Transport of Iodide Ion in Compacted Bentonite Containing Ag_2O - 12111

Sung Paal Yim*, Ji-Hyun Lee*, Heui-Joo Choi*, Jong-Won Choi*,
Cheo Kyung Lee**
Korea Atomic Energy Research Institute,
Yusong, Daejon, Republic of Korea
** Handong Global University,
Heunghae-eub, Buk-gu, Pohang, Kyungbuk, Republic of Korea

ABSTRACT

Observations of the transport of iodide through compacted bentonite containing Ag_2O as additive and that without additive were made. Compacted bentonite samples with densities of 1.41 g/cm^3 and 1.60 g/cm^3 were used in the experiment. The amount of Ag_2O added to the compacted bentonite was in the range of 0.0064 ~ 0.0468 wt/wt%. Two diffusion solutions were used: one in which iodide ion was dissolved in demineralized water (pure iodide solution), and one in which iodide ion was dissolved in 0.1 M NaCl solution (0.1 M NaCl-iodide solution). Experimental results confirmed that iodide ion was transported by the diffusion process in the compacted bentonite containing Ag_2O as well as in the compacted bentonite without Ag_2O. The time-lag of diffusion of iodide ion in the compacted bentonite containing Ag_2O is larger than that in the compacted bentonite without Ag_2O. The increase of the time-lag of diffusion was observed in pure iodide ion solution as well as in 0.1M NaCl-iodide solution. The apparent diffusion coefficient of iodide ion in the compacted bentonite containing Ag_2O was smaller than in the compacted bentonite without Ag_2O. The effective diffusion coefficient decreased as the amount of Ag_2O in the compacted bentonite increased.

INTRODUCTION

Iodine-129 is one of the major nuclides concern with the disposal of high level radioactive waste because it has a very long half life and is radiologically hazardous. Iodine usually exists as iodide ion under the usual condition of a disposal site and it is not strongly adsorbed onto bentonite, a buffer material recommended to retard the release of radionuclides in disposal sites. An interest in diffusion of iodide ion in bentonite comes from the question whether bentonite has an enough retardation capacity of diffusion of iodide because it is not capable of adsorption on the anion.

According to a recent investigation [1], iodide ion is adsorbed well onto a bentonite containing Ag_2O, and this could be used as a buffer material in the disposal site to retard the transport of iodide ion from the container to the environment.

If iodide is adsorbed well onto the bentonite containing Ag_2O, it is assumed that the transport of iodide ion in bentonite would be more delayed than that in the bentonite without Ag_2O. Experimentation is needed to quantify the effect of Ag_2O on the rate of iodide ion release and the diffusion property through the bentonite buffer material.

In this study, by using through-diffusion method, the time-lag of diffusion, apparent diffusion coefficient and effective diffusion coefficient were measured to evaluate the transport property of iodide ion in compacted bentonite containing Ag_2O.

EXPERIMENTALS

Through-diffusion method

A through-diffusion method was used to observe the transport property of iodide ion in a compacted bentonite containing Ag_2O and a compacted bentonite without Ag_2O. The through-diffusion method for obtaining a time-lag of diffusion, an apparent diffusion coefficient and an effective diffusion coefficient of ions in clay layer such as bentonite is well established by Oscarson et al. [2] and Lee et al. [3].

At steady state, time-lag of diffusion, t_e and apparent diffusion, D_a can be obtained from the following equation derived from Fick's second law.

$$\frac{Q}{A} = \frac{D_a \alpha C_o}{L} t - \frac{L \alpha C_o}{6} \qquad (Eq.1)$$

Where Q is the cumulative amount of ion transported through the bentonite, A the cross sectional area, D_a is the apparent diffusion coefficient, α is the capacity factor, L is the thickness of betonite, C_o is the initial concentration, and t is the time.

Using graph of Q/A vs t for (Eq. 1), the intercept of the slope at the axis (Q/A = 0) is the time-lag of diffusion, t_e. Subsequently, D_a is derived from the following equation.

$$D_a = \frac{L^2}{6 t_e} \qquad (Eq.2)$$

Also, as the effective diffusion coefficient, D_e has a following relationship with the apparent diffusion.

$$D_e = \alpha D_a \qquad (Eq.3)$$

The effective diffusion coefficient can be calculated from the following equation.

$$D_e = \frac{S L}{C_0} \qquad (Eq.4)$$

where S is the slope of the graph for Q/A vs t.

Material

The powdered bentonite used in this study was sampled from the Kyeongju, the southeastern area of Korea and screened through a 200 mesh sieve. The bentonite was the same as that used in the study by Lee et al.[4] and the chemical composition of the bentonite is summarized in Table I. The XRD pattern is also shown Figure 1.

Table I. Chemical composition of bentonite [4]

Chemical Constituents	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	FeO	SO_3	MnO
Weight %	56.80	19.96	6.03	2.59	0.77	0.93	1.25	0.15	1.28	0.04

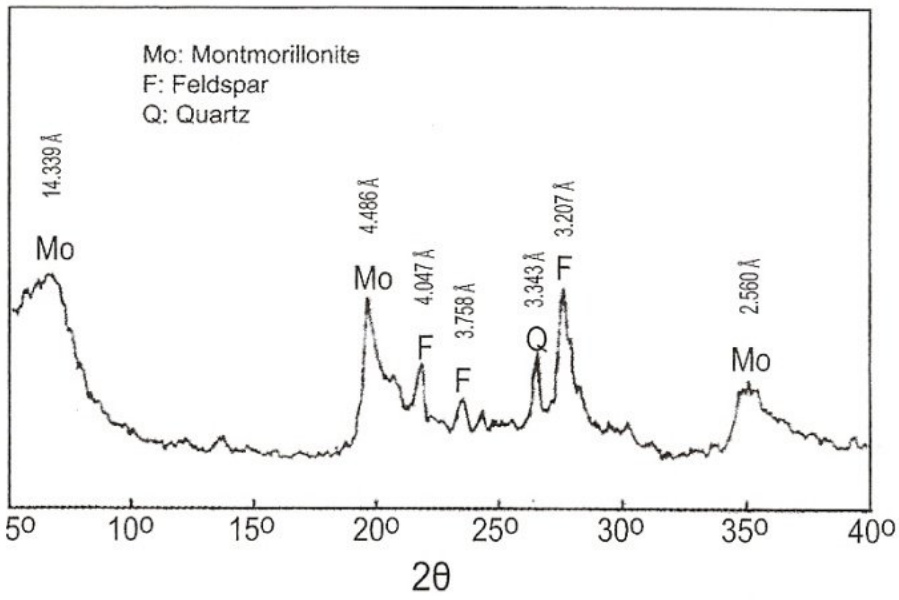

Fig 1. XRD Pattern of bentonite [4]

Experimental Apparatus

The apparatus used in the experiment is shown in Figure 2. A diffusion cell was made of polyacrylates. A sample ring made of stainless steel with 50 mm diameter and 7 mm thickness is located in the inner shell to fix compacted bentonite. A metal filter made of stainless steel with 50 mm diameter, 3.2 mm thickness and 25 μm average pore is installed on both sides of the sample ring. Four tubes are connected to the diffusion cell. Two tubes are used for supplying the diffusion solution to the diffusion cell and the others are for collecting the solution containing the iodide ion diffused from bentonite.

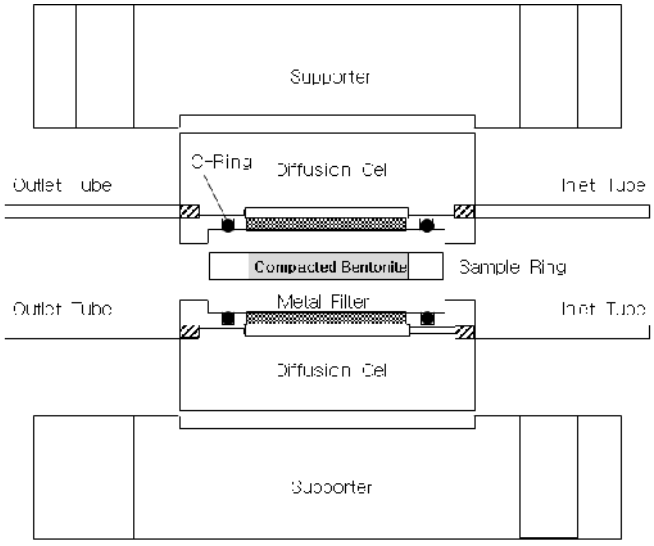

Fig 2. Experimental apparatus used in through-diffusion method

Compacted Bentonite

Bentonite powder was compacted into the sample ring by using a compression mold. Compacted bentonites with densities of 1.41 g/cm^3 and 1.60 g/cm^3 were prepared for the experiment. Compacted bentonite containing Ag_2O was made by the same way after addition of Ag_2O to the bentonite powder. The amount of Ag_2O added to compacted bentonite was in the range of 0.0064 ~ 0.0468 wt/wt%.

Diffusion Solution and Collection Solution

Two diffusion solutions were prepared: A pure iodide solution in which iodide ion was dissolved in demineralized water, and a 0.1 M NaCl-iodide solution in which iodide ion was dissolved in 0.1 M NaCl solution. The concentration of iodide ion in each solution was 100 mg/l. Demineralized water was used as collection solution when the diffusion solution was a pure iodide solution. A 0.1 M NaCl solution was used as a collection solution when the diffusion solution was a 0.1 M NaCl-iodide solution.

Experimental Method

The diffusion cell was placed in a supporter and the compacted bentonite was allowed to be saturated with demineralized water or 0.1 M NaCl solution for 6 weeks. Diffusion solution was then passed over one side of the bentonite at 1 ml/hr and collection solution was passed over the opposite side of the bentonite at the same rate. Each of the two solutions was injected into the diffusion cell using a syringe pump. The collection solution passed through the diffusion cell was received in a volumetric flask and analyzed by the ion meter with an ion-selective electrode.

RESULTS

Pure Iodide Solution As Diffusion Solution

Results of the experiments on compacted bentonites with and without Ag_2O using a pure iodide solution are shown in Figure 3. The density of all the bentonites was 1.41 g/cm^3. After an intial time lag, linear data on plot of Q/A shows that the iodide ion is transported in compacted bentonite by diffusion process. The time-lag of diffusion, t_e, apparent diffusion coefficient, D_a, effective diffusion coefficient, D_e are summarized in Table II.

The time-lag of diffusion of iodide ion, t_e, delayed as the amount of Ag_2O in bentonite increased. While time-lag of diffusion of iodide ion in the compacted bentonite without Ag_2O was 0.4 day or 2.1days, those in compacted bentonite containing 0.0064, 0.0128 and 0.0468 wt/wt% Ag_2O increased to 12.0 days, 60.4 days, and 220.3 days, respectively.

The apparent diffusion coefficients of iodide ion in the compacted bentonites containing Ag_2O were smaller than that in the compacted bentonite without Ag_2O.

The effective diffusion coefficient in compacted bentonite containing 0.0064 wt/wt% Ag_2O was slightly larger than that in compacted bentonite without Ag_2O, but those in compacted bentonites containing 0.0128 wt/wt% Ag_2O and 0.0468 wt/wt% Ag_2O were smaller than that in compacted bentonite without Ag_2O. The effective diffusion coefficient decreased as the amount of Ag_2O added increased.

The effective diffusion coefficients in compacted bentonite without Ag_2O were about one-tenth of measured by Oscarson et al. [2] (D_e : 2.1×10^{-11} under the condition of the ionic strength 0.22 M and the dry density of 1.39 g/cm^3). However, these results were similar to those reported by Lee et al. [3] (D_e : 4.99×10^{-12} under the condition of the ionic strength 0.0015 M and the dry density of 1.4 g/cm^3). Because the ionic strength of the diffusion solution was 0.008 M in this

experiment, the differences with the literature may be related to the ionic strength of the diffusion solution used in the experiment.

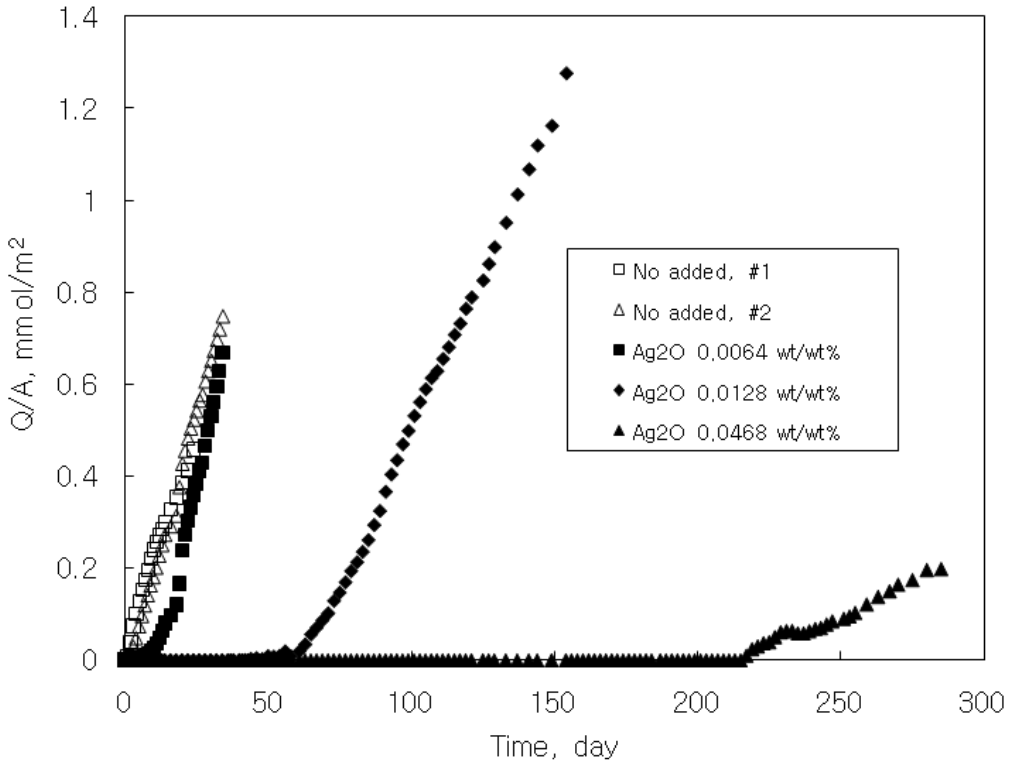

Fig 3. Breakthrough curves of iodide ion in compacted bentonites
- Dry density of the compacted bentonite : 1.41 g/cm^3
- Diffusion solution : pure iodide solution

Table II. Time-lags and diffusion coefficients of iodide ion in compacted bentonites

Compacted bentonite (Dry density : 1.41 g/cm^3)	Pure iodide ion solution (100 mg of iodide ion in 1 L of demineralized water)		
	Time-lag(t_e), day	Apparent diffusion coefficient(D_a), m^2/s	Effective diffusion coefficient(D_e), m^2/s
No added, #1	2.1	4.4×10^{-11}	2.4×10^{-12}
No added, #2	0.4	2.2×10^{-10}	2.7×10^{-12}
0.0064 wt/wt% Ag_2O	12.0	7.9×10^{-11}	3.1×10^{-12}
0.0128 wt/wt% Ag_2O	60.4	1.6×10^{-12}	1.4×10^{-12}
0.0468 wt/wt% Ag_2O	220.3	4.3×10^{-13}	3.3×10^{-13}

- #1, #2: Duplicate

0.1 M NaCl-iodide Solution As Diffusion Solution

An experimental was performed using 0.1 M NaCl-iodide solution as the diffusion solution to confirm the effect of addition of Ag_2O in the compacted bentonite on the time-lag of diffusion, the apparent diffusion coefficient and the effective diffusion coefficient. The ionic strength of 0.1 M NaCl-iodide solution is 0.2 M and the concentration of chloride ion (3,500 mg/l) in the solution is 35 times larger than that of the iodide ion (100 mg/l).

Results of the experiments are shown in Figure 4. Similar to the results of the experiment using a pure iodide solution, the breakthrough curves in the figure show that the iodide ion is transported in compacted bentonite by a diffusion process. The time-lag of diffusion, t_e, the apparent diffusion coefficient, D_a, the effective diffusion coefficient, D_e, are summarized in Table III.

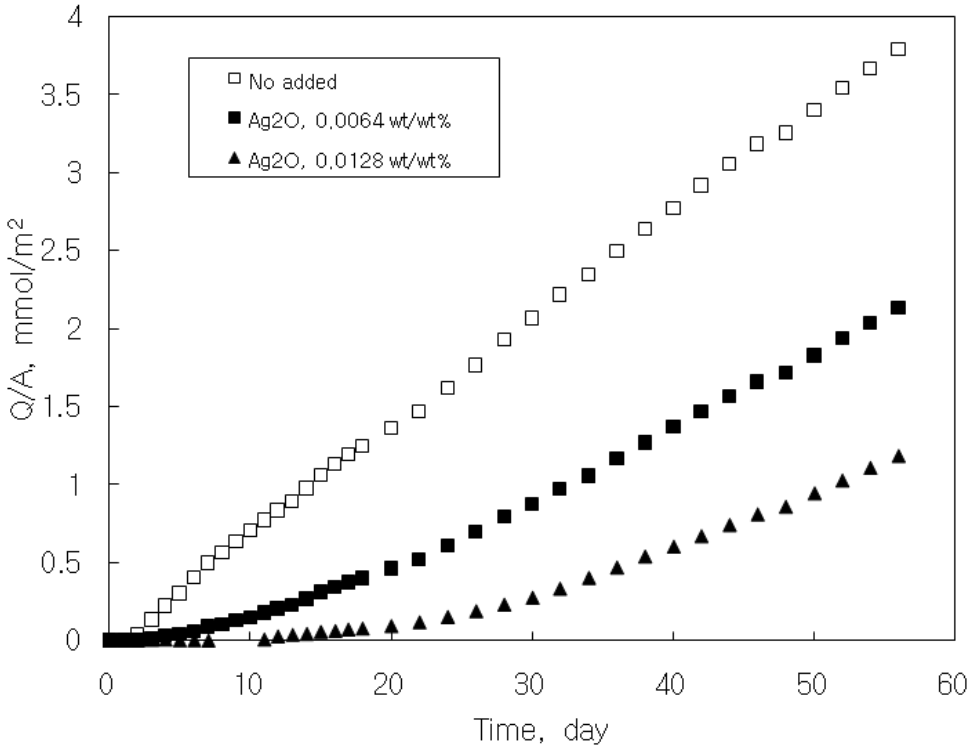

Fig 4. Breakthrough curves of iodide ion in compacted bentonites
- Dry density of the compacted bentonite: 1.60 g/cm^3
- Diffusion solution: 0.1 M NaCl-iodide ion solution

In 0.1 M NaCl-iodide solution as diffusion solution, when the dry density of compacted bentonites was 1.41 g/cm^3, the time-lag of diffusion of iodide ion in compacted bentonite containing 0.01wt/wt% Ag_2O increased about 8days as compared to that of compacted bentonite without Ag_2O. And when the dry density of compected bentonites was 1.60 g/cm^3, while time-lag of diffusion in the compacted bentonite without Ag_2O was only 0.3 day, those in compacted bentonite containing 0.0064 and 0.0128 wt/wt% Ag_2O increased to 11.5 days and 23.6 days, respectively. However, in the compacted bentonite containing Ag_2O, time-lags of diffusion measured using 1 M NaCl-iodide diffusion solution were lower than those measured using pure iodide solution.

Table III. Time-lags and diffusion coefficients of iodide ion in compacted bentonites

Compacted bentonite	Dry density, g/cm^3	0.1 M NaCl-iodide ion solution (100 mg of iodide ion in 0.1 M NaCl Solution)		
		Time-lag(t_e), day	Apparent diffusion coefficient (D_e), m^2/s	Effective Diffusion coefficient (D_a), m^2/s
No added	1.41	1.6	5.8×10^{-11}	1.7×10^{-11}
0.01 wt/wt% Ag_2O	1.41	9.1	1.0×10^{-11}	3.3×10^{-11}
No added	1.60	1.3	7.0×10^{-11}	8.7×10^{-12}
0.0064 wt/wt% Ag_2O	1.60	11.5	8.3×10^{-12}	4.9×10^{-12}
0.0128 wt/wt% Ag_2O	1.60	23.6	4.3×10^{-12}	3.7×10^{-12}

The apparent diffusion coefficients of iodide ion in the compacted bentonites containing Ag_2O have smaller values than that in the compacted bentonite without Ag_2O. In the compacted bentonite containing Ag_2O, the apparent diffusion coefficients of iodide ion measured using 1 M NaCl-iodide diffusion solution were higher than those measured using pure iodide ion solution.

The effective diffusion coefficient in compacted bentonite without Ag_2O obtained in this experiment is similar to the results measured under the ionic strength of 0.2 M by Oscarson et al. [1] (D_e: 2.1×10^{-11} m^2/s in dry density of 1.39 g/cm^3, D_e: $0.1 \times 10^{-11} \sim 0.68 \times 10^{-11}$ m^2/s in dry density of 1.61~1.63 g/cm^3).

At a dry density of compacted bentonite of 1.41 g/cm^3, the effective diffusion coefficient of compacted bentonite containing 0.01wt/wt% Ag_2O was slightly larger than that of compacted bentonite without Ag_2O. However, at a dry density of compacted bentonite of 1.60 g/cm^3, the effective diffusion coefficients of compacted bentonite containing 0.0064 wt/wt% and 0.0128 wt/wt Ag_2O were smaller than that of compacted bentonite without Ag_2O. The effective diffusion coefficient decreased as the amount of Ag_2O added increased. In addition, the effective diffusion coefficients measured using 1 M NaCl-iodide diffusion solution were higher than those compared to those measured using pure iodide solution.

SUMMARY

The transport of iodide ion in compacted bentonite containing Ag_2O was investigated as an attempt to improve the safety of the environment when the high level radioactive waste is disposed.

Compacted bentonite samples with densities of 1.41 g/cm^3 and 1.60 g/cm^3 were used in the experiment. The amount of Ag_2O added in compacted bentonite was in the range of 0.0064 ~ 0.0468 wt/wt%. Two diffusion solutions were used: a solution in which iodide ion was dissolved in demineralized water (pure iodide solution), and one in which iodide ion was dissolved in 0.1 M NaCl solution (0.1 M NaCl-iodide solution).

Results confirmed that iodide ion is transported by a diffusion process in the compacted bentonite containing Ag_2O as well as in the compacted bentonite without Ag_2O. The time-lag of diffusion of iodide ion in the compacted bentonite containing Ag_2O is larger than that in the

compacted bentonite without Ag_2O. The increase of the lag-time of diffusion was observed in pure iodide solution as well as in 0.1M NaCl-iodide solution.

In both diffusion solutions, the apparent diffusion coefficient of iodide ion in the compacted bentonite containing Ag_2O was smaller than that in the compacted bentonite without Ag_2O. Also, the effective diffusion coefficient decreased as the amount of Ag_2O in compacted bentonite increased. The effective diffusion coefficients of iodide ion in the compacted bentonites without Ag_2O obtained in this study were similar to those reported in the literature.

To clearly understand the diffusion of iodide ion in compacted bentonite containing Ag_2O, the effect of amount of Ag_2O in the compacted bentonite on the time-lag of diffusion and the effective coefficient should be more quantitatively analyzed through further experiment.

Based on these efforts to evaluate the transport of iodide ion in the compacted bentonite containing Ag_2O, it is expected that the compacted bentonite containing Ag_2O could be utilized to increase the safety of radioactive waste disposal.

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