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**DIFFUSIVE TRANSPORT OF STRONTIUM-85  
IN SAND-BENTONITE MIXTURES**

**MIGRATION PAR DIFFUSION DU STRONTIUM-85  
DANS DES MELANGES DE SABLE ET BENTONITE**

**R. W. Gillham, M. J. L. Robin and D. J. Dytynyshyn**

**Whiteshell Nuclear Research  
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**Etablissement de recherches  
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RÉSUMÉ

On a effectué des essais de diffusion pour déterminer la migration du  $^{85}\text{Sr}$  dans des mélanges de sable et bentonite. Ces essais ont mis en jeu un sol naturel (sable de Chalk River) et sept mélanges de bentonite et sable de sélice à teneur de 0% à 100% de bentonite. On s'est servi de deux solutés non réactifs ( $^{36}\text{Cl}$  et  $^3\text{H}$ ) et d'un soluté réactif ( $^{85}\text{Sr}$ ) lors de l'étude. Les essais effectués avec les solutés non réactifs ont donné des estimations de facteurs de sinuosité. On a obtenu les facteurs de ralentissement à partir des valeurs d'essais de porosité et d'essais de densité apparente et à partir des coefficients de distribution discontinue ( $K_d$ ). Ces valeurs  $K_d$  sont un moyen simple de décrire la réaction soluté/milieu et sont basées sur l'hypothèse où la réaction d'échange de cations peut être décrite par une isotherme d'adsorption linéaire passant par l'origine.

Les résultats démontrent que, à des fins pratiques et pour les conditions d'essais qui nous intéressent, l'emploi du coefficient de distribution est un moyen convenable pour calculer le coefficient de diffusion efficace dans le cas du  $^{85}\text{Sr}$ . En outre, on a constaté que la porosité et la densité apparente ont un très grand effet sur le coefficient de diffusion efficace du fait du facteur de ralentissement. On a constaté que les mélanges à teneur de 5-10% de bentonite ralentissent le  $^{85}\text{Sr}$  avec une plus grande efficacité que le sable à lui seul ou les mélanges à plus forte teneur en bentonite. Dans les sols à plus forte teneur en bentonite, l'effet de la capacité accrue d'échange de cations est équilibré par le rapport décroissant de la densité apparente à la porosité.

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ABSTRACT

Diffusion experiments have been used to determine the transport of  $^{85}\text{Sr}$  in sand-bentonite mixtures. The diffusion experiments were performed on one natural soil (Chalk River sand) and on seven mixtures of bentonite and silica sand, containing from 0% to 100% bentonite. Two non-reactive solutes ( $^{36}\text{Cl}$  and  $^3\text{H}$ ) and one reactive solute ( $^{85}\text{Sr}$ ) were used in the study. The experiments with non-reactive solutes yielded estimates of tortuosity factors. Retardation factors were obtained from experimental porosities, experimental bulk densities, and from batch distribution coefficients ( $K_d$ ). These  $K_d$  values are a simple way of describing the solute/medium reaction, and are based on the assumption that the cation-exchange reaction may be described by a linear adsorption isotherm passing through the origin.

The results demonstrate that, for practical purposes and for our experimental conditions, the use of the distribution coefficient provides a convenient means of calculating the effective diffusion coefficient for  $^{85}\text{Sr}$ . The porosity and bulk density were also found to have a considerable influence on the effective diffusion coefficient, through the retardation factor. Mixtures containing 5-10% bentonite were found to be more effective in retarding  $^{85}\text{Sr}$  than either sand alone, or mixtures containing more bentonite. In the soils of higher bentonite content, the effect of increased cation-exchange capacity was balanced by a decreasing ratio of bulk density to porosity.

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## CONTENTS

	<u>Page</u>
1. INTRODUCTION	1
2. MATHEMATICAL DEVELOPMENT	1
3. MATERIALS AND METHODS	5
3.1 POROUS MEDIA	5
3.2 SOLUBLE CATIONS AND $K_d$ DETERMINATIONS	6
3.2.1 Centrifugation Procedure	6
3.2.2 Soluble Cations	6
3.2.3 Strontium-85 Batch Distribution Coefficients ( $K_d$ )	8
3.3 DIFFUSION EXPERIMENTS	8
3.3.1 Diffusion Cells	8
3.3.2 Soil Preparation	10
3.3.3 Diffusion Cell Preparation	10
3.3.4 Sectioning, Extraction and Analysis	12
3.3.5 Normalized Concentration Profiles	14
4. RESULTS AND DISCUSSION	15
4.1 RETARDATION PARAMETERS	15
4.2 TORTUOSITY FACTORS	20
4.3 CALCULATED AND EXPERIMENTAL STRONTIUM-85 DIFFUSION COEFFICIENTS	23
5. CONCLUSIONS	32
ACKNOWLEDGEMENTS	33
REFERENCES	34

## 1. INTRODUCTION

It is anticipated that fine-grained geological materials will be used in the buffer and backfill of a high-level radioactive waste disposal vault (Bird and Cameron, 1982). It is therefore reasonable to expect molecular diffusion to be the predominant mechanism of contaminant transport through these materials. The diffusion coefficient of the radionuclides of interest in the materials used therefore becomes a critical parameter for predicting migration rates through the buffer and backfill. The objective of this study was to determine if calculated diffusion coefficients could be used for predictive purposes, or if the longer and more costly approach of measuring diffusion coefficients directly would be required.

## 2. MATHEMATICAL DEVELOPMENT

Studies of solute transport in porous media have shown that there are many situations where molecular diffusion is the dominant migration mechanism (Gillham and Cherry, 1981; Smiles and Philip, 1978). In these situations the transport characteristics can be described by the diffusion coefficient of the solute in the medium. However, the transport behaviour is somewhat more complex for solutes that react with the porous medium.

The usual equation used to describe the transport of a solute in a porous medium is the result of the combination of a mass-balance equation and a solute-flux equation. The solute-flux equation is based on a phenomenological analogy to Fick's law. Using the continuum approach (Bear, 1972), the one-dimensional equation describing the transport of a reactive solute in a porous medium is:

$$\frac{\partial c}{\partial t} + \frac{\rho}{\eta} \frac{\partial q}{\partial t} = D \frac{\partial^2 c}{\partial x^2} , \quad (1)$$

where  $\eta$  is the porosity of the medium, or the volumetric water content in the unsaturated condition ( $L^3 \cdot L^{-3}$ );  $c$  is the concentration of solute in the liquid ( $M \cdot L^{-3}$ );  $\rho$  is the bulk density of the medium ( $M \cdot L^{-3}$ );  $q$  is the amount of solute associated with the solid phase ( $M \cdot M^{-1}$ );  $D$  is the molecular diffusion coefficient of the solute in the medium ( $L^2 \cdot T^{-1}$ );  $x$  is the space coordinate (L); and  $t$  is time (T).

In equation (1), the first term on the left-hand side describes the rate of accumulation of solute in solution per unit volume of the porous medium, and the second term, referred to as the reaction term, accounts for the rate of accumulation of solute in the solid phase of the porous medium. The term on the right-hand side of equation (1) describes the flux of solute into and out of the volume element (i.e. the divergence of the flux). It is pertinent to point out that reactions other than those that can be described by the reaction term of equation (1) must be represented by separate terms.

In many natural porous media, one of the most common reactions is cation exchange, wherein one cationic species associated with the surface of the porous medium is replaced by another. There are many ways of expressing the reaction term to account for cation exchange (Boast, 1973; Bolt, 1979; Robin, 1980). One of the simplest assumes that the amount of solute associated with the porous material is proportional to the concentration of the solute in solution, i.e.

$$q = K_d c \tag{2}$$

where  $K_d$  is referred to as the distribution coefficient, and has units of  $L^3 \cdot M^{-1}$ . The important assumptions underlying the use of a reaction term expressed in this manner are:

- the reaction is instantaneous, i.e.  $q$  is not a function of time, except through  $c$ ,
- $q$  is a unique function of  $c$ , i.e. the reaction is fully reversible, and non-hysteretic,



- $q$  is a unique function of  $c$ , i.e. other factors that may affect the reaction, such as the concentrations and types of other cations and anions present in solution, remain unchanged in the course of the reaction,
- $q$  is a linear function of  $c$ , i.e.  $K_d$  is a constant.

One can use cation-exchange theory to show that these assumptions are justified if the concentration of the species of interest is low with respect to the concentration(s) of the competing cation(s).

If the use of equation (2) is justified, equation (1) reduces to the same form as the non-reactive solute diffusion equation:

$$\frac{\partial c}{\partial t} = D_s \frac{\partial^2 c}{\partial x^2} \quad , \quad (3)$$

where  $D_s$  is the effective diffusion coefficient of the reacting solute in the medium.  $D_s$  is given by:

$$D_s = D/R \quad , \quad (4)$$

where  $R$ , the retardation factor, is defined as:

$$R = 1 + \frac{\rho}{\eta} K_d \quad . \quad (5)$$

In equations (1) and (4),  $D$  is usually assumed to be related by a "tortuosity factor" to the molecular diffusion coefficient of the solute in free solution. That is:

$$D = \tau D_o \quad (6)$$

where  $D_o$  is the molecular diffusion coefficient of the solute in free solution ( $L^2 \cdot T^{-1}$ ), and  $\tau$  is the tortuosity factor ( $L^2 \cdot L^{-2}$ ) that reflects the effects of physical properties of the porous medium on the diffusion process.

For a porous medium of infinite length in which, at time zero, one half of the medium contains the tracer and the other half does not, the following initial and boundary conditions are applicable:

$$c(x,0) = c_0 \text{ for } x < 0, \text{ and } c(x,0) = 0 \text{ for } x > 0 \quad , \quad (7a)$$

and

$$c(-\infty,t) = c_0 \quad , \text{ and } c(\infty,t) = 0 \quad , \quad (7b)$$

where  $c_0$  is the concentration of solute in the "tagged" part of the porous medium. Solving equation (3) for these conditions gives:

$$c/c_0 = 1/2 \operatorname{erfc} (x/2\sqrt{D_s t}) \quad . \quad (8)$$

This solution is also applicable to porous media of finite length, as long as the diffusion times are short enough that the concentrations at the boundaries remain unchanged. If this is not the case, another solution must be used. For a porous medium of length  $L$ , the applicable initial and boundary conditions are:

$$c(x,0) = c_0 \text{ for } 0 < x < x_0 \quad , \text{ and } c = 0 \text{ for } x_0 < x < L \quad , \quad (9a)$$

and

$$\frac{\partial c}{\partial x} = 0 \quad , \text{ at } x = 0 \text{ and } x = L \quad . \quad (9b)$$

In this case the solution to equation (3) is (Carslaw and Jaeger, 1959):

$$\frac{c}{c_0} = \frac{x_0}{L} + \frac{2}{\pi} \sum_{\eta}^{\infty} \frac{\exp \left( - \frac{D_s \eta^2 \pi^2 t}{L^2} \right) \cos \left( \frac{\eta \pi x}{L} \right) \sin \left( \frac{\eta \pi x_0}{L} \right)}{\eta} \quad . \quad (10)$$

It should be noted that equation (8) gives the plane through which diffusion occurs at  $x = 0$ , whereas the equivalent plane is situated at  $x = x_0$  for equation (10). For a given time and  $D_s$  value, it is then possible to calculate a diffusion profile from equation (8), or equation (10), depending on the conditions of the experiment.

Values of  $D_s$  may be obtained from diffusion experiments, but if  $K_d$  is large the time required for a measurable diffusion profile to develop

may be several months. An alternative and much shorter approach is to obtain  $K_d$  values from batch experiments (requiring less than a few days), and to calculate  $D_s$ . The calculated  $D_s$  may be obtained from the molecular diffusion coefficient of the solute in free solution and from a measured or estimated tortuosity factor.

In the early part of this study  $K_d$  values were measured using batch techniques, and tortuosity factors were determined from the results of non-reacting solute diffusion experiments. Values of  $D_s$ , and concentration profiles, were then calculated. These calculated profiles were compared to experimental diffusion data obtained for the reactive solute. The first objectives were to establish reliable experimental techniques and to obtain preliminary comparisons of measured and calculated diffusion coefficients for a single soil and a single radionuclide ( $^{85}\text{Sr}$ ). The results of the first year's investigations are given in Johnston and Gillham (1980). In the second year, the investigations, reported here, were extended to include materials that could be used for buffer and backfill in a high-level waste disposal vault. In particular, measured and calculated diffusion coefficients were compared for various mixtures of bentonite and silica sand.

### 3. MATERIALS AND METHODS

#### 3.1 POROUS MEDIA

The porous media (soils) used in this study were a Chalk River sand and seven mixtures of silica sand and bentonite clay. The silica sand was sieved and particles larger than 63  $\mu\text{m}$  were retained and washed with deionized water. The bentonite clay was air-dried and passed through a 62.5- $\mu\text{m}$  mesh. The sand-bentonite mixtures contained 0, 5, 10, 15, 25, 50, and 100% bentonite by weight.

## 3.2 SOLUBLE CATIONS AND $K_d$ DETERMINATIONS

### 3.2.1 Centrifugation Procedure

The centrifugation device used for these determinations consisted of a filtering centrifuge tube (modified 50-mL syringe) held over a sample collection vial. Both the tube and vial were held in a container that fit the "head" of the centrifuge. The inside of the container was designed to bear the weight of the tube while keeping it centered over the sample vial.

Each filtering centrifuge tube was made by first removing the plunger and cutting off the tip of a 50-mL syringe. A perforated nylon plug of a diameter and shape matching that of the inside of the syringe was inserted at the bottom of the syringe. A 0.45- $\mu$ m membrane filter (25-mm diameter) was placed immediately on top of the nylon plug, followed by a number 540 filter paper of the same diameter. A rubber O-ring (2.54 cm x 2.22 cm) was used to hold the filters in place and to prevent solution from seeping along the edge of the nylon plug (see Figure 3.1).

### 3.2.2 Soluble Cations

To obtain an estimate of the "native" soil-solution composition, the concentrations of water-soluble calcium, magnesium, sodium and potassium were determined for all soils. Deionized water was added to duplicate samples of each soil, in soil-to-solution ratios comparable to those in the diffusion experiments. The soil-water mixture was allowed to equilibrate, with frequent stirring, for five days in plastic beakers with screw-on lids. After five days, the soil-water mixtures were transferred to the "filtering" centrifuge tubes described previously. The water was extracted from the soils by centrifuging at 11 000 r/min (20 000 G) for 25 minutes.

To obtain a sufficient volume for analysis, the solutions thus obtained in the sampling vials were weighed and diluted with glass-distilled water. The diluted extracts were analysed for calcium, magnesium,

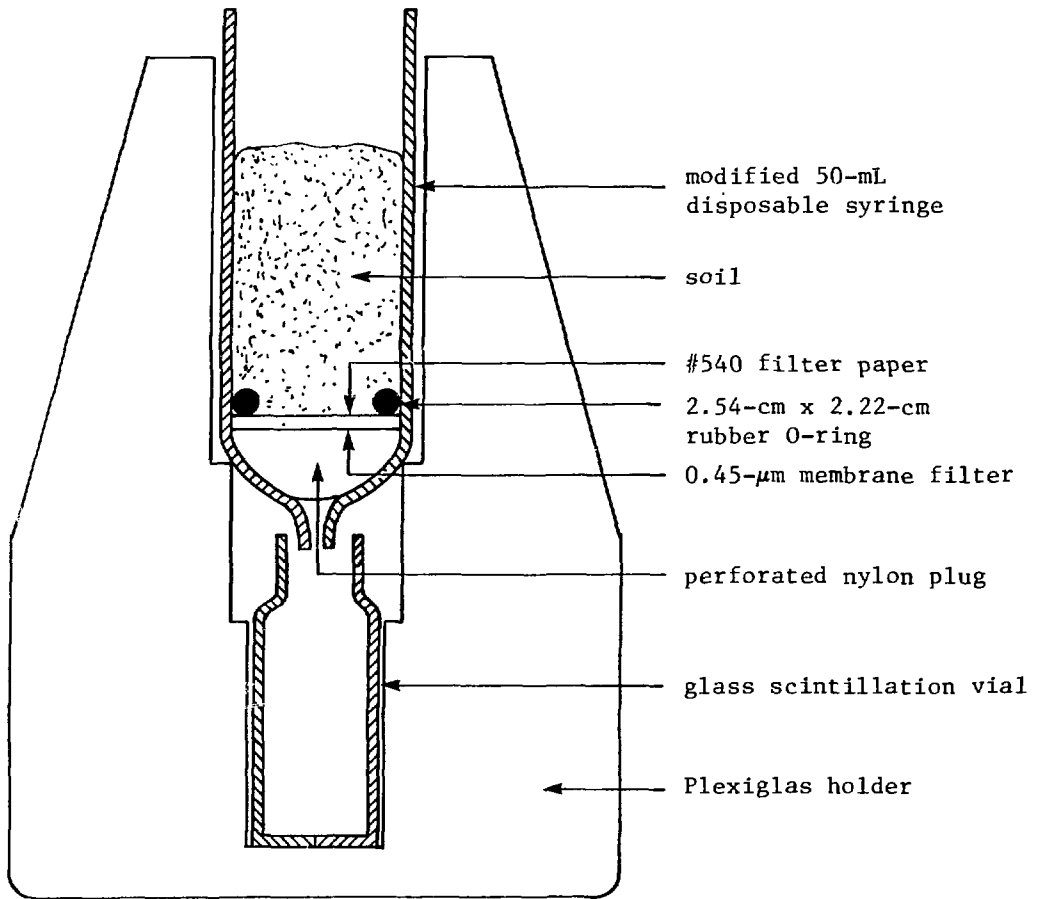


FIGURE 3.1: Centrifugation Device Used for Extracting Water from Soil for Soluble Cation and  $K_d$  Determinations

sodium and potassium using spectrophotometric techniques, and the extract concentrations were back-calculated using the dilution factor. The results are given in Table 3.1.

### 3.2.3 Strontium-85 Batch Distribution Coefficients ( $K_d$ )

Distribution coefficients of  $^{85}\text{Sr}$  were determined in duplicate for all soils. The soil-to-solution ratio used for each soil was close to that measured in the diffusion experiments. The  $^{85}\text{Sr}$  solution used was a  $1.15 \times 10^{-6}$  mol/L solution of strontium chloride. The equilibration and extraction procedures used were those described in the previous section.

The extracted soil solution was diluted with 4.0 mL of deionized water. Three mL of the resulting diluted soil solution were placed in a gamma radiation counter. The  $^{85}\text{Sr}$  activity obtained was expressed in counts per minute (cpm) per mL of soil solution.  $K_d$  values were then calculated from:

$$K_d = \left( \frac{c_1}{c} - 1 \right) \frac{V}{M} , \quad (11)$$

where  $c_1$  is the number of cpm per mL of stock solution added to the soil initially,  $c$  is the number of cpm per mL of equilibrium soil solution,  $V$  is the volume of  $^{85}\text{Sr}$  solution added, and  $M$  is the mass of dry soil used. The results of the  $K_d$  measurements are presented in Table 4.3 and discussed in Section 4.1.

## 3.3 DIFFUSION EXPERIMENTS

### 3.3.1 Diffusion Cells

Each diffusion cell consisted of two Plastipak 50-mL plastic syringes with an inner diameter of 2.65 cm. The syringes were modified by removing the needle fittings and then removing the ridge around the inner

TABLE 3.1

SOLUBLE CATION CONTENT OF EACH SOIL, USING SOIL-TO-SOLUTION RATIOS  
COMPARABLE TO THOSE FOUND IN THE DIFFUSION EXPERIMENTS

Soil	Solution:Soil Ratio	<u>Soluble Cation (<math>\mu\text{g/mL}</math>)</u>				<u>Soluble Cations (<math>\mu\text{g/g}</math>)</u>			
		Ca	Mg	Na	K	Ca	Mg	Na	K
Chalk River Sand	0.19:1	48.0	6.0	14.0	6.2	9.1	1.1	2.7	1.2
0%*	0.19:1	38.3	4.7	7.2	10.6	7.3	0.9	1.3	2.0
5%	0.17:1	136.0	7.9	772.5	11.7	23.1	1.3	131.3	2.0
10%	0.25:1	286.0	17.8	1345.0	23.2	71.5	4.4	336.3	5.8
15%	0.38:1	305.0	16.8	1475.0	36.4	115.9	6.4	560.5	44.0
25%	0.54:1	387.0	21.5	1547.5	24.1	209.0	11.6	835.7	13.0
50%	1.00:1	433.6	23.9	1643.3	15.3	433.6	23.9	1643.3	15.3
100%	2.33:1	363.2	20.3	1602.1	21.2	846.3	47.3	3732.9	49.5

\* % bentonite in the bentonite-silica sand mixtures

diameter to provide for easy sample extraction. When assembled, the two syringes were held together by machine screws passing through holes drilled in the finger grips.

Plexiglass plugs, 2.0 cm long, were machined to fit inside each syringe. These were positioned along the barrel of the syringe according to the amount of soil available and the anticipated diffusion distances, and were held in place by stainless-steel pins passing through the syringe barrel (see Figure 3.2).

### 3.3.2 Soil Preparation

Tracer-free soils were prepared by adding a known volume of deionized water sufficient to saturate the soil. Tagged soils were saturated with solutions of either  $^3\text{H}$ ,  $^{36}\text{Cl}$  or  $^{85}\text{Sr}$ . The specific activity of the  $^3\text{H}$  solution used was approximately 10 microcuries\* per litre, and that of the  $^{36}\text{Cl}$  solution was approximately 25 microcuries per litre. The  $^{85}\text{Sr}$  solution used was the same as that used in the batch tests.

The tagged and tracer-free soil-solution mixtures were allowed to equilibrate for a minimum of two weeks in parafilm-covered plastic beakers. The soils were mixed with a spatula periodically to ensure equilibration.

### 3.3.3 Diffusion Cell Preparation

Following equilibration, the moisture content of each soil was determined by drying small duplicate samples to a constant weight at 105°C. Half-cells were weighed and the soil was then packed to achieve as high a bulk density as possible. Care was taken to ensure the same bulk density for tagged and tracer-free half-cells of the same soil, and to avoid air bubbles within the packed soil. Once packed, each half-cell was reweighed, and the mass of the soil determined. Problems were encountered when packing the Chalk River sand and the silica sand in that some solution would seep out between the plug and syringe wall. Attempts were made to correct this by using paraffin as a sealant.

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\*1 microcurie  $\equiv$  37 kilobecquerel



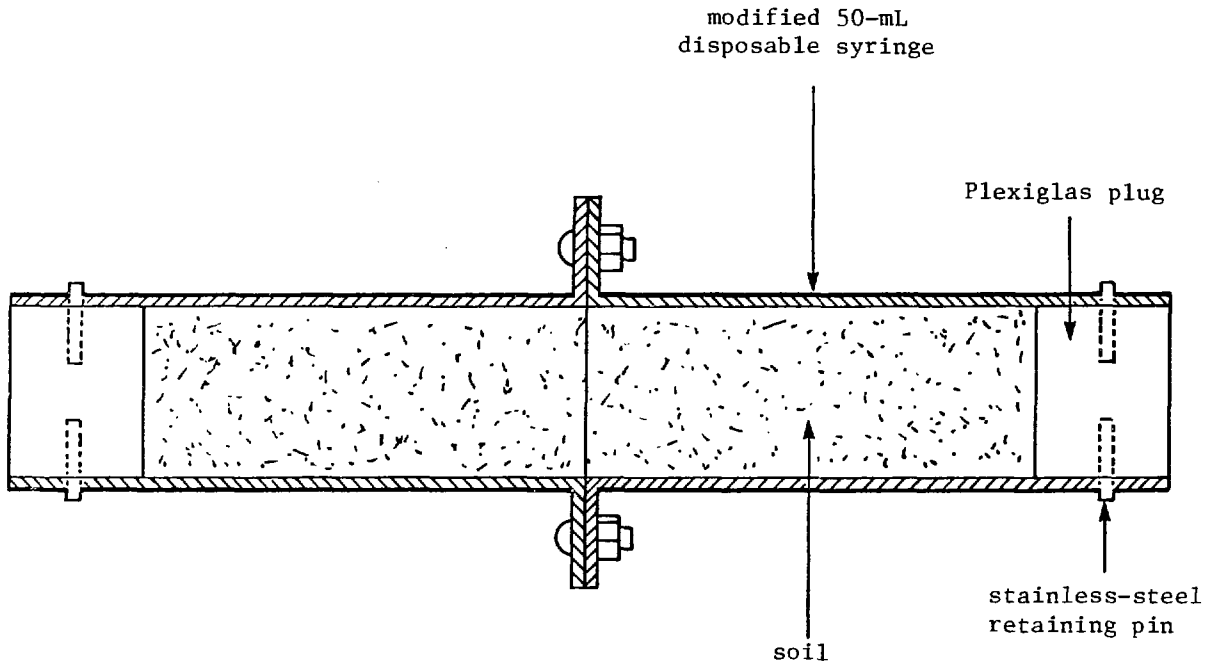


FIGURE 3.2: Schematic of an Assembled Diffusion Cell

In joining the tracer-free and tagged half-cells, Dow Corning high vacuum grease was added to the face of the moulded plastic grip of each syringe. The two half-cells were then brought together so that the soil surfaces made complete contact, and were held in place by bolts passing through the plastic grips.

To reduce moisture loss during the diffusion period, the assembled cells were dipped in warm paraffin wax. After the wax hardened, the cells were placed horizontally on racks in a controlled-environment chamber at  $22 \pm 1^\circ\text{C}$  where a high humidity was maintained by trays of water in the chamber.

#### 3.3.4 Sectioning, Extraction and Analysis

Estimates of the sectioning time (i.e. termination time of the diffusion experiment) were obtained from equation (8) using  $D_s = 9.1 \times 10^{-6} \text{ cm}^2/\text{s}$  for both non-reactive solutes. The  $D_s$  values for  $^{85}\text{Sr}$  were estimated by combining equations (4), (5), and (6) to give:

$$D_s = \tau D_o / (1 + \rho / \eta K_d) \quad , \quad (12)$$

and setting  $\tau = 0.67$ ,  $D_o = 1.3 \times 10^{-5} \text{ cm}^2/\text{s}$ ,  $\eta = 0.33$ , and using the  $\rho$  values calculated for each soil and the  $K_d$  values obtained from preliminary batch experiments. For all cases, the time (t) required to obtain  $c/c_o = 0.9$  at a distance (x) from the interface equal to 2/3 of the length of the half-cell was calculated from equation (8). The preliminary  $K_d$  values were determined using a solution-to-soil ratio of 5:1 and this gave  $K_d$  values substantially larger than those obtained from the procedure described above. As a result, in most cases the calculated diffusion times were too long, allowing the diffusion profiles to reach the ends of the cell. In these cases the medium could not be considered infinite and thus equation (10) was used to analyze the data rather than the simpler solution given by equation (8).

At the end of the diffusion experiment (i.e. at the termination time) the half-cells were separated. Each half-cell was then sectioned by removing the pins from the plexiglass plug and pushing the plug outward with the aid of a long threaded bolt. The apparatus was similar to that described in Johnston and Gillham, 1980. The bolt, turned by hand, extruded 0.254 cm of soil per revolution. To save time and cost, the number of sample sections analyzed per half-cell was limited to approximately 24. Section length was either 0.254 cm or 0.508 cm.

The soil was sectioned using a stainless-steel spatula run along the surface of the plastic grip. Samples were put into preweighed 20 mL glass scintillation vials, each containing 10 mL of water. For soils containing  $^{85}\text{Sr}$ , the vials contained 10 mL of 6 mol/L HCl instead of water. Acid extraction was used to release  $^{85}\text{Sr}$  from the exchange sites, and therefore concentrations obtained for  $^{85}\text{Sr}$  in the extract were in terms of total  $^{85}\text{Sr}$  in the sample. Whether the concentration of  $^{85}\text{Sr}$  is expressed in terms of  $^{85}\text{Sr}$  in solution only, or in terms of "total" concentration (i.e.  $^{85}\text{Sr}$  on the exchange complex plus  $^{85}\text{Sr}$  in solution) is immaterial because we assume  $K_d$  to be constant.

Two samples from each half-cell were taken to determine the moisture content. One sample was taken a few sections from the contact plane and the other was taken at the opposite end of the half-cell adjacent to the plug. Following extraction and equilibration, the supernatant solution in each glass vial was decanted into a Sorvall polyethylene centrifuge tube and centrifuged at 15 000 r/min for 20 minutes. Samples containing more than 25% bentonite usually required an additional decanting and centrifugation before an appropriate volume of clear solution could be recovered. After centrifuging, 5 mL of the supernatant solutions of the two non-reactive tracers were transferred, along with 10 mL of scintillation cocktail, into glass scintillation vials. The vials were allowed to cool for a few hours in a Mark III Automatic Liquid Scintillation Counter (Model 6880Z) and then counted to 10 000 counts.

The  $^{85}\text{Sr}$  was analyzed by transferring three mL of supernatant solutions to counting tubes, and counting to 10 000 counts, using a Nuclear-Chicago dual-channel, automatic gamma counter (Model 1085). The data for all solutes were then transformed to counts per minute per gram of soil, and plotted as a function of distance from the interface.

### 3.3.5 Normalized Concentration Profiles

In order to transform the data into normalized concentration profiles, we required values of  $c_0$ , the initial concentration in the tagged half-cell. There are several ways of obtaining  $c_0$  from the "absolute" concentration profiles. The usual and most straightforward ways are directly from the absolute concentrations at the smallest values of  $x$ , or to obtain and double the concentration at  $x \approx 0$  (the interface). For a combination of reasons, neither method could be used consistently for all experiments. Most concentration profiles showed enough scatter at low values of  $x$  to make direct reading difficult and, in many experiments, diffusion extended to the ends of the diffusion cells, in which case the first method is not applicable. In addition to the diffusion profile reaching the ends of the cell, the concentration profiles in many diffusion cells were asymmetrical so that neither method was applicable. Consequently,  $c_0$  was obtained as a mass balance on the concentration profiles, using the following equation:

$$c_0 = \left( \sum_{i=1}^n c_i g_i \right) / \left( \sum_{i=1}^{n_0} g_i \right) \quad (13)$$

where  $n$  is the total number of sample sections analyzed,  $n_0$  is the number of sections in the tagged half-cells,  $c_i$  is the concentration in the  $i$ th section (in cpm per gram of soil), and  $g_i$  is the number of grams of soil in the  $i$ th section.

#### 4. RESULTS AND DISCUSSION

##### 4.1 RETARDATION PARAMETERS

The porosity and bulk density for each diffusion cell are given in Table 4.1. It can be seen that, for a given soil, they did not vary appreciably from cell to cell. Variations were also very slight within each cell (data not shown). One may recall (see Section 3) that each half-cell was packed to as high a bulk density as possible. The data indicate that this packing technique yielded consistent results for a given soil, and that the bulk density and porosity varied in a consistent manner with clay content.

The porosity and bulk density values of the sand-bentonite mixtures are plotted as a function of bentonite content in Figure 4.1. The porosity decreased from about 0.34 at 0% bentonite to 0.30 at 5% and then increased to 0.84 at 100% bentonite. Porosities of fine-textured soils are usually higher than those of coarser-textured soils, although the size of the individual pores may be larger in the coarser material. The initial decrease can be attributed to clay-size particles filling the pore space between the sand grains. As the proportion of clay increased, the porosity reached a minimum when all the large pores between the sand grains were occupied by the clay particles. The porosity increased again as the clay occupied more space than that provided by the larger pores of the sand.

As expected, the bulk densities followed the opposite trend, increasing from about  $1.64 \text{ g/cm}^3$  at 0% bentonite to about  $1.73 \text{ g/cm}^3$  at 5%, and then decreasing to  $0.39 \text{ g/cm}^3$  at 100% bentonite.

Batch distribution coefficients ( $K_d$ ) were measured for  $^{85}\text{Sr}$  using soil-to-solution ratios similar to those measured in the diffusion cells. The results are given in Table 4.2, and are plotted as a function of bentonite content in Figure 4.2. The average  $K_d$  value obtained for the Chalk River sand (4.85 mL/g) agrees very well with values reported earlier

TABLE 4.1

POROSITIES AND BULK DENSITIES FOR ALL DIFFUSION CELLS\*

Soil	Repli- cates	<u>Chlorine-36</u>		<u>Tritium</u>		<u>Strontium-85</u>	
		Porosity (cm <sup>3</sup> /cm <sup>3</sup> )	Bulk Density (g/cm <sup>3</sup> )	Porosity (cm <sup>3</sup> /cm <sup>3</sup> )	Bulk Density (g/cm <sup>3</sup> )	Porosity (cm <sup>3</sup> /cm <sup>3</sup> )	Bulk Density (g/cm <sup>3</sup> )
Chalk River Sand	1	0.32	1.70	0.33	1.68	0.32	1.65
	2	0.32	1.67	0.32	1.68	0.32	1.67
0%**	1	0.34	1.62	0.35	1.64	0.31	1.61
	2	0.34	1.64	0.33	1.65	0.34	1.65
5%	1	0.30	1.72	0.31	1.73	0.29	1.73
	2	0.31	1.79	0.30	1.74	0.29	1.72
10%	1	0.40	1.57	0.40	1.60	0.41	1.59
	2	0.39	1.58	0.42	1.57	0.36	1.63
15%	1	0.51	1.32	0.50	1.32	0.50	1.35
	2	0.50	1.32	0.51	1.32	0.50	1.34
25%	1	0.60	1.07	0.59	1.07	0.59	1.07
	2	0.59	1.08	0.59	1.07	0.59	1.06
50%	1	0.75	0.72	0.73	0.72	0.73	0.72
	2	0.73	0.72	0.74	0.73	0.74	0.73
100%	1	0.86	0.39	0.86	0.39	0.86	0.39
	2	0.86	0.39	0.86	0.39	0.86	0.39

\* Each value listed is the average of four separate values: two sub-samples per half-cell (four sub-samples per diffusion cell).

\*\* % bentonite in the bentonite-silica sand mixture.

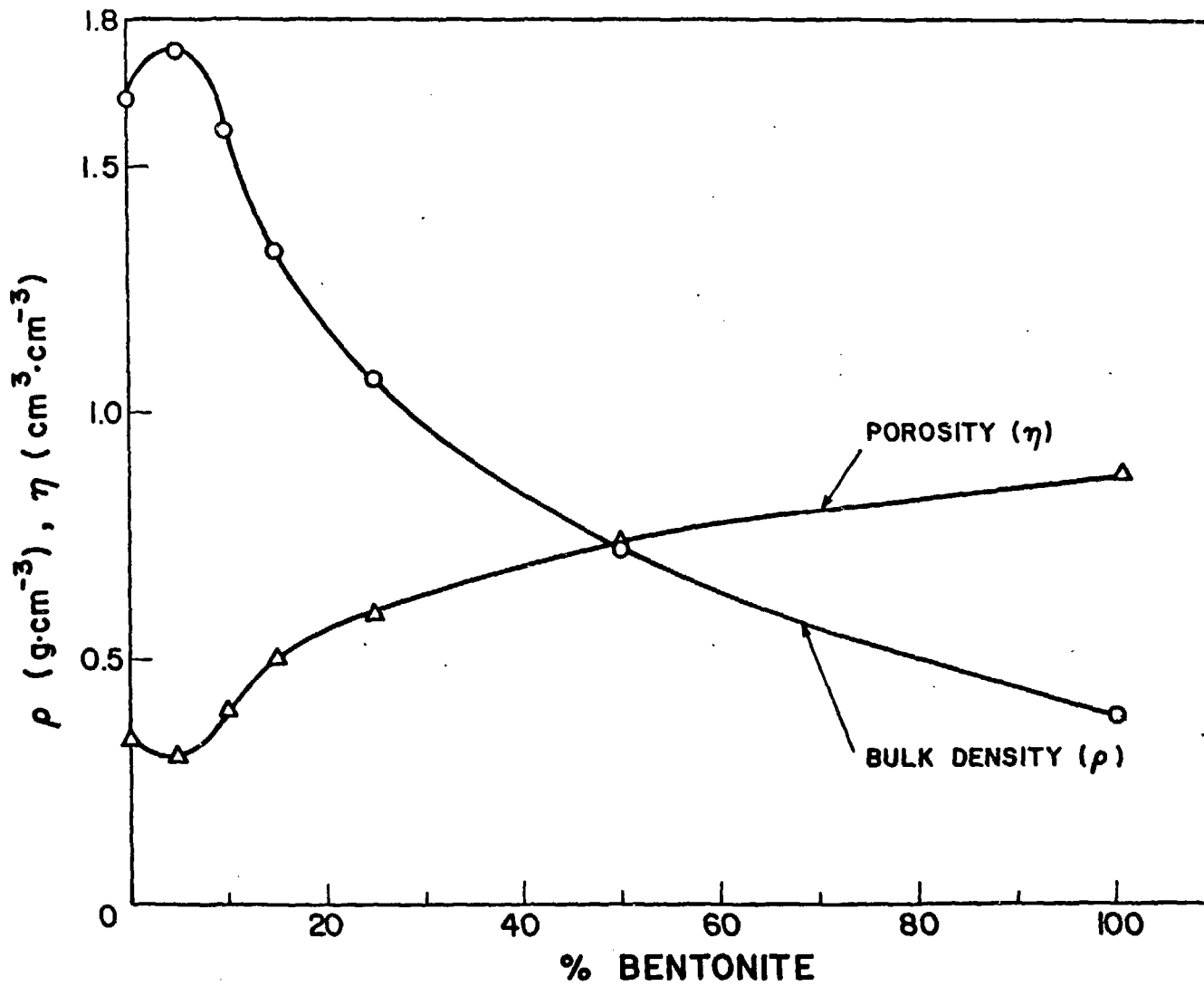


FIGURE 4.1: Porosity and Bulk Density of Sand-Bentonite Mixtures versus Bentonite Content

TABLE 4.2

BATCH DISTRIBUTION COEFFICIENTS,  $\rho/\eta$  RATIOS AND  
RETARDATION FACTORS (R)\* FOR STRONTIUM-85

Soil	Repli- cates	Batch $K_d$ (mL/g)		$\rho/\eta$ (g/mL)		$R = 1 + (\rho/\eta) K_d$	
Chalk River Sand	1	5.00	4.85	5.16	5.19	25.98	26.13
	2	4.69		5.22			
0%**	1	0.26	0.29	5.19	5.02	2.48	2.43
	2	0.31		4.85			
5%	1	2.36	2.25	5.97	5.95	14.39	14.36
	2	2.13		5.93			
10%	1	3.30	3.59	3.88	4.21	14.93	16.09
	2	3.88		4.53			
15%	1	-	4.10	2.70	2.69	12.07	12.04
	2	4.10		2.68			
25%	1	5.10	5.10	1.81	1.81	10.24	10.20
	2	5.09		1.80			
50%	1	11.46	11.68	0.99	0.99	12.52	12.52
	2	11.90		0.99			
100%	1	-	20.85	0.45	0.45	10.46	10.46
	2	20.85		0.45			

\* The retardation factors were calculated using the  $\rho/\eta$  ratio of the given cell and the average  $K_d$  value obtained for that soil.

\*\* % bentonite in the bentonite-silica sand mixture.



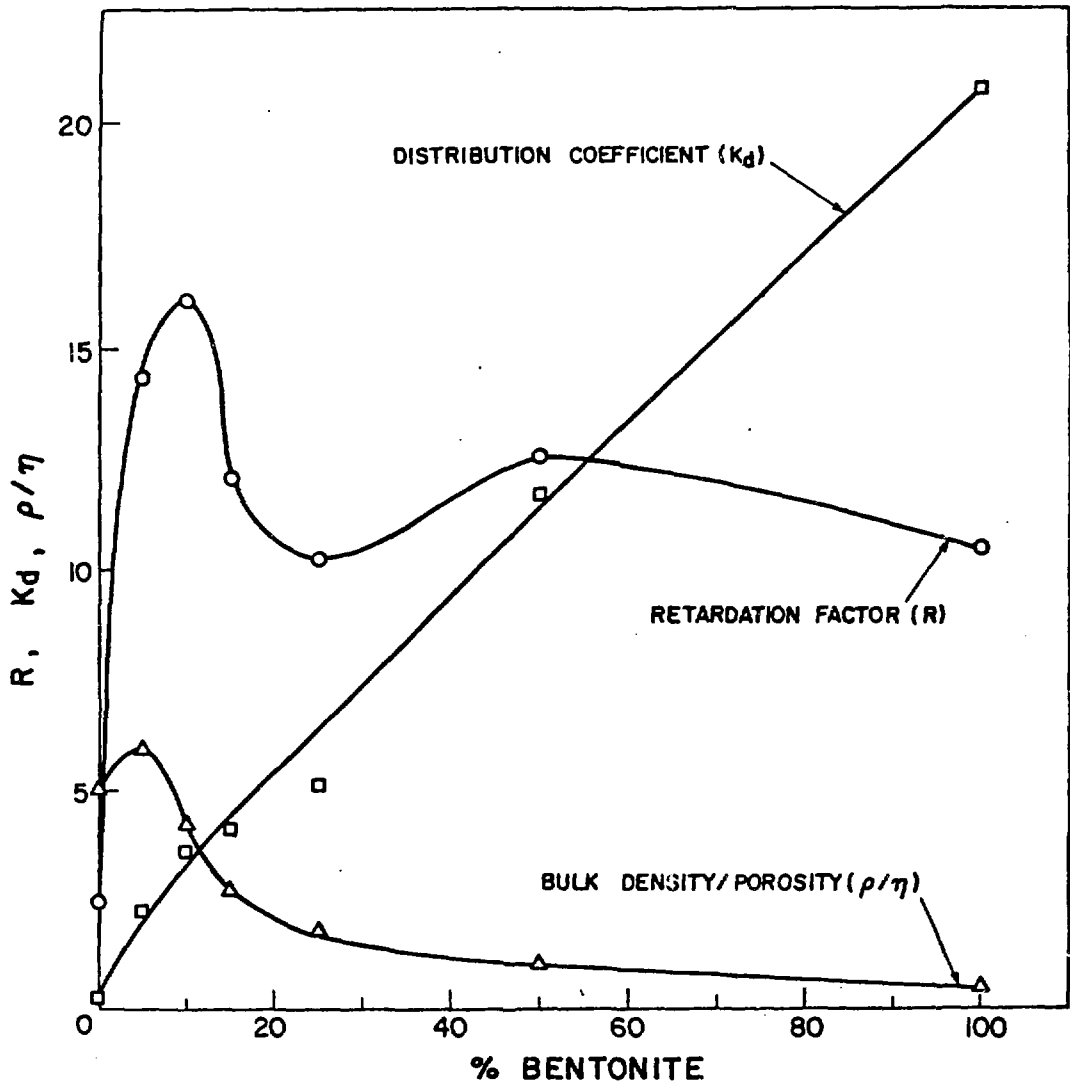


FIGURE 4.2: Strontium-85 Distribution Coefficients, Bulk Density-to-Porosity Ratios, and Retardation Factors versus Bentonite Content

(Johnston and Gillham, 1980): 6.7 mL/g for batch  $K_d$  values obtained using a different soil-to-solution ratio, and 4.9 mL/g for  $K_d$  values obtained from diffusion experiments. As expected, the  $K_d$  values increased with clay content, with values ranging from 0.29 mL/g at 0% bentonite to 20.85 mL/g at 100% bentonite.

Also presented in Table 4.2 and Figure 4.2 are values of  $\rho/\eta$  for the diffusion cells containing  $^{85}\text{Sr}$ , and the calculated retardation factors (R). Equation (5) shows that the retardation factor is proportional to the distribution coefficient ( $K_d$ ) and to the bulk density ( $\rho$ ), but inversely proportional to the porosity ( $\eta$ ). Although  $K_d$  increased by about seventy times as the bentonite content increased from 0% to 100%, the bulk density decreased and the porosity increased with clay content (see Table 3.1). As shown in Table 4.2 and Figure 4.2, the resulting bulk density to porosity ratios increased from about 5.0 at 0% bentonite to about 5.9 at 5% bentonite, then decreased to 0.45 at 100%. The resulting R values increased six-fold, from about 2.45 at 0% bentonite to 14.35 at 5% bentonite, and then decreased in an irregular manner to a value of 10.46 at 100% bentonite.

It is interesting to note that in the 5% to 100% bentonite range, the bulk density-to-porosity ratio affected the R values to about the same degree as  $K_d$ . In fact, an increase of almost an order of magnitude in  $K_d$  was counterbalanced by the decrease in the bulk density-to-porosity ratio. For coarse-grained geological materials, it is frequently assumed that the bulk density-to-porosity ratio varies within relatively narrow limits (two to six). The above results show that this may not be the case for repacked fine-textured materials, and variations in the ratio may have a significant effect on the retardation factor.

#### 4.2 TORTUOSITY FACTORS

Tortuosity factors were determined from the results of the non-reactive solute-diffusion experiments. The data for some of the diffusion cells were discarded when air entrapment at the interface was obvious. An

example of a non-reactive diffusion profile is shown in Figure 4.3 for tritium in the 50% bentonite mixture.

It is apparent from Figure 4.3 that the experimental data showed a fair amount of scatter, particularly in the tagged half-cell, where the concentrations were higher. This trend was present in all diffusion cells, and it is difficult to assess whether it was due to physical aberrations, or the result of the extraction and/or analysis procedures. The scatter caused problems in the estimation of  $c_0$  (see Section 3.4) and in the estimation of diffusion coefficients. Generally, the data showed a high degree of symmetry about  $x = 0$ ,  $c/c_0 = 0.5$ , for all non-reactive diffusion cells.

The diffusion coefficient for each non-reactive diffusion cell was determined by superimposing several "calculated diffusion profiles" onto the experimental data. Calculated profiles were obtained from equation (10) by varying the diffusion coefficient. The coefficient giving the best "eye-fit" of the experimental data was taken as the "experimental" diffusion coefficient. More weight was placed on the data at positive values of  $x$ , where the scatter was usually less, than on the tagged half-cell side of the interface. This procedure is illustrated in Figure 4.3 for tritium in the 50% bentonite mixture where a value  $D = 1.1 \times 10^{-5} \text{ cm}^2/\text{s}$  gives the best fit.

Tortuosity factors were calculated for each non-reactive diffusion cell as the ratio of the experimental diffusion coefficient of the solute in the soil ( $D$ ), to the molecular diffusion coefficient of the solute in free solution ( $D_0$ ) (see equation (6)). The values of  $D_0$  used were  $1.19 \times 10^{-5} \text{ cm}^2/\text{s}$  for  $^{36}\text{Cl}$ , which is the molecular diffusion coefficient of 0.1 mol/L  $\text{CaCl}_2$  at 25°C (Handbook of Chemistry and Physics, 1975) and  $2.44 \times 10^{-5} \text{ cm}^2/\text{s}$  for tritium (Klitzsche et al., 1976). The value for tritium was the only one found in the literature, and its accuracy is uncertain.

The values of the experimental diffusion coefficient and tortuosity factor obtained for each diffusion cell of the non-reactive solutes

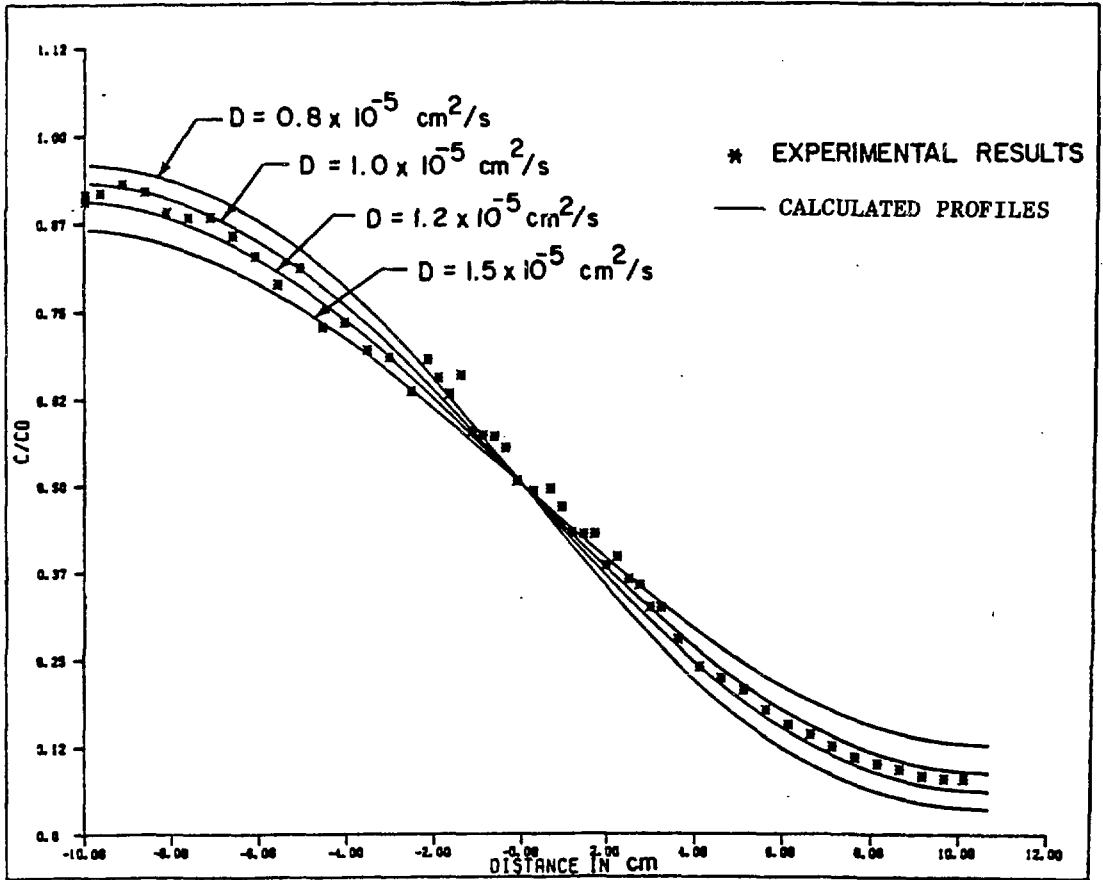


FIGURE 4.3: Diffusion Data for  $^3\text{H}$  in the 50% Bentonite Mixture

are presented in Table 4.3. It is important to point out that, because of the scatter in the data, and because of the moderate insensitivity of the calculated diffusion profiles to small variations in  $D$ , the values of the experimental diffusion coefficient, and thus of the tortuosity factors, carry an uncertainty of about  $\pm 10\%$ .

In spite of the uncertainty, slight trends are evident. As can be seen from Table 4.3, the average tortuosity factors for  $^{36}\text{Cl}$  first decreased from 0.74 to 0.59 as the clay content increased from 0% to 10% bentonite and then increased to 0.84 at 100%. The trend was similar for tritium. This behaviour is expected, if one views the tortuosity factor as the ratio of the net distance travelled in the porous medium to the length of the tortuous pathway followed by the molecules in the medium. At 0% bentonite, the molecules have to diffuse around the sand grains through fairly large pores. When a small amount of clay is added, the clay particles occupy the space between the sand grains and the length of the molecular pathway increases. As the clay content increases further, the effect of the sand grains decreases, and the pathlength decreases.

The observation that the trends were similar for the two non-reactive solutes as a function of clay content indicates that they behaved in a similar manner in the soil. Therefore, for these experimental conditions, phenomena such as anion-exclusion and anion-adsorption did not seem to affect the diffusion of chloride to a noticeable extent. The fact that the tortuosity factors were usually lower for  $^3\text{H}$  than for  $^{36}\text{Cl}$  may be due to the uncertainty in the free-solution diffusion coefficient ( $D_0$ ) for tritium.

#### 4.3 CALCULATED AND EXPERIMENTAL STRONTIUM-85 DIFFUSION COEFFICIENTS

The tortuosity factors obtained from the  $^{36}\text{Cl}$  and the  $^3\text{H}$  diffusion experiments were used in equation (12) to calculate values of the  $^{85}\text{Sr}$  diffusion coefficient ( $D_g$ ), for each soil. The other parameters used in equation (12) were: the measured porosity and bulk density in the  $^{85}\text{Sr}$  diffusion cells; the average  $K_d$  values obtained from the batch tests for

TABLE 4.3

EXPERIMENTAL DIFFUSION COEFFICIENTS AND TORTUOSITY FACTORS  
FOR THE NON-REACTING SOLUTES <sup>36</sup>Cl and <sup>3</sup>H

Soil	Repli- cates	<u>Chlorine-36</u>			<u>Tritium</u>		
		Experimental Diffusion Coefficient (10 <sup>-5</sup> cm <sup>2</sup> /s)	Tortuosity Factor		Experimental Diffusion Coefficient (10 <sup>-5</sup> cm <sup>2</sup> /s)	Tortuosity Factor	
Chalk River Sand	1	1.0	0.84	0.84	1.4	0.57	0.57
	2	1.0	0.84		1.4	0.57	
0%*	1	1.0	0.63	0.74	1.8	0.72	0.67
	2	0.9	0.84		1.5	0.61	
5%	1	0.7	0.59	0.62	-	-	0.33
	2	0.8	0.65		0.8	0.33	
10%	1	0.7	0.59	0.59	0.9	0.37	0.36
	2	0.7	0.59		0.9	0.35	
15%	1	0.7	0.59	0.61	1.0	0.41	0.39
	2	0.8	0.63		0.9	0.37	
25%	1	0.7	0.59	0.63	1.0	0.41	0.41
	2	0.8	0.67		1.0	0.41	
50%	1	-	-	0.76	1.1	0.45	0.45
	2	0.9	0.76		1.1	0.45	
100%	1	-	-	0.84	1.2	0.49	0.48
	2	1.0	0.84		1.2	0.47	

\* % bentonite in the bentonite-silica sand mixture

each soil; and a value of  $1.3 \times 10^{-5} \text{ cm}^2/\text{s}$  for the molecular diffusion coefficient of  $^{85}\text{Sr}$  in free solution. The tortuosity factors for  $^{36}\text{Cl}$ , the retardation factors and the resulting  $^{85}\text{Sr}$  diffusion coefficients are plotted in Figure 4.4. The results of the same calculation using the  $^3\text{H}$  tortuosities are given in Figure 4.5. It is interesting to note from the shape of these graphs that the calculated diffusion coefficient ( $D_g$ ) was influenced very strongly by the retardation factor (R), but somewhat less by the tortuosity factor. This was expected, since R was usually higher than 10, making  $1/R$  less than 0.1, whereas the tortuosity factor was usually higher than 0.3. It should be pointed out that the calculated values of  $D_g$  carry, through the tortuosity factor, the uncertainties of the experimental diffusion coefficients of the non-reactive solutes.

An example of an experimental diffusion profile for  $^{85}\text{Sr}$  is given in Figure 4.6, for the 25% bentonite mixture. The same general observations may be made for these profiles as were made for those of the non-reactive solutes; that is, the scatter was usually more pronounced at the higher concentration values (tagged half-cells), and the data were usually fairly symmetrical about the point  $x = 0$ ,  $c/c_o = 0.5$  for symmetrical cells. This symmetry is expected since the reaction term is assumed to be a function of neither concentration nor time.

Experimental values of the diffusion coefficient of  $^{85}\text{Sr}$  for each diffusion cell were obtained as for the non-reactive solutes by "eye-fitting" simulated curves to the experimental data. More weight was placed on the data at positive values of  $x$ . The experimental diffusion coefficient values thus carry an uncertainty of about 5-10% for the same reasons as explained for the non-reactive solutes. The simulated concentration profile judged to give the best fit to the data is that given by the solid line in Figure 4.6. Also presented in these figures are the concentration profiles obtained using each of the two calculated diffusion coefficients (dashed lines). The experimental and calculated coefficients are given in Table 4.4 for all soils, and the averages of those for the sand-bentonite mixtures are plotted versus clay content in Figure 4.7.

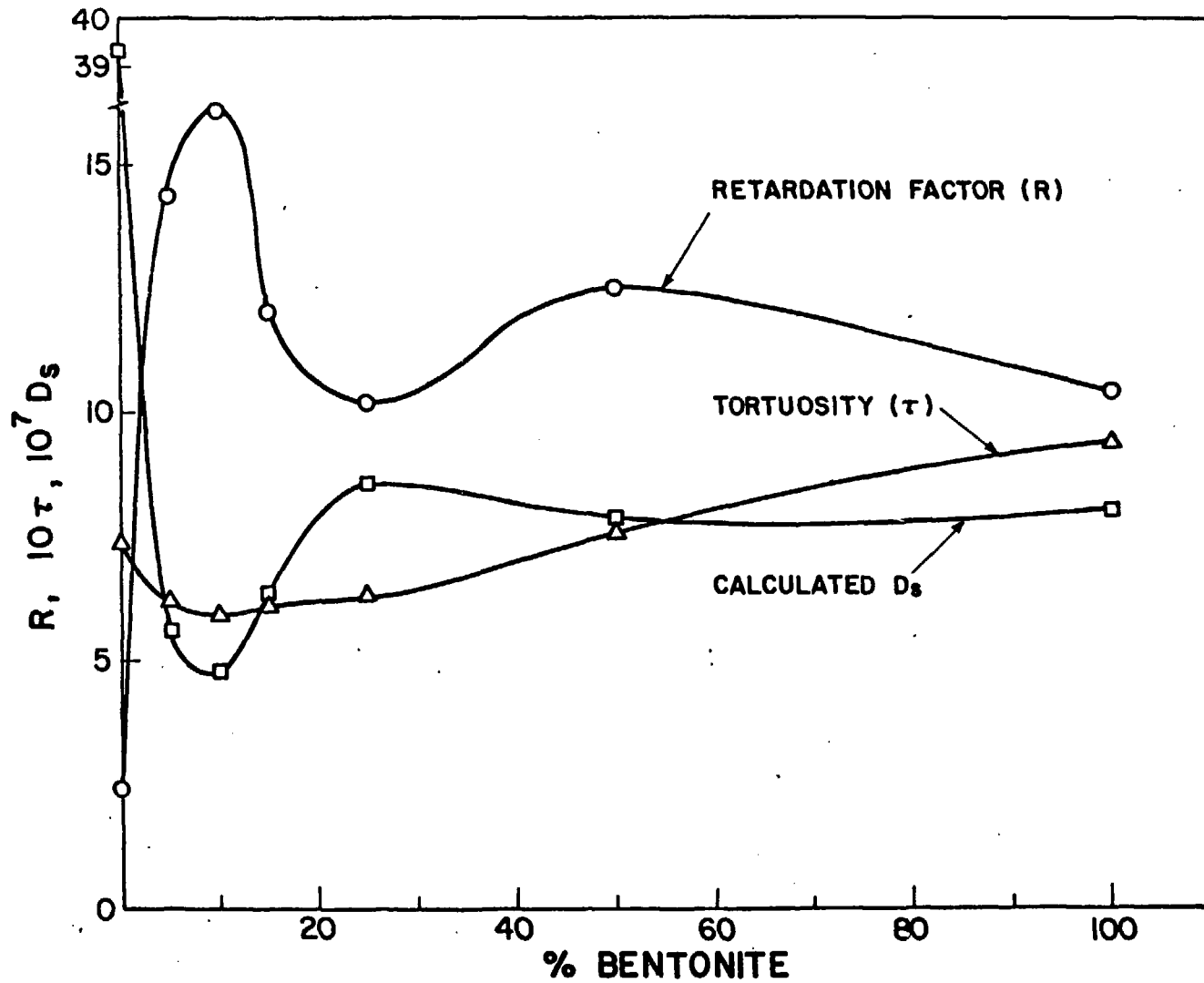


FIGURE 4.4: Retardation Factors,  $^{36}\text{Cl}$  Tortuosity Factors, and Calculated  $D_s$  Values for  $^{85}\text{Sr}$  versus Bentonite Content



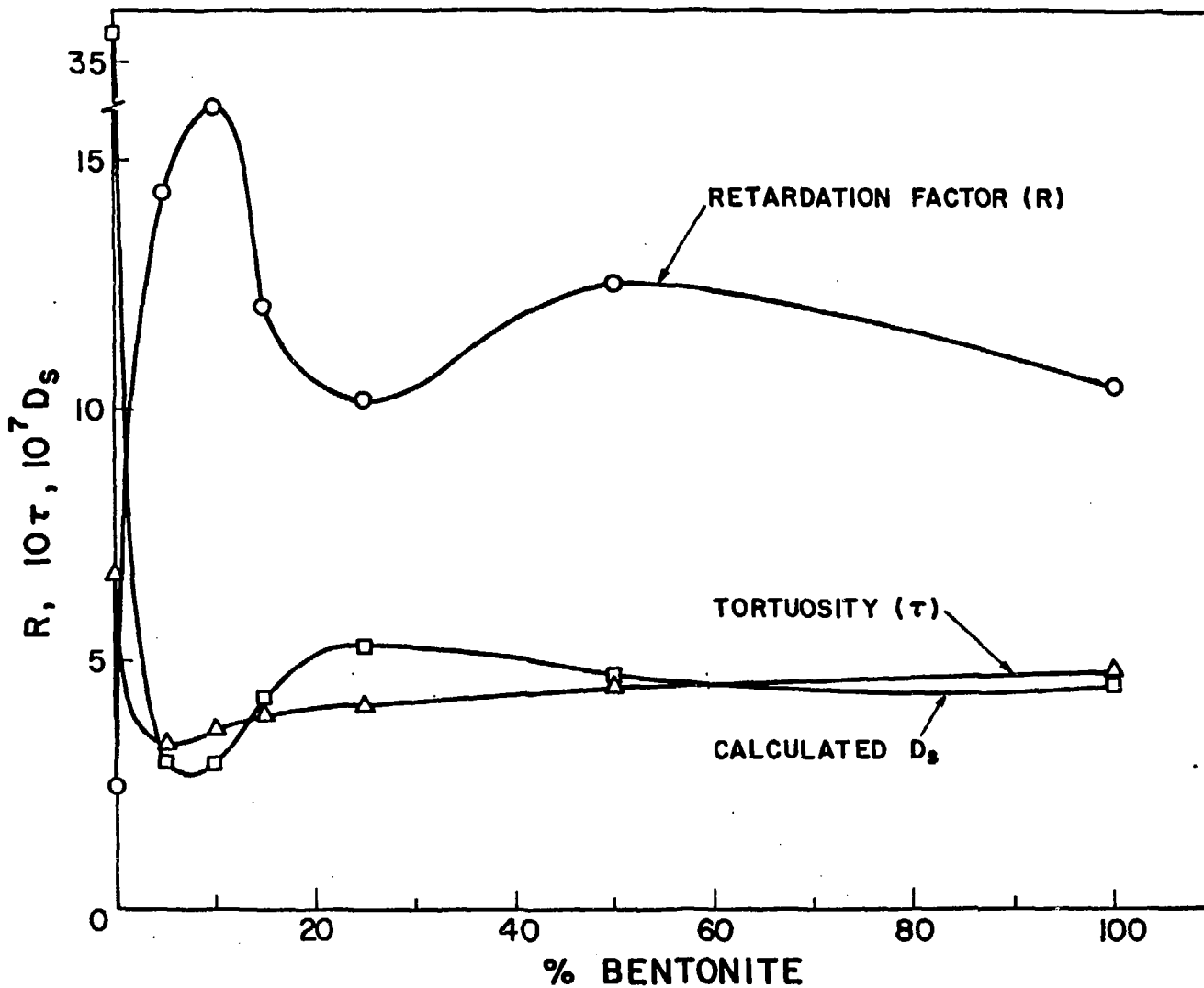


FIGURE 4.5: Retardation Factors,  $^3\text{H}$  Tortuosity Factors, and Calculated  $D_s$  Values for  $^{85}\text{Sr}$  versus Bentonite Content

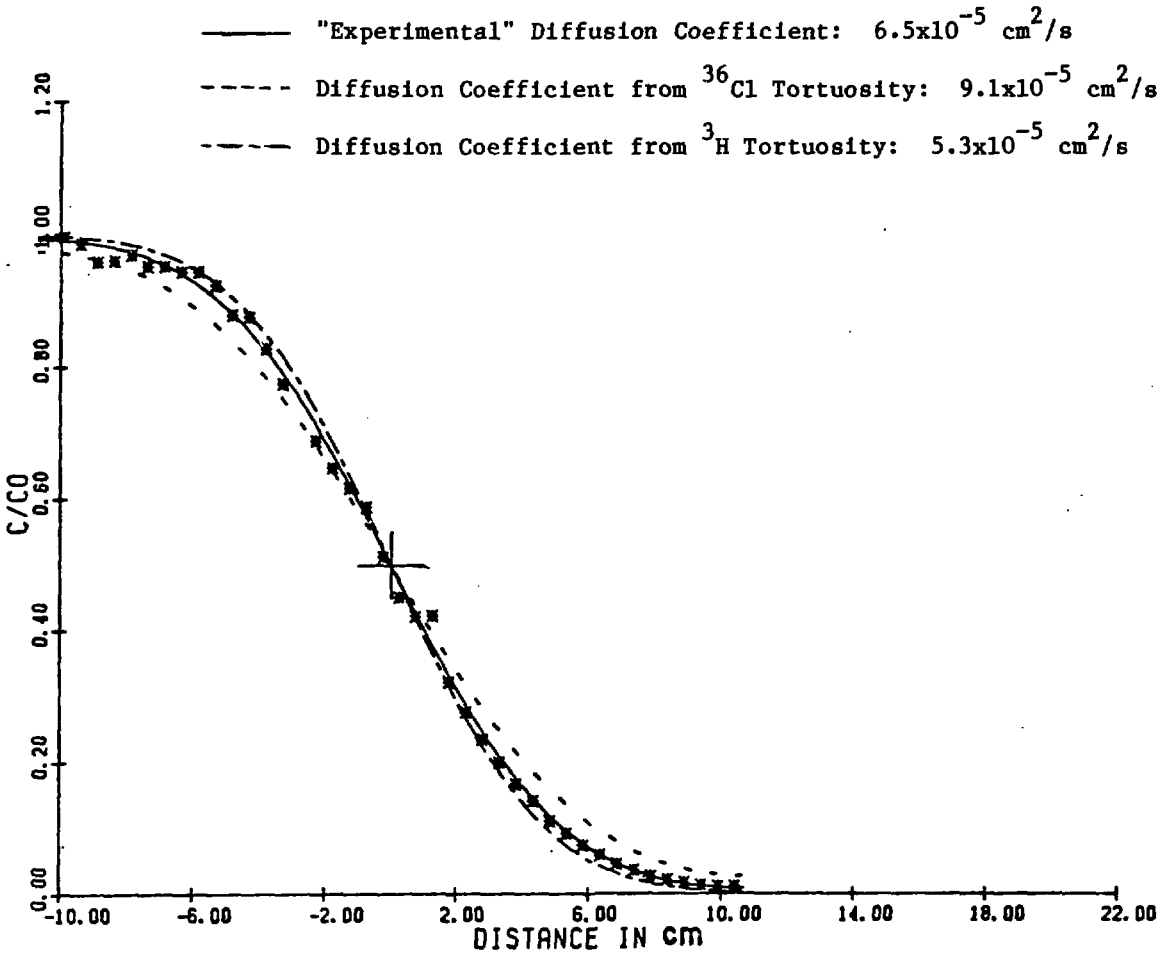


FIGURE 4.6: Diffusion Data for  $^{85}\text{Sr}$  in the 25% Bentonite Mixture

TABLE 4.4

EXPERIMENTAL AND CALCULATED DIFFUSION COEFFICIENTS FOR STRONTIUM-85

Soil	Repli- cates	Experimental Diffusion Coefficient ( $10^{-7}$ cm <sup>2</sup> /s)		Calculated Diffusion Coefficient ( <sup>36</sup> Cl tortuosity) ( $10^{-7}$ cm <sup>2</sup> /s)		Calculated Diffusion Coefficient ( <sup>3</sup> H tortuosity) ( $10^{-7}$ cm <sup>2</sup> /s)	
Chalk River Sand	1	0.5	0.6	4.18	4.14	2.80	2.79
	2	0.6		4.09		2.78	
0%	1	20.0	20.0	38.74	39.35	35.06	35.61
	2	20.0		39.96		36.15	
5%	1	4.0	4.0	5.65	5.60	3.01	2.98
	2	-		5.54		2.95	
10%	1	5.0	5.0	5.09	4.78	3.11	2.92
	2	5.0		4.47		2.73	
15%	1	5.5	5.8	5.81	6.34	4.21	4.23
	2	6.0		9.12		5.28	
25%	1	-	6.5	8.06	8.59	5.24	5.26
	2	6.5		9.12		5.28	
50%	1	7.0	6.8	7.92	7.91	4.69	4.68
	2	6.5		7.89		4.67	
100%	1	7.0	7.3	5.72	8.03	3.27	4.59
	2	7.05		10.34		5.91	

\* % bentonite in the bentonite-silica sand mixture

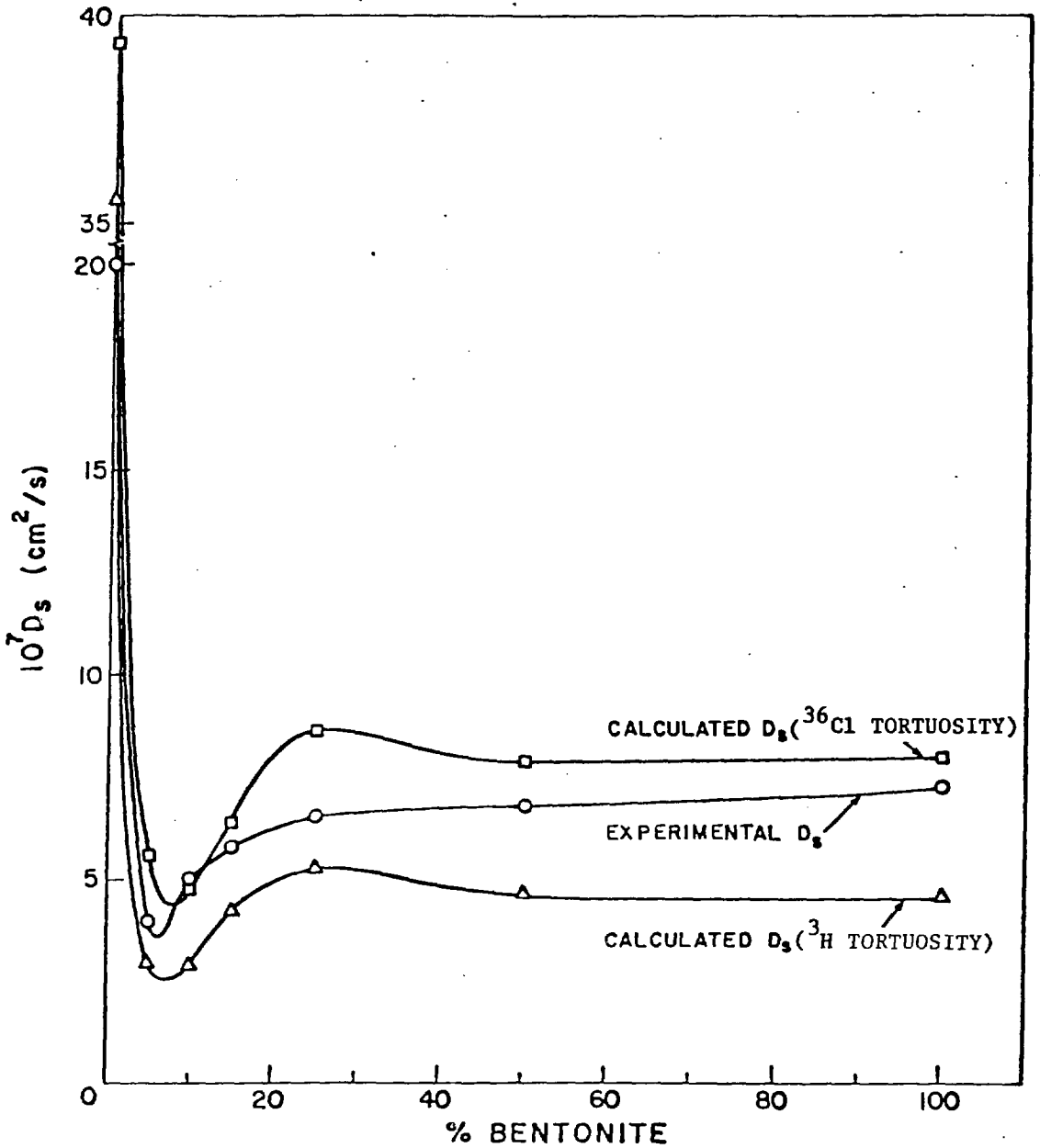


FIGURE 4.7: Strontium-85 Diffusion Coefficients: Experimental and Calculated Using  $^{36}\text{Cl}$  and  $^3\text{H}$  Tortuosities versus Bentonite Content

From Table 4.4 it can be seen that the experimental values of the diffusion coefficient for the Chalk River sand are very low; i.e. five to ten times lower than predicted, and at least one order of magnitude lower than those obtained for any of the sand-bentonite mixtures. This does not appear to be related to the  $K_d$  value, since the values obtained for the Chalk River sand were of the same order of magnitude as those of the other soils. The tortuosity factors obtained from the non-reacting solutes were somewhat high, but not by enough to cause this large discrepancy. A plausible explanation may be the incomplete saturation of the half-cells with water, which would reduce the area for diffusion, and could induce water movement. This might also explain the large discrepancy (a factor of almost two) between the calculated and experimental values of the diffusion coefficients for the silica sand (0% bentonite). This possibility is strongly supported by the fact that problems were encountered when packing the sands into the respective half-cells (see Section 3).

Table 4.4 and Figure 4.7 show that the experimental and calculated diffusion coefficients behaved in a similar manner with increasing clay content. In all cases, the diffusion coefficients decreased from high values (greater than  $20 \times 10^{-7} \text{ cm}^2/\text{s}$ ) at 0% bentonite to a minimum of less than  $5 \times 10^{-7} \text{ cm}^2/\text{s}$  between 5% and 10% bentonite. The diffusion coefficients then increased and leveled off between 50% and 100% bentonite. At 100% bentonite, the experimental  $D_s$  value and the average of the  $D_s$  values calculated from the  $^{36}\text{Cl}$  tortuosity factors were about 7.0 and  $8.0 \times 10^{-7} \text{ cm}^2/\text{s}$ , respectively, and about  $4.5 \times 10^{-7} \text{ cm}^2/\text{s}$  for the  $D_s$  calculated from the  $^3\text{H}$  tortuosity factors. Between 5% and 100% bentonite one would expect the diffusion coefficient to decrease as the clay content increased because the cation-exchange capacity, and thus the  $K_d$  value of the porous material, would increase. However, because of the strong influences of bulk density and porosity on the retardation factor (see Section 4.3.2), the opposite trend was observed. It would seem that, if a maximum  $^{85}\text{Sr}$  retardation (i.e. a minimal diffusion coefficient) is desired, a mixture containing 5% to 10% bentonite would be preferable to either pure sand or a mixture containing more than 10% bentonite. Improved retardation at higher bentonite contents could only be realized in situations where higher degrees of compaction could be achieved.

Except for the Chalk River sand, the agreement between the experimental data and the concentration profiles generated from the calculated diffusion coefficients was very good. The agreement was usually somewhat better with the diffusion coefficient values calculated using the  $^{36}\text{Cl}$  tortuosity factors than with those using  $^3\text{H}$  tortuosity values. As noted previously, this may be due to the uncertainty in the molecular diffusion coefficient of  $^3\text{H}$  in free solution used to calculate the tritium tortuosity factors.

In spite of the differences between the calculated and experimental diffusion coefficients, and except for the two sands, the calculated concentration profiles showed good agreement with the experimental data. This good fit is at least partly due to the insensitivity of the concentration profiles to relatively small variations in the diffusion coefficient. It should be pointed out that using the commonly assumed value of 0.67 for the tortuosity factor would have given equally good results, since 0.67 is close to the tortuosity factors calculated for the two non-reactive solutes.

#### 4. CONCLUSIONS

The results of this study indicate that, for practical purposes, distribution coefficients ( $K_d$ ) provide a convenient and simple means of estimating the reaction term of the diffusion equation for  $^{85}\text{Sr}$  in sand-bentonite mixtures. One would expect similar behaviour for other solutes and soil materials in which the reaction with the porous medium can be described by a distribution coefficient. It would be desirable to attempt to validate the use of  $K_d$  for a wider range of reactive solutes and soil materials.

The results also showed that a small amount of clay mixed with sand (5-10% bentonite) was more effective in retarding  $^{85}\text{Sr}$  than either pure silica sand or mixtures of higher clay content. As the clay content

increased from 5% to 100%, even though the soils were packed as tightly as possible, within the experimental constraints of this study the effect of increasing porosity and decreasing bulk density on the retardation factor countered the effect of a nine-fold increase in the distribution coefficient. This could have significant implications for the design of optimum buffer and backfill materials.

The insensitivity of the calculated concentration profiles to variations in the diffusion coefficient was such that, in many cases, variations of as much as two-fold still produced profiles giving reasonable fits to the experimental data. The tortuosity factors obtained from the non-reacting solute diffusion experiments did not vary considerably and were close to 0.67, the commonly assumed value. Based on this fact and on previous observations, it appears that, for practical purposes, using the value 0.67 as a tortuosity factor, together with the diffusion coefficient of  $^{85}\text{Sr}$  in free solution, and the retardation factor, could yield very satisfactory predictions of the diffusion coefficient.

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