8–11 Migration Behavior of Carbon-14 in Buffer Material

—Diffusion Experiments of Carboxylic Acid and Alcohol in Compacted Bentonite—

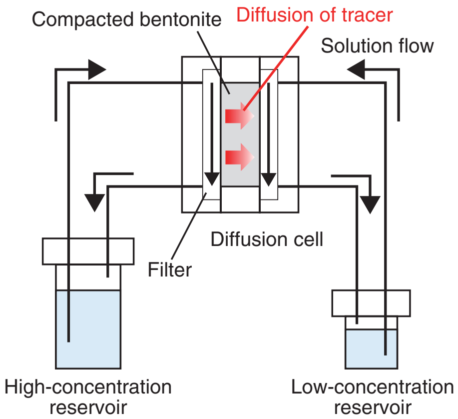

Fig.8-26 Schematic view of diffusion experiment
The solution from the reservoirs was recirculated through the filters placed on both sides of compacted bentonite. The tracers (acetic acid, butyric acid, and butanol) in the high-concentration reservoir diffuse through the compacted bentonite into the low-concentration reservoir. The diffusion behavior of the tracers can be observed from the change in tracer concentration in the low-concentration reservoir.

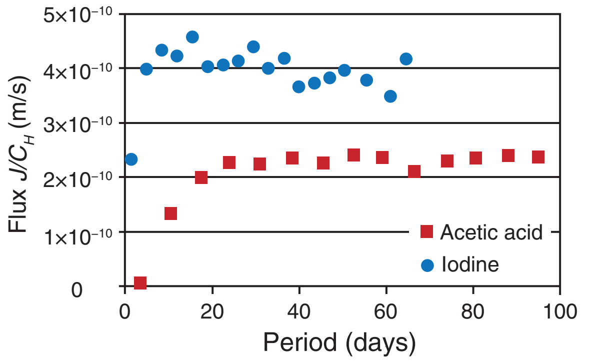

Fig.8-27 Results of diffusion experiments
Change in tracer flux with time for acetic acid and iodine. The flux is defined as the amount of tracer diffused from compacted bentonite per unit surface area and per unit time. The flux shown in the figure is normalized by the tracer concentration in the high-concentration reservoir.

Table 8-2 Effective diffusion coefficients (D_e) and formation factors (*FF*) measured by the diffusion experiments
The *FF* values of acetic acid (No.1, 2) and butyric acid (No.3, 4) were in good agreement with those of iodine, while the *FF* values of butanol were different from those of deuterated water (The *FF* values obtained from the same number sample were compared). These results indicate that the diffusion path of carboxylic acid was similar to that of iodine, while the diffusion property of butanol was different from that of deuterated water.

No.	Acetic acid		Butyric acid		Iodine	
	D_e (m²/s)	*FF* (×10⁻³)	D_e (m²/s)	*FF* (×10⁻³)	D_e (m²/s)	*FF* (×10⁻³)
1	2.3×10^{-12}	2.1	—	—	4.0×10^{-12}	1.9
2	2.6×10^{-12}	2.4	—	—	4.4×10^{-12}	2.1
3	—	—	8.0×10^{-13}	0.93	2.0×10^{-12}	1.0
4	—	—	6.2×10^{-13}	0.71	1.5×10^{-12}	0.74

No.	Butanol		Deuterated water	
	D_e (m²/s)	*FF* (×10⁻³)	D_e (m²/s)	*FF* (×10⁻³)
5	2.5×10^{-12}	4.5	1.7×10^{-11}	7.7
6	3.2×10^{-12}	5.7	2.0×10^{-11}	8.8

Carbon-14 (^{14}C) is one of the radionuclides present in radioactive waste in the form of activated metal waste. In the geological disposal of radioactive waste, the radionuclides released from the waste are retarded in the compacted bentonite used as the buffer material. However, the retention of ^{14}C in compacted bentonite is hardly expected because of the weak sorption of ^{14}C on bentonite. Therefore, ^{14}C is one of the key radionuclides in the safety assessment of geological disposal. A part of the ^{14}C in the activated metals is released as low-molecular-weight organic compounds. In this study, the diffusion behavior of carboxylic acids (acetic acid and butyric acid) and alcohol (butanol) in compacted bentonite was investigated via diffusion experiments. The diffusion behavior was compared with that of iodine and deuterated water, which has been widely reported.

The changes in fluxes with time measured by the diffusion experiment (Fig.8-26) are shown in Fig.8-27. The fluxes increased with time and reached a steady state. The effective diffusion coefficient (D_e) was calculated from the flux at the steady state based on Fick's first law. The formation factor (*FF*), which indicates the diffusion path properties such as tortuosity, was calculated by dividing D_e by the self-diffusion coefficient of the tracer in water. From Table 8-2, the *FF* values of acetic acid and butyric acid are in good agreement with those of iodine, which diffuses as an anion similar to acetic acid and butyric acid. In contrast, the *FF* values of butanol were different from those of deuterated water, though both tracers diffuse as electrically neutral species. (Note that the *FF* values obtained from the same number sample were compared (Table 8-2) as the *FF* values vary depending on the condition of the compacted bentonite sample even if the samples are prepared under identical conditions.). These results indicate that the diffusion path of carboxylic acid was similar to that of iodine, while the diffusion property of butanol differed from that of deuterated water. Thus, the difference in D_e values between carboxylic acid and iodine originates from the difference between their self-diffusion coefficients. The D_e values of carboxylic acids can be calculated from those of iodides by using their self-diffusion coefficients.

In the safety assessment for geological disposal, the D_e values of radionuclides in compacted bentonite should be evaluated with high reliability. Nevertheless, few studies reported the D_e values of carboxylic acids. The results of this study indicate that the widely reported D_e values of iodine can be utilized for setting the D_e for ^{14}C in the form of carboxylic acids. This knowledge will contribute to increasing the confidence in setting the D_e for ^{14}C.

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Reference

Ishidera, T., Diffusion of Acetic Acid, Butyric Acid, and Butanol in Compacted Montmorillonite, Journal of Radioanalytical and Nuclear Chemistry, vol.330, issue 1, 2021, p.149–158.