9–6 Evaluation of Radionuclide Migration by a Diffusion Model
—Diffusion Test and Model Development for Compacted Ca-Bentonite—

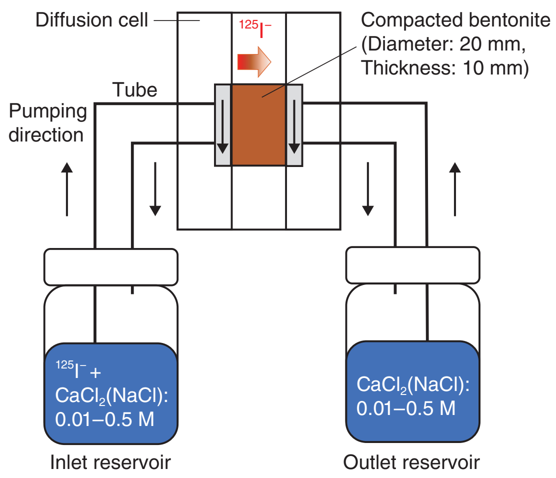

Fig.1 Schematic illustration of a diffusion test
The test setup comprises an electrolyte solution containing tracer in the inlet reservoir and an electrolyte solution without the tracer in the outlet reservoir in contact with compacted bentonite. The tracer diffuses through the compacted bentonite because of its concentration gradient. The change in the concentration of both solutions during the test and the concentration profile in the bentonite after the test are fitted with Fick's diffusion equation to calculate the effective diffusion coefficient (D_e).

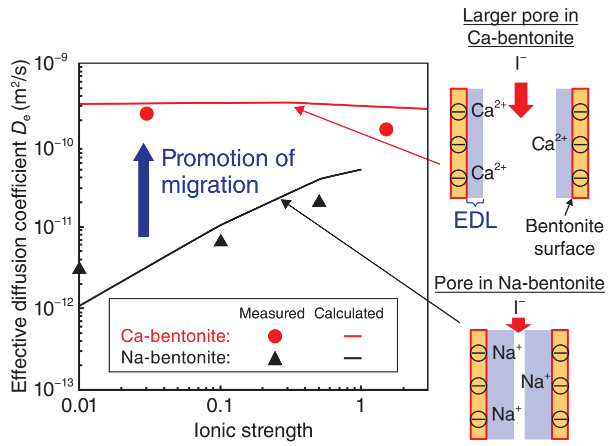

Fig.2 Measured D_e of $^{125}I^-$ in Na- and Ca-bentonite (dry density = 0.8 Mg/m³) and evaluation by a diffusion model
A diffusion model that takes into account the pore structure and electrostatic interaction on the bentonite surface can evaluate the D_e for radionuclide migration in Ca-bentonite.

In the geological disposal of high-level radioactive waste, compacted Na-bentonite is placed around the vitrified waste as a buffer material. When Na-bentonite comes in contact with groundwater, it swells and forms a microstructure with micropores, thereby effectively inhibiting radionuclide migration. However, the Ca^{2+} in groundwater leached from cement materials, which are used as repository components, can possibly be exchanged with the Na^+ in bentonite, transforming Na-bentonite to Ca-bentonite. This alteration may reduce the ability of the buffer to inhibit radionuclide migration, because Ca-bentonite has large pores as it has a poor swelling property.

In this study, tests were conducted to study the diffusion of radioiodine ($^{125}I^-$) in Na- and Ca-bentonite (Fig.1). The effective diffusion coefficient (D_e), which indicates the diffusion rate of radionuclides, was obtained. The impact of alteration from Na-bentonite to Ca-bentonite on radionuclide migration was investigated by comparing the D_e values in Na- and Ca-bentonite. We also evaluated the effect of ionic strength, which is promotional to the Ca^{2+} or Na^+ concentrations, in the pore water on D_e. Using these results, we developed an evaluation model for D_e values under a variety of groundwater conditions.

Fig.2 shows D_e values of $^{125}I^-$ for Na- and Ca-bentonite obtained by diffusion tests. As shown in the figure, the D_e of $^{125}I^-$ is higher in Ca-bentonite (● in Fig.2) than in Na-bentonite (▲ in Fig.2), indicating that the migration of $^{125}I^-$ is enhanced in Ca-bentonite. With regard to the dependence of D_e on ionic strength, the D_e of $^{125}I^-$ in Na-bentonite tended to increase with increasing ionic strength (▲ in Fig.2). In contrast, the D_e of $^{125}I^-$ in Ca-bentonite hardly affected by ionic strength (● in Fig.2).

These results can be explained by the difference in pore size between Na- and Ca-bentonite and the formation of the electric double layer (EDL) by the negative charges on the bentonite surface. Anions such as $^{125}I^-$ are electrically excluded from the EDL. As the thickness of EDL decreases with increasing ionic strength, the fraction of pore space available for anion migration increases, leading to an increase in D_e. However, in Ca-bentonite, the large pores result in a smaller EDL–total pore space ratio (Fig.2), and hence, the migration pathway for anions is larger than that in Na-bentonite. Thus, ionic strength has less influence on D_e in the case of Ca-bentonite than in the case of Na-bentonite.

We developed a diffusion model that considers the large pores and EDLs that are characteristic of Ca-bentonite and evaluated the D_e of $^{125}I^-$ (the solid line in Fig.2) using this model. As shown in the figure, the model mostly reproduced the experimental values of ionic strength. Thus, this model can evaluate the D_e values and is particularly applicable for evaluating the differences in D_e value caused by a transformation from Na-bentonite to Ca-bentonite. D_e is an important parameter for predicting radionuclide migration in the safety assessment of geological disposal. The knowledge obtained in this study will contribute to improving the reliability of setting D_e for compacted bentonite.

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Reference

Fukatsu, Y. et al., Diffusion of Tritiated Water, $^{137}Cs^+$, and $^{125}I^-$ in Compacted Ca-Montmorillonite: Experimental and Modeling Approaches, Applied Clay Science, vol.211, 2021, 106176, 10p.