.0 · 10 ⁻⁸	85
		10 ⁻⁶	0.8 · 10 ⁻⁸	270
		10 ⁻⁴	1.4 · 10 ⁻⁸	270
		10 ⁻²	1.1 · 10 ⁻⁸	450
		10 ⁻⁸	1.1 · 10 ⁻⁸	450

Distribution coefficients for Cs⁺ and Sr²⁺ measured in batch experiments using bentonite suspended in NaCl solutions and ground waters with different ionic strength (I) are shown in Log K_d versus Log I plots in Figures 7 and 8 respectively. In the strontium plot (Figure 8) we have included data from ionic-strength dependence studies by Muurinen (1994) and Oscarson (1984).

We have also plotted the K_d-values obtained by simulation of the diffusion break through curves and concentration profiles in the solid phase into Figures 7 and 8. Inspection of the plots in Figures 7 and 8 reveals that the diffusion K_d-values fall well within the range of distribution coefficients obtained in batch sorption experiments. The assumption of lower K_d-values for diffusive transport in compacted bentonite than for sorption in suspensions is therefore not borne out in this study.

The apparent diffusion constants for Cs⁺ and Sr²⁺ are, together with data from earlier studies by Muurinen (1994) and Eriksen and Jacobsson (1984), shown in Log D_a versus Log K_d plots in Figures 9 and 10, respectively. Inspection of these plots shows that in the K_d interval 10 to 1000 the apparent Cs⁺ diffusivity decreases by approximately one order of magnitude whereas the apparent Sr²⁺ diffusivity is virtually constant. This is clearly not in accordance with a diffusion model for porous media encompassing diffusion in the pore water and complete immobilization of sorbed species, yielding equation (7)

$$D_a = \varepsilon D_p / (\varepsilon + K_d \rho) \quad (7)$$

where D_p is the pore diffusion constant, D_a the apparent diffusion constant, K_d the distribution coefficient and ρ the dry bulk density of the porous medium. According to equation (7) the apparent diffusivity should decrease nearly linearly with increasing K_d. Instead assuming diffusion of sorbed cations to take place and to be Fickian, equation (7) can be modified to read

$$D_a = \varepsilon D_p / (\varepsilon + K_d \rho) + K_d \rho D_s / (\varepsilon + K_d \rho) \quad (8)$$

where D_s is the surface diffusivity.

The effective diffusivity is given by equation (9)

$$D_e = D_a (\varepsilon + K_d \rho) \quad (9)$$

Fitted D_a and D_e curves calculated using equations (8) and (9) are plotted into Figures 9 and 10. As can be seen, good fits to experimental data were obtained using $D_p = 8 \cdot 10^{-6}$, $D_s = 6 \cdot 10^{-9}$ $\text{cm}^2 \text{s}^{-1}$ for Cs^+ and $D_p = 3 \cdot 10^{-6}$ and $D_s = 1 \cdot 10^{-7}$ $\text{cm}^2 \text{s}^{-1}$ for Sr^{2+} . These surface diffusivities are in fair agreement with results obtained by Muurinen (1994) and Choi et al (1992).

It should be pointed out that our calculations are based on the water porosity of the bentonite ($\varepsilon \pm 0.32$). It may be argued that the diffusion porosity is smaller than the water porosity, but we have presently not sufficient information on the ionic strength dependent diffusion porosity. The different diffusion constants for Cs^+ and Sr^{2+} are summarized in Table 4.

Table 4

Diffusion constants for Cs^+ and Sr^{2+} in aqueous solution and compacted bentonite.

Ion	D_w $\text{cm}^2 \text{s}^{-1}$	D_p $\text{cm}^2 \text{s}^{-1}$	D_s $\text{cm}^2 \text{s}^{-1}$
Cs^+	$2.1 \cdot 10^{-5}$	$8.0 \cdot 10^{-6}$	$6.0 \cdot 10^{-9}$
Sr^{2+}	$0.78 \cdot 10^{-5}$	$3.0 \cdot 10^{-6}$	$1.0 \cdot 10^{-7}$

The ratio D_p/D_w is found to be the same for both ions indicating that the overall effect of tortuosity and constrictivity is the same.

The ionic strength dependence of the distribution coefficients clearly points at ion exchange as the main sorption mechanism. The Cs^+ ion, having a larger radius than Sr^{2+} , is very weakly solvated and may be expected to be more specifically bound closer to the solid surface and thereby less mobile than the strongly solvated Sr^{2+} which is separated from the surface by at least one molecule of water.

5. ACKNOWLEDGEMENTS

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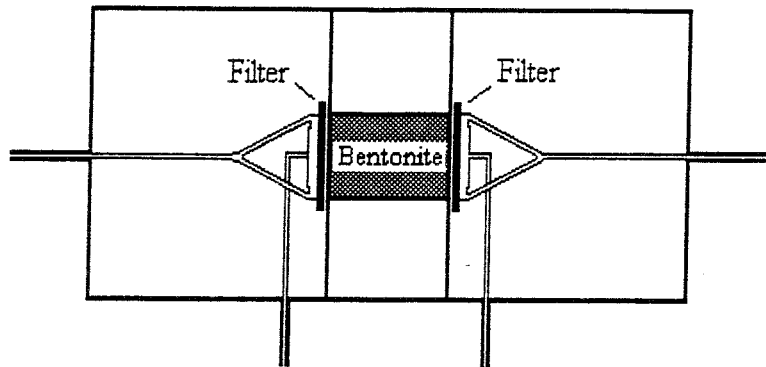


Figure 1

Schematic drawing of diffusion cell.

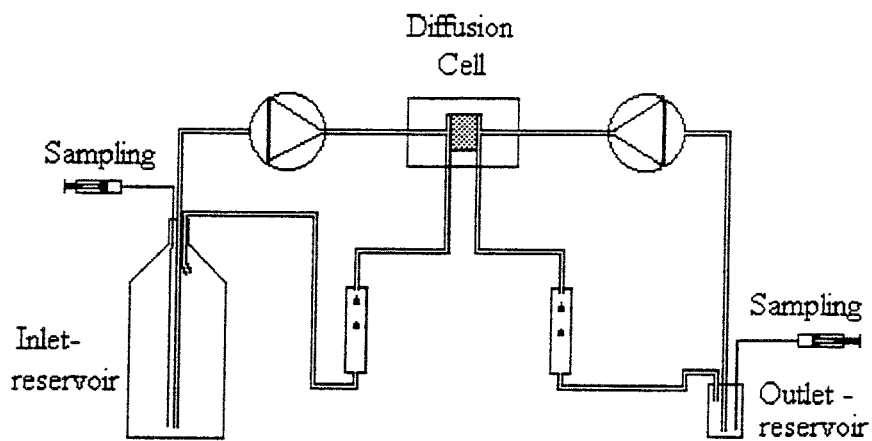


Figure 2

Arrangement of through diffusion experiments.

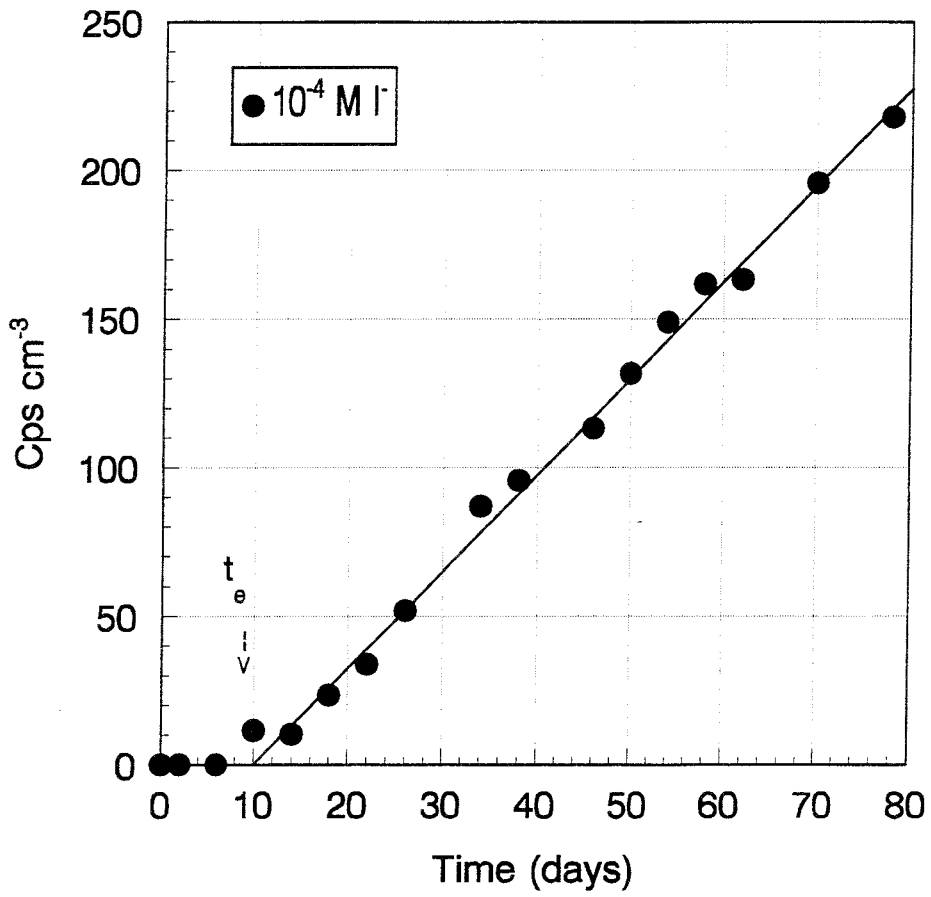


Figure 3.

Cumulative flux plot from I through diffusion experiment.
 Dry density 1.8 g cm⁻³, NASK-solution.

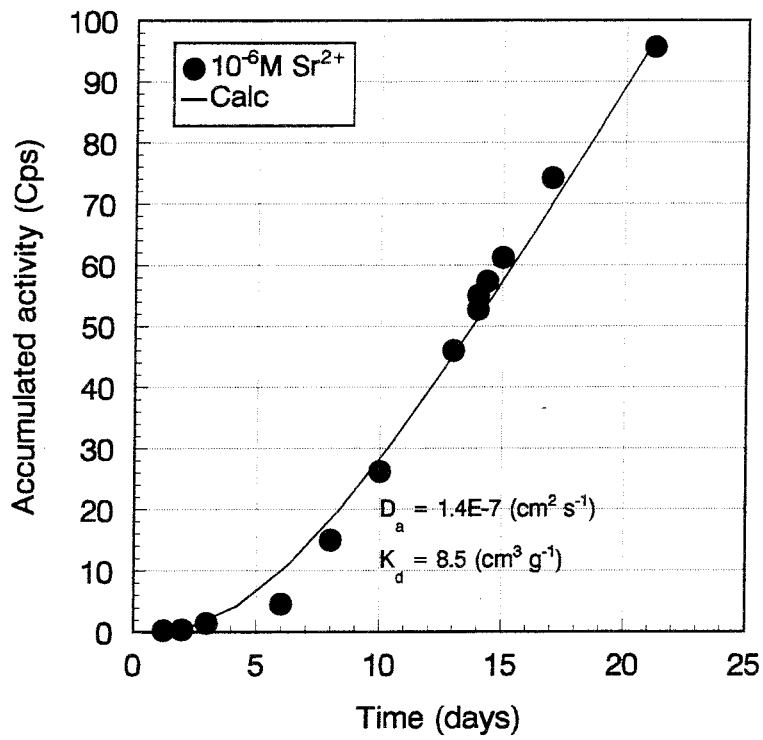


Figure 5a.

Experimental and calculated break through curves for 10^{-6} M Sr^{2+} .
Dry density 1.8 g cm^{-3} , NASK-solution.

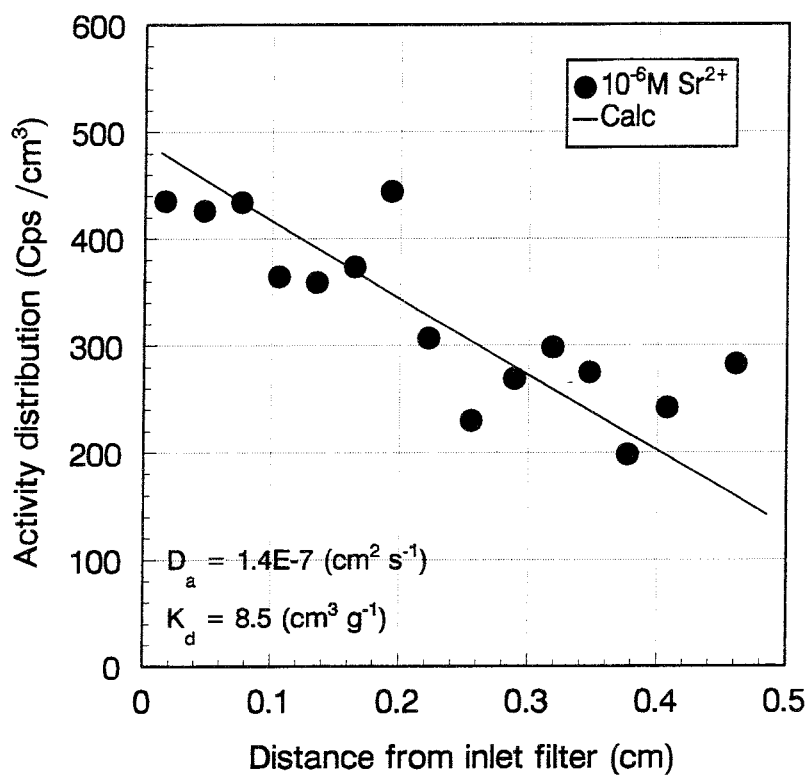


Figure 5b.

Experimental and calculated activity profiles in bentonite plug.
 $10^{-6} \text{ M Sr}^{2+}$, dry density 1.8 g cm^{-3} , NASK-solution.

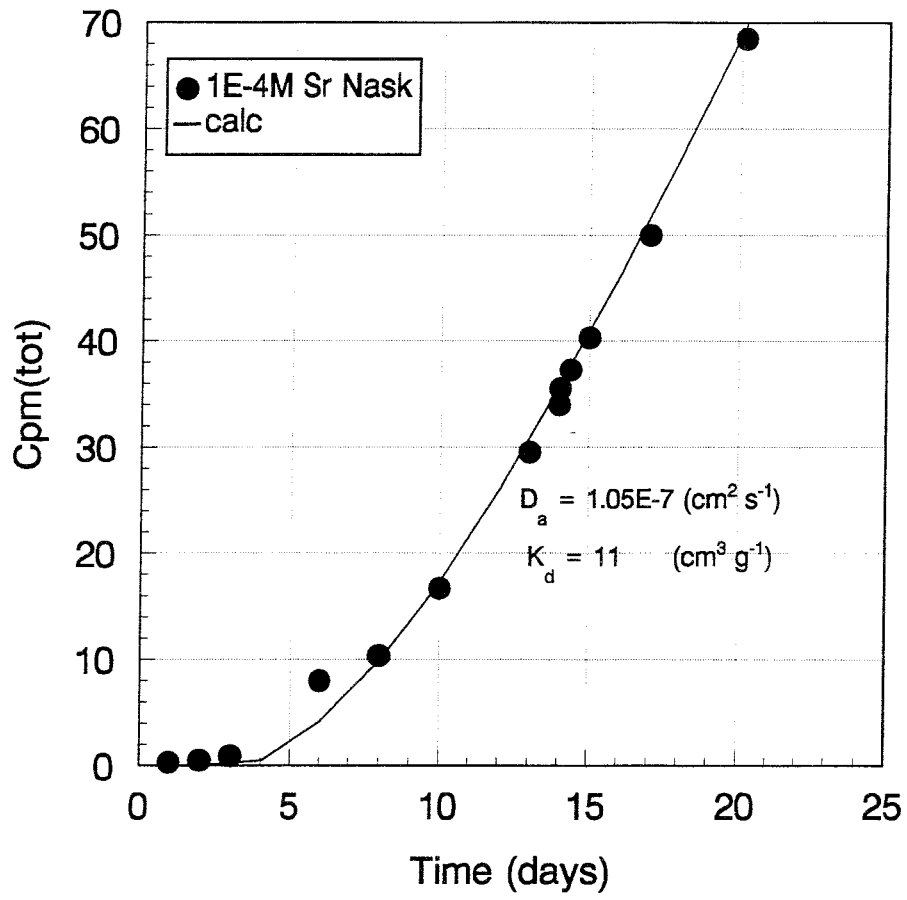


Figure 6a.

Experimental and calculated break through curves for 10^{-4} M Sr^{2+} .
 Dry density 1.8 g cm^{-3} , NASK-solution.

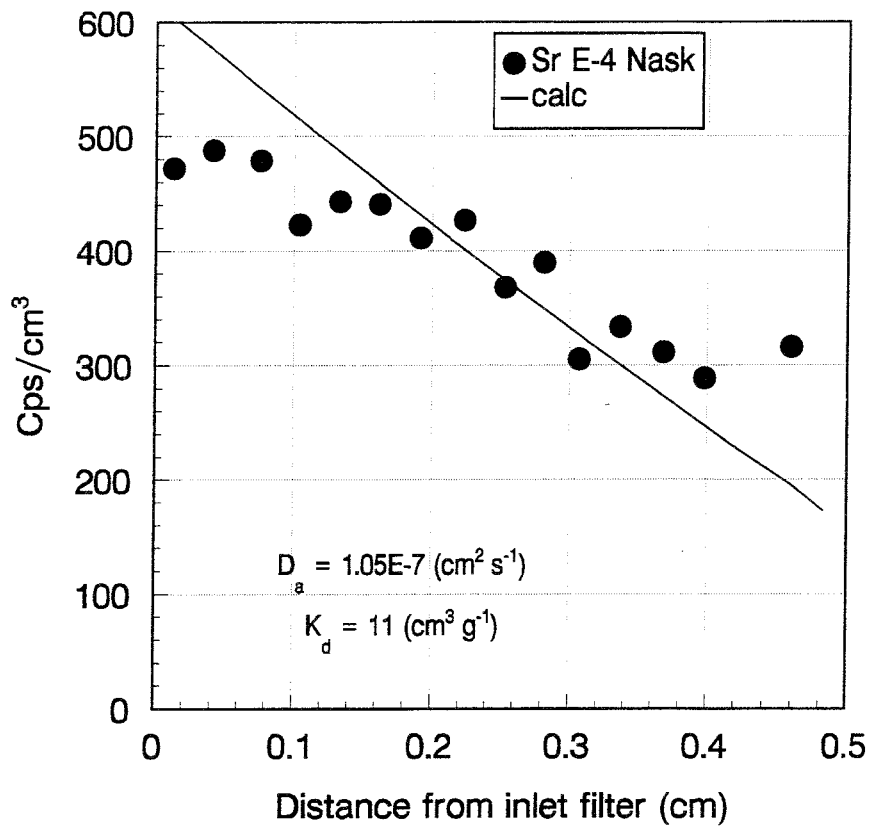


Figure 6b.

Experimental and calculated activity profiles in bentonite plug.
 10^{-4} M Sr^{2+} , dry density 1.8 g cm^{-3} , NASK-solution.

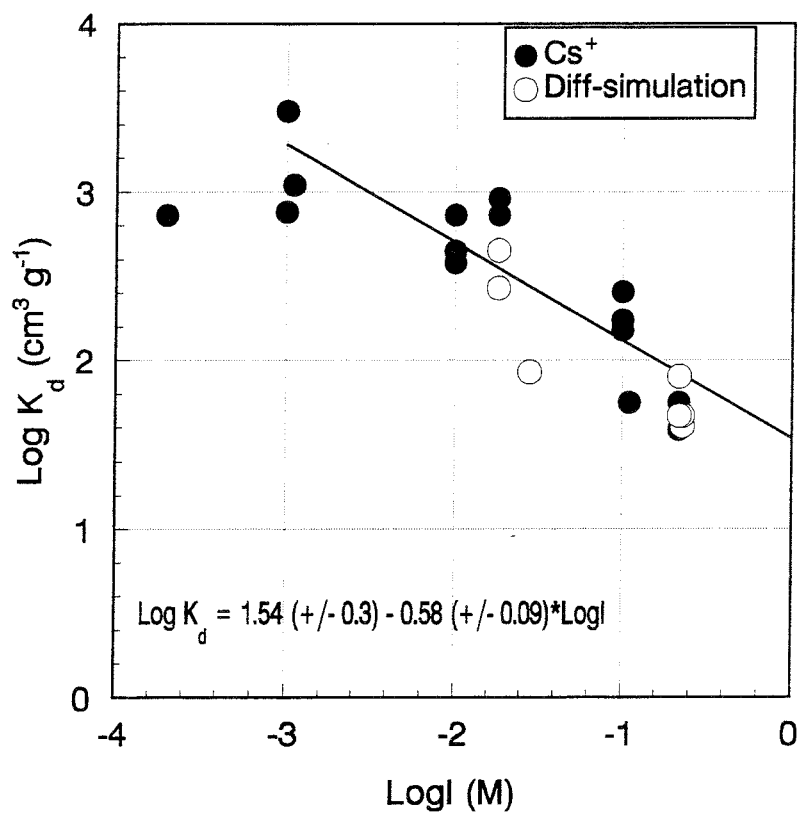


Figure 7.

Logarithmic plot of distribution coefficients (K_d) as function of ionic strength of solutions used in Cs⁺ sorption and diffusion experiments. Cs⁺ concentrations 10^{-2} to 10^{-8} mol dm⁻³.

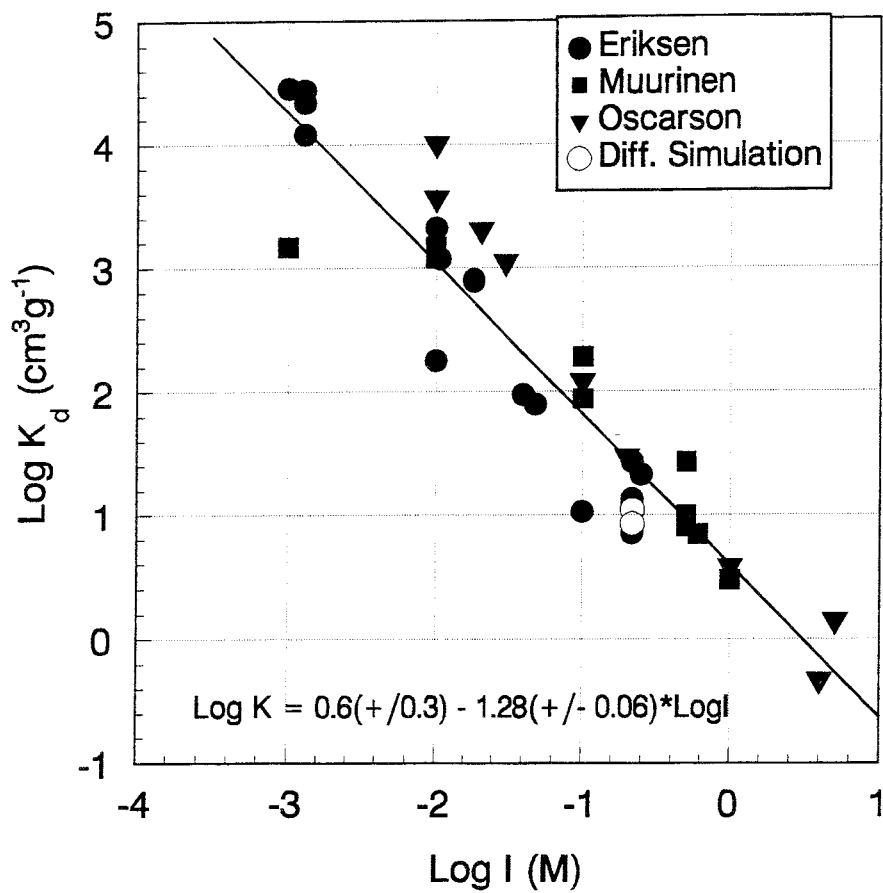


Figure 8.

Logarithmic plot of distribution coefficients (K_d) as function of ionic strength of solutions used in Sr^{2+} sorption and diffusion experiments.

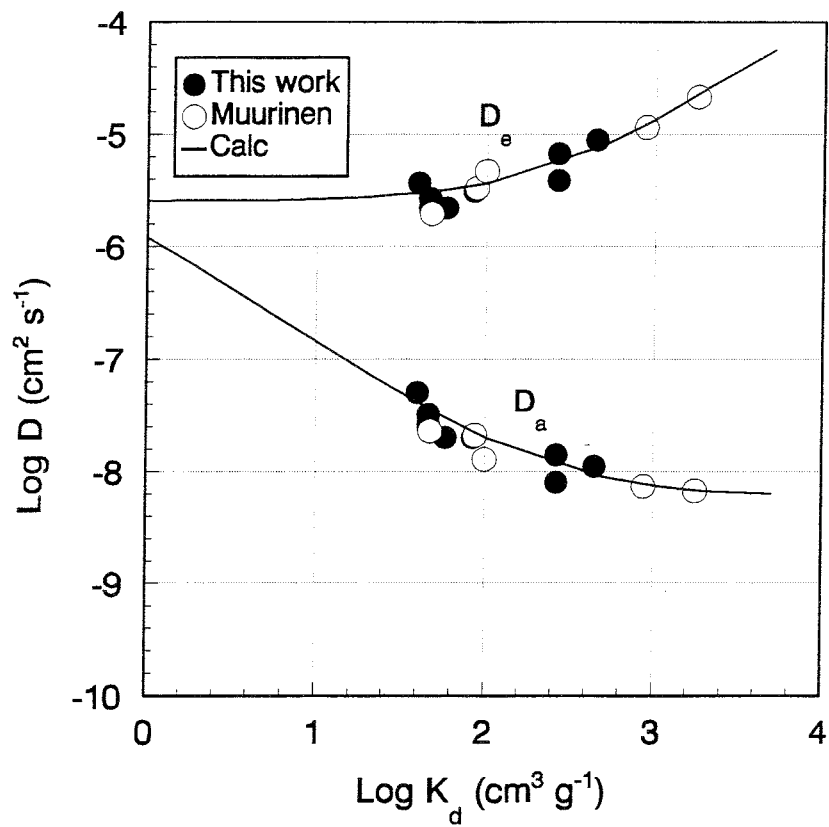


Figure 9.

Logarithmic plot of apparent and effective Cs⁺ diffusivity as function of distribution coefficient (K_d).

The lines are calculated using $D_p = 8 \cdot 10^{-6}$ and $D_s = 6 \cdot 10^{-9} \text{ cm}^2 \cdot \text{s}^{-1}$.

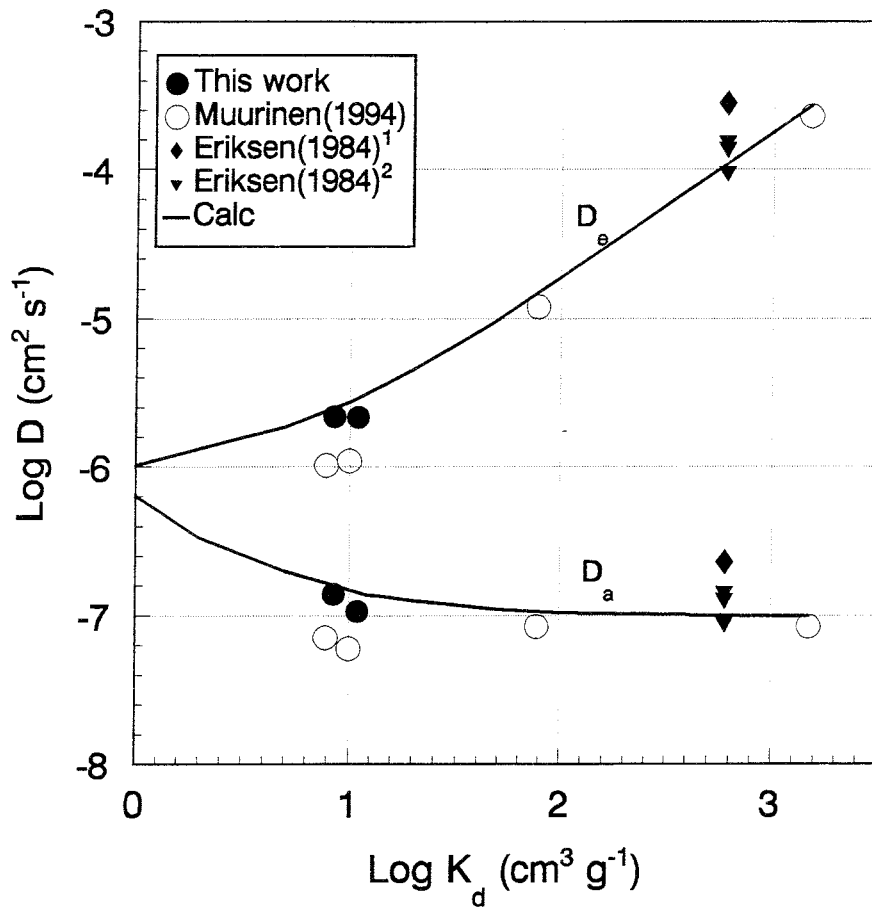


Figure 10.

Logarithmic plot of apparent and effective Sr^{2+} diffusivity as function of distribution coefficient (K_d). The lines are calculated using $D_p = 3 \cdot 10^{-6}$ and $D_s = 1 \cdot 10^{-7} \text{ cm}^2 \text{ s}^{-1}$. The data from Eriksen (1984) are from experiments with different techniques.

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(GRAM), Aix-en-Provence, France
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