

Diffusion of Water in Bentonite

by

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Abstract

Because of the low permeability and the high sorption coefficient, compacted sodium bentonite has been proposed as an effective backfill material in the repository of high-level radioactive waste. The only possible mechanism of nuclide transport in bentonite must be the diffusion. The diffusivity has been considered to have a close relationship with the retardation factor and the self-diffusivity of water. Then, we performed the experiments to determine the self-diffusivity of water in bentonite by using tritiated water as a tracer. The obtained self-diffusivity can be written as

$$Da = (3.4 \pm 0.2) \times 10^{-6} \text{ [cm}^2\text{/s]}$$

under the condition of temperature $T = 25^\circ\text{C}$ and compacted density of bentonite $\rho = 2.7 \text{ [g/cm}^3\text{]}$. It was revealed quantitatively that the self-diffusivity of water in bentonite is much smaller than that of free water ($2.13 \times 10^{-5} \text{ [cm}^2\text{/s]}$ at $T = 25^\circ\text{C}$).

Also discussed is the analysis of experimental results on the basis of a new solution derived for the diffusion in one dimensional finite zone.

1. Introduction

Compacted sodium bentonite has been proposed as an effective backfill material which surrounds the waste canister in the repository of high-level radioactive waste¹⁻⁴⁾. The backfill material is expected to form a low-permeability barrier against the flow of groundwater and to retard the migration of radio nuclides by sorption.

The permeability of water through bentonite is so low that the only possible mechanism of nuclide transport must be the diffusion. The diffusivities of some ions in bentonite, Da , have been measured by previous investigators⁵⁻⁹⁾,

$$Da = D/\alpha \left(1 + \frac{\rho}{\epsilon} K_d \right), \quad (1)$$

where D , K_d , α , ϵ and ρ denote the diffusivity of the ion in free water, distribution coefficient, tortuosity, void fraction and packing density, respectively.

Assuming the validity of the above relation, the diffusivity of water in bentonite would be almost the same as the self diffusivity of free water. The water in compacted bentonite, however, is thought to be composed of the surface water, interlayer water and interstitial water¹⁰⁾. The surface water refers to the first few layers of water on a particle surface, whose properties are highly influenced by the solid surface. The diffusion in surface water must be much restricted in comparison with that in free water. Then, the self-diffusion of water in bentonite will be possibly different from that in free water.

In the study on the deep geologic disposal, the engineering barrier becomes more and more important in these days. The diffusive migration of various nuclides will be one of the key subjects for the safety assessment. The diffusion of water is important as the basis of the transport of radionuclides in bentonite.

We made diffusion cells which resemble those of B. Torstenfelt⁵⁾, M. Kumagai⁶⁾, and E. J. Nowak⁷⁾. Tritiated water was used as a tracer. The analysis of the experimental results was performed on the basis of the solution derived for the diffusion in one dimensional finite zone.

2. Experimental

Equipments for the study on the diffusion of tritiated water in compacted bentonite were manufactured with reference to those of previous researchers. By using the equipments, the compaction of bentonite, saturation of water, diffusion of tritiated water, and the cutting into thin slices were carried out successfully. The diffusion cells made of stainless steel were prepared as shown in Fig. 1. The columns of compacted bentonite were 4 cm long and 2 cm in diameter. Tritiated water was spiked on the center surface of the column. After a predetermined time interval, the bentonite column was sectioned and the tritium content was analyzed to determine the concentration profile. Sectioning was accomplished by forcing a column out of its diffusion tube in 1 mm increments and cutting off each exposed section with a sharp blade. Tritium was analyzed by means of a liquid scintillation counter. Experiments were performed at room temperature ($\sim 25^\circ\text{C}$).

The homogeneity of the packed density and the full saturation of water was verified for a few columns by the weight measurements of the sliced sections as shown in Fig. 2. It shows also that the diffusion column can be cut into sections of almost the same width by a sharp blade.

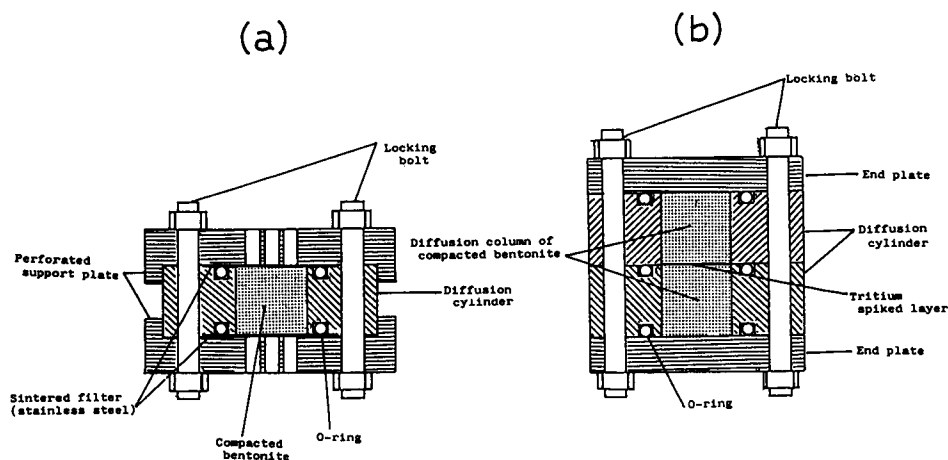


Fig. 1 Equipments for diffusion study :

- (a) immersion in water for pre-saturation of water in compacted bentonite,
 (b) diffusion of tritiated water in compacted bentonite.

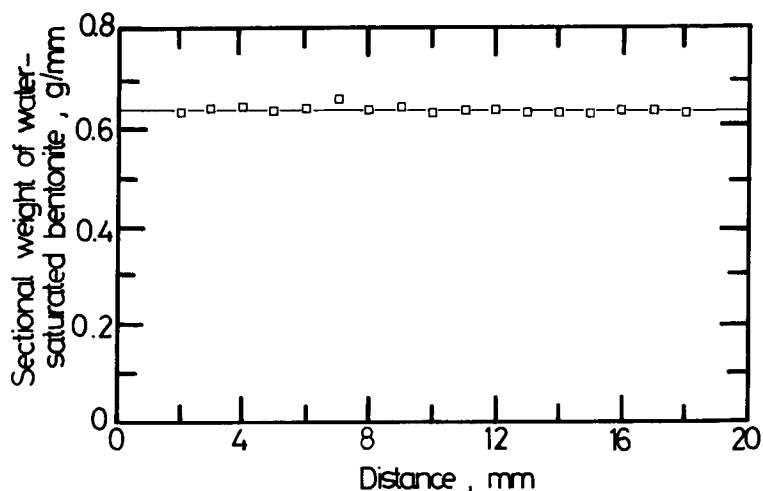


Fig. 2 The weight of sectioned bentonite was measured to confirm the homogeneity of water content after immersion in water.

In the tritium analysis, the sectioned piece of bentonite was in the liquid scintillator. As shown in Fig. 3, it was confirmed that even in such a condition, the counting rate of the scintillation counter was exactly proportional to the tritium concentration in the sectioned bentonite. The standard deviation of the twelve points was 2.4% .

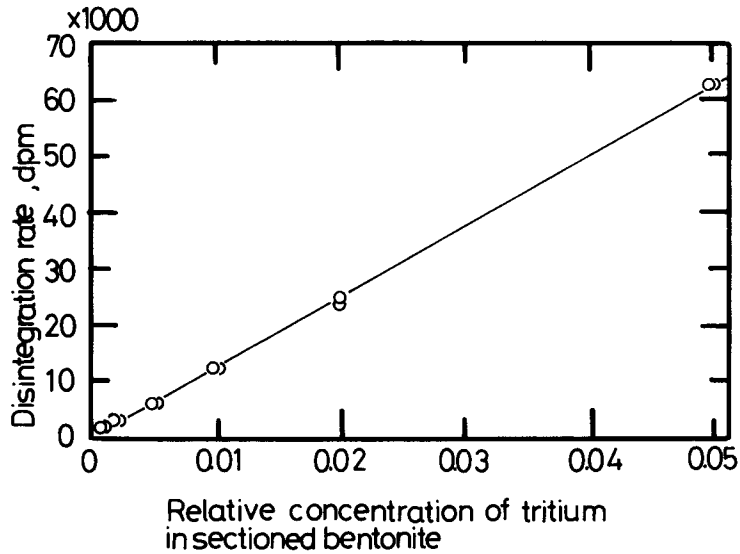


Fig. 3 It was confirmed that the disintegration rate measured by a liquid scintillation counter was sufficiently proportional to the amount of tritium spiked in the sectioned bentonite.

3. Analysis

In previous experiments, the following equation has been applied for the analysis^{5-7, 9)};

$$C/M = \exp[-x^2/4Dat]/\lambda/(\pi Dat) \quad (2)$$

where,

C : concentration of diffusing substance at distance x , [mole/cm³]

M : total amount of diffusing substance in instantaneous source per unit area normal to direction of diffusion, [mole/cm²]

Da : apparent diffusion coefficient [cm²/s]

t : time after introduction of instantaneous source, [s].

λ : 1 for the source located at one end of a column, and 2 for the source located at the middle of a column. [-]

The above equation describes a one-dimensional diffusion from an instantaneous planar source into a semi-infinite medium. Actually, however, the diffusion medium is finite. The solution for the diffusion in such a finite medium can be derived as follows by applying the mathematical procedure of the so-called

reflection at the impermeable boundaries,

$$C/M = [\exp[-x^2/4Dat] + \sum_{n=1}^{\infty} [\exp[-(2nl-x)^2/4Dat] + \exp[-(2nl+x)^2/4Dat]]] / \lambda \sqrt{\pi Dat} \quad (3)$$

In the present analysis, the resulting concentration profiles were fitted to curves drawn by using Eq. (3). The value of Da that yielded the best fit was chosen as the diffusivity of tritiated water in compacted bentonite under a given condition.

The difference of Eq. 3 from Eq. 2 is illustrated in Fig.4. It should be noticed that in the concentration profiles of Eq.3, the gradient becomes zero at the boundaries as well as at the center.

4. Results and Discussion

Two columns of bentonite of 2 cm length and 2 cm diameter were immersed in water for the pre-saturation of water (cf. Fig. 1). Then, tritiated water was spiked on the surface between two columns contacted with one another. The diffusion cell was closed tightly as shown in Fig.1, and stored in a bottle of 100% relative humidity for a predetermined time interval.

The concentration profiles obtained for the different intervals are shown in Fig.5. The diffusivity was chosen by applying Eq. 3 to each concentration profile of a different time interval. The resulting diffusivity can be written as

$$Da = (3.4 \pm 0.2) \times 10^{-6} \text{ cm}^2/\text{s} \quad (4)$$

The solid lines in Fig. 5 show the profiles calculated from Eq. 3 with the diffusivity of $Da = 3.4 \times 10^{-6} \text{ cm}^2/\text{s}$.

According to the experimental results by J.H. Wang¹¹⁾, the self-diffusivity of free water at 25°C is $2.13 \times 10^{-5} \text{ cm}^2/\text{s}$. This was obtained by using heavy water (D_2O) as the tracer. Comparing with this self-diffusivity, the diffusivity of water in bentonite obtained in the present work is smaller by about one order. The tortuosity will be around $\sqrt{2}$ ⁷⁾. The two tracers, D_2O in the experiments by J. W. Wang and HTO in the present work, have the same molecular weight with one another. So, the isotope effect due to the difference of the molecular weight may not be considered. Then, it can be concluded that the diffusivity of water in compacted bentonite is much smaller than the self-diffusivity of free water. It suggests that the simple relationship between the apparent diffusivity Da and the distribution coefficient Kd of the sorbable species in bentonite, Eq. (1),

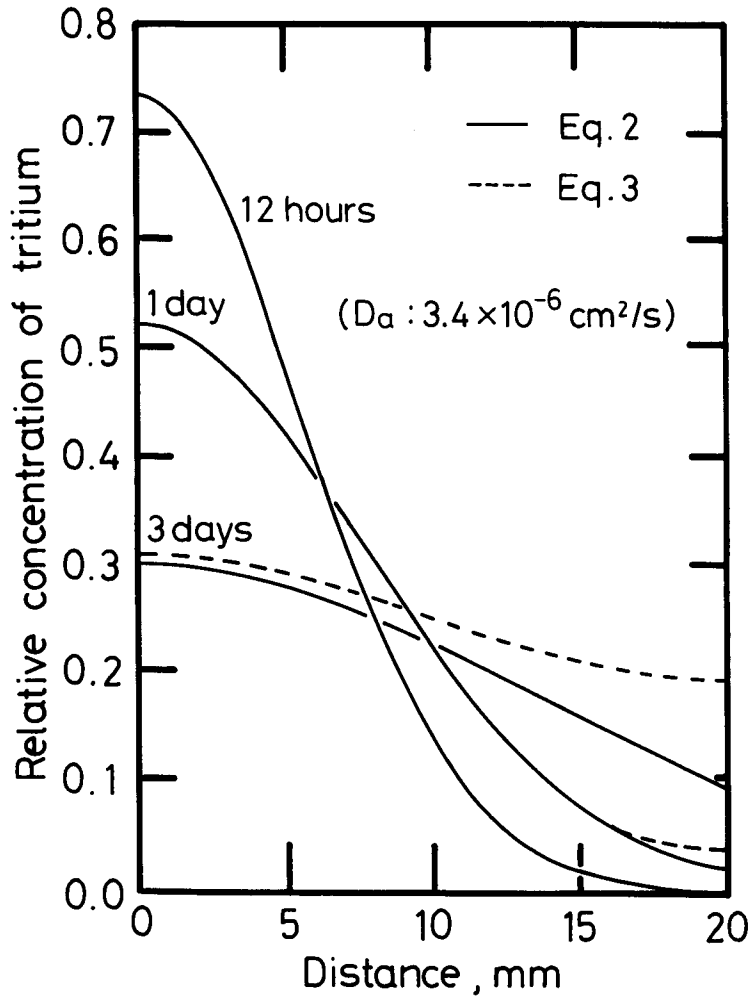


Fig. 4 Difference of calculated profiles depending on the applied solution (Eq. 2 or Eq. 3).

should be modified through the compilation of various results of further experiments.

5. Acknowledgments

The helpful discussions on the design of experimental equipments by Y.

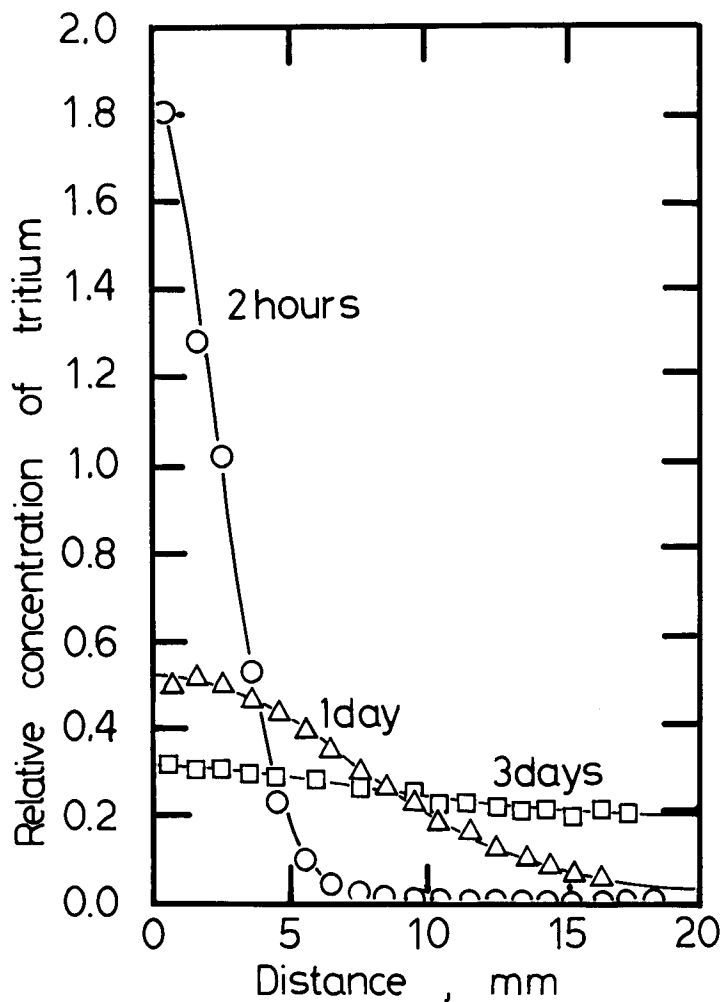


Fig.5 Concentration profiles of tritiated water for different time-intervals.

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6. References

- 1) I. Neretnieks: KBS technical report 79 (1978).
- 2) R. Push: Nucl. Technol., **45**, 153 (1979).
- 3) Annual Report of Swedish Nuclear Fuel Safety Project KBS 8026 (1981).

- 4) B.J. Wood, Nucl. Technol., **59**, 390 (1982).
- 5) B. Torstenfelt, K. Andersson, H. Kipatsi, B. Allard, and U. Olofsson: The Scientific Basis for Nuclear Waste Management, **6**, 295 (1982).
- 6) M. Kumagai, M. Sazarashi, K. Idemitsu, Kagawa, H. Umeki and K. Ishigure: Proc. Intern. Conf. Waste Management 89, vol.1, pp.45-421, Arizona Board Regents, (1989).
- 7) E.J. Nowak: Mat. Res. Soc. Symp. Proc. vol. 26, pp. 59-68, (1984).
- 8) I. Neretnieks: KBS technical report 30 (1977).
- 9) B. Torstenfelt et al.: Scientific Basis for Radioactive Waste Management, vol., pp.659-667, Elsevier (1982).
- 10) S.C.H. Cheung, D.W. Oscarson and R.S. Lopez: Mat. Res. Soc. Symp. Proc. **26**, 711 (1984).
- 11) J.H. Wang: J. Am. Chem. Soc. **73**, 510 (1951).