Flow Rate Dependency of Permeability Changes with the Deposition of CSH under the Highly Alkaline Ca-rich Condition – 17175

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ABSTRACT

The formation of calcium silicate hydrate (CSH) around the geological repository is one of the notable factors for the assessment of the radionuclide migration. This study discussed the clogging effect of CSH in flow path with the change in the flow rate based on the flow experiments using micro flow cell. In the experiment, the change of the permeability with the alteration of granite under the highly alkaline and Ca-rich condition was examined. As a result, the permeability in flow path gradually decreased with the periodical increase and decrease of the permeability. In addition, under the condition of the lower flow rate (0.06 ml/h), the permeability became much smaller than those for other conditions of the higher flow rates. These suggest that the clogging of flow path will progress with the local deposition/moving of CSH, in particular, under the condition of the lower flow rate. Retardation effect of radionuclide migration will be expected because it is prospected that flow rate of groundwater is lower than that of this study in the actual condition of repository.

INTRODUCTION

High-level radioactive wastes (HLW) will be disposed in the deep geological repository, around which the host rock is expected to retard the migration of radionuclides as a natural barrier [1]. This geological disposal system is based on the concept that HLW containing the radionuclides of long half-life must be isolated from the biosphere for a long period by a multiple barrier system [2], until the radioactivity of HLW decays enough not to affect human beings on the ground. In Japan, for assessing the performance of the barrier system, it is important to comprehend the migration of radionuclides through the fractures in the host rock with groundwater because the underground environment is a groundwater-rich condition as in other countries. In addition, groundwater is altered to highly alkaline condition by alkaline species contained in cementitious materials used for the construction of the repository [3]. Under such a condition, the formation of calcium silicate hydrate (CSH) alters the chemical and physical properties of the fractures in the host rock. Here, it is predicted that CSH as a secondary mineral is formed by the reaction of Ca ions leached from cementitious materials with silicate species dissolved from rocks (granite) under the highly alkaline condition (≈ pH 13) [3, 4]. The deposition of CSH in the fractures of the rock might decrease the migration of groundwater including radionuclides.

According to these backgrounds, this study examined this retardation effect with the deposition of CSH in the fracture (flow paths) of the rock at relatively low flow rate. So far, in a preliminary study [5], the authors have reported that micro flow paths on the surface of granite were clogged by the deposition of CSH under the condition of high pH (=12.5) and Ca-rich. However, the experimental results have shown that the deposited CSH might be flowed out of the micro flow path under the conditions of relatively high flow rate within the range of from 0.2 to 2.0 ml/h. Therefore, this study examined the clogging effect, i.e., the deposition behavior of CSH in flow paths, at lower flow rates (0.06 ml/h and 0.2 ml/h), so that the relation between the deposition of CSH and the clogging effect is appropriately estimated under the condition with little CSH flowing out of the micro flow cell.

EXPERIMENTAL METHOD

Flow Experiments Using Micro Flow Cell

Figures 1 and 2 show an illustration of the experimental apparatus including the micro flow cell. So far, Okuyama et al. [6, 7] have also applied the micro flow cell to the estimation of radionuclide migration with the sorption and diffusion. In general, micro flow cell is well-known as an apparatus to simulate flow-paths with aperture less than 100 µm. As shown in Fig.2, in order to simulate flow-paths on the surface of granite [8], this study prepared the micro flow cell by nipping the Teflon sheet of 80 µm in thickness between the granite chip and the basement of Teflon. The Teflon sheet had a slit in which fluid can flow.

As in our previous studies [5, 8], its area size of the slit was 5 mm in width and 60 mm in length. The granite chip (from Makabe area, Ibaraki Prefecture, Japan) was roughly ground, so the surface of granite chip was grinded with #2000 (SiC grinding powder was 7.9 µm of a particle diameter). The permeability was monitored by a pressure gauge attached at the inlet of the micro flow cell (The estimation of permeability is described later). The flow rate was set to 0.06 ml/h and 0.20 ml/h by a constant flow pomp. For preventing the change in the hydraulic head pressure, all experimental devices were on the same level. The experimental period was about 10 days. The temperature of the micro flow cell was 298 K (room temperature). As an injection solution, 8.5 mM $Ca(OH)_2$ solution was adjusted to 12.5 in pH with a NaOH solution. This concentration of Ca was set by considering that of the pore water in cementitious materials [3]. In order to avoid the formation of calcium carbonate, this injection solution was prepared in a globe bag filled with nitrogen gas. Moreover, pure water used for this preparation was removed carbon dioxide by bubbling with nitrogen gas.

During the experiments, the concentrations of Ca and Si in the solution eluted at the outlet of the micro flow cell were measured by inductively coupled plasma atomic emission spectrometry (ICP-AES, SPS7800), and the pH value was also monitored. Under these experimental conditions, the elution of Si was not detected, in other words, CSH formed in the micro flow cell did not elute. Furthermore, the concentration of Ca ions and pH at the outlet were almost the same as the injection conditions.

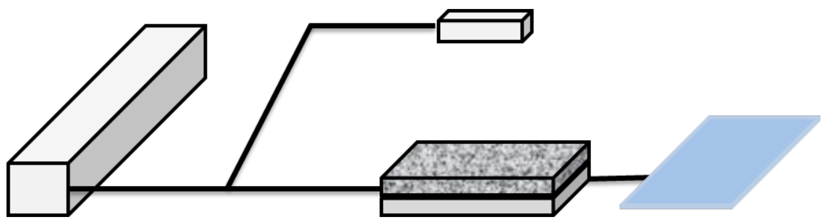

Fig. 1. Schematic view of the experimental apparatus.

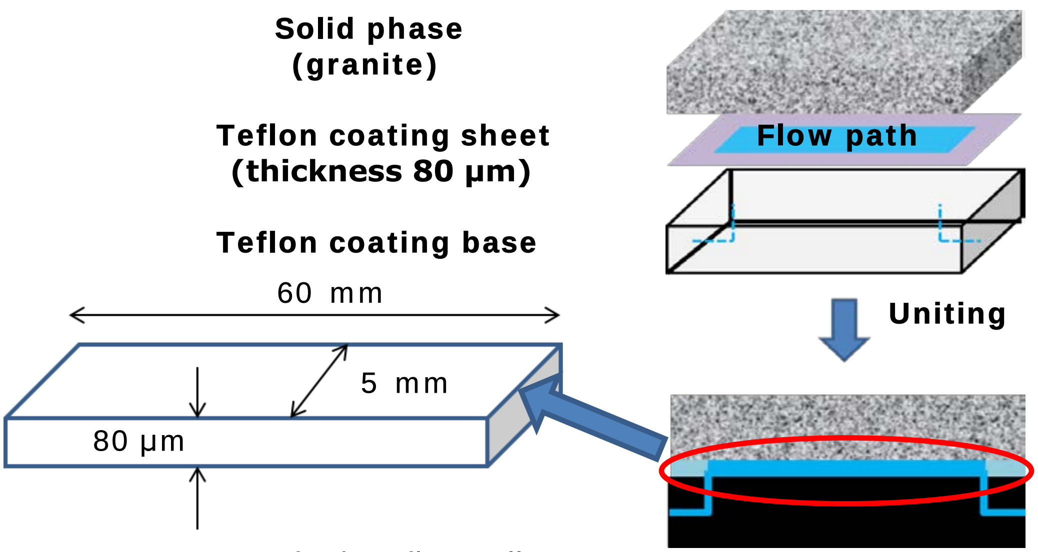

Fig. 2. Inner structure of micro flow cell.

Estimation of Permeability

In this study, the permeability in micro flow cell was calculated by the same way as the previous study [5]. Considering Darcy's law, the overall permeability, k [m^2] of the micro flow cell is defined by

$$v = \frac{k}{\mu}\frac{p_{in}-p_{out}}{x_1}, \qquad \Delta p \equiv p_{in} - p_{out} \quad , \tag{1}$$

where v is Darcy's fluid flow velocity [m/s], μ is fluid viscosity [Pa·s], p is pressure [Pa], x_1 is the length of flow cell along flow direction [m]. P_{in} and P_{out} mean the pressure at the inlet ant outlet of micro flow cell.

Here, the volume flow rate, Q (m^3/s) yields

$$Q = Av = vwb, \tag{2}$$

where w is the width of the slit in the micro flow cell [m], b is the aperture of flow-path [m]. *A* [m^2] is the *wb*, which means a cross section of the flow path. Furthermore, as reported by the previous study [10], the permeability, *k*, of a parallel flat board layer as shown in Fig. 2 is approximated by

$$k = \frac{b^2}{12} \quad . \tag{3}$$

From equations (1) and (2), equation (4) is given, i.e.,

$$Q = \frac{k}{\mu} bw \frac{\Delta p}{x_1} \quad . \tag{4}$$

In addition, equation (4) is rewritten with equation (3) as

$$Q = \frac{b^3 w}{12\mu} \frac{\Delta p}{x_1} \quad . \tag{5}$$

Finally, *b* can be estimated by

$$b = \left(\frac{12\mu Q x_1}{w \Delta p}\right)^{\frac{1}{3}} \quad . \tag{6}$$

Furthermore, using this estimated *b* and equation (3), the overall permeability, *k*, can be obtained.

RESULTS AND DISCUSSION

Figures 3 and 4 show the permeability change for each flow rate (0.06 ml/h and 0.20 ml/h). The vertical axis in Figs. 3 and 4 is the normalized permeability. The estimated permeability at each measured point is normalized by the maximum value of the permeability during the experiments. As mentioned above, for the conditions of these flow rates in Figs. 3 and 4, the elution of Si was not detected at the outlet of micro flow cell, while the concentration of Ca and the pH were kept constant as the injection condition, the formation of CSH is stable in a range of pH larger than 10, i.e. the condition for the formation of CSH was kept enough. This means that the CSH formed in the micro flow cell did not elute because the elution of Si at the outlet reflects that of CSH. In Figs. 3 and 4, from the view point of the long term of these experimental results, the permeability was gradually decreasing. On the other hand, from the view point of short term of that, the permeability was periodically changing decrease and increase. If CSH uniformly deposits on the surface of granite in micro flow cell, the decrease in the permeability would be constant with time. These permeability changes as shown in Figs. 3 and 4 suggest that the micro flow path in the cell will be locally clogged by the heterogeneous deposition of CSH on the surface of granite and the re-distribution of CSH particles drifted with the flow.

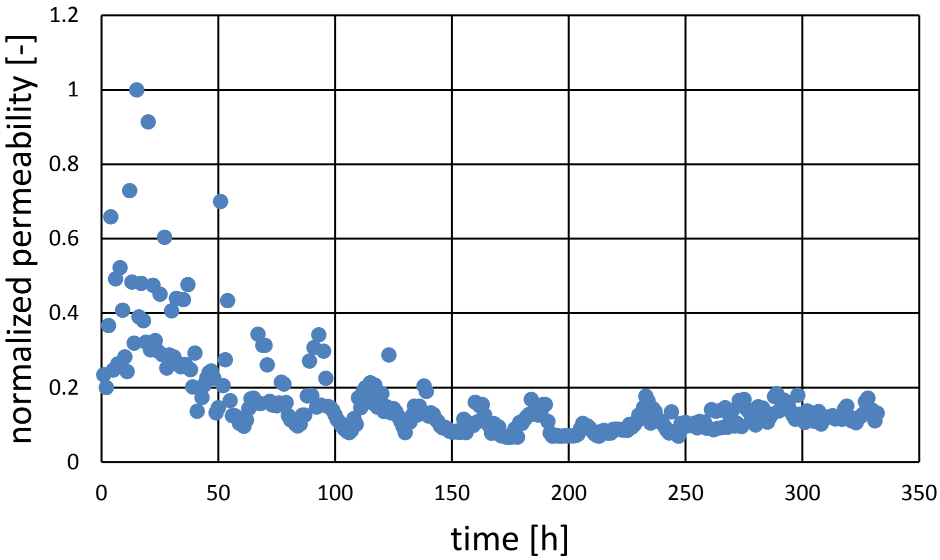

Fig. 3. Change of permeability. (Flow rate = 0.06 ml/h)

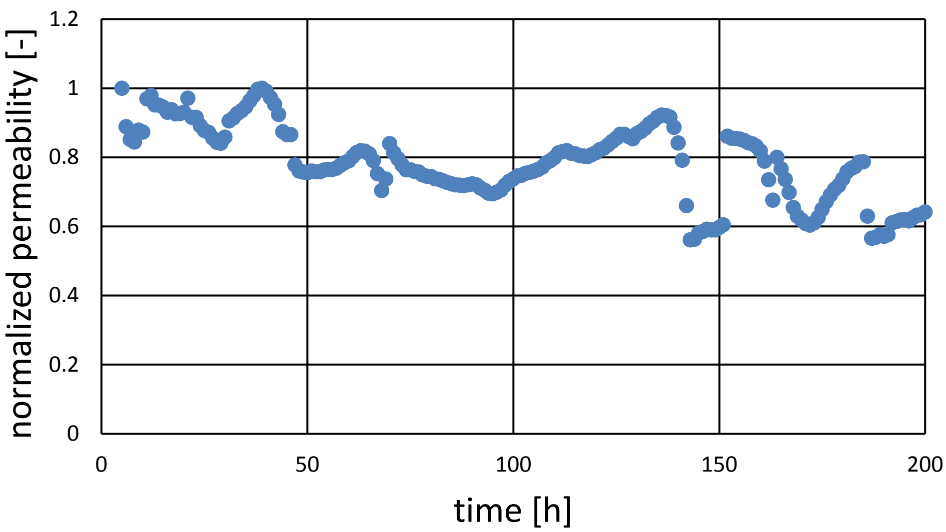

Fig. 4. Change of permeability. (Flow rate = 0.2 ml/h)

Figures 5 and 6 show the permeability change for the condition of the higher flow rates (1 ml/h and 2 ml/h) reported in our previous study [5]. Under the conditions of the higher flow rates in Figs. 5 and 6, the decrease fraction of the permeability was about 20% or less. Besides, the permeability for 2 ml/h apparently recovered to the maximum permeability after once the permeability decreased with time. Such changes in the permeability suggest that CSH cannot clog stably because most of CSH formed in micro flow cell are eluted under the condition of the high flow rate.

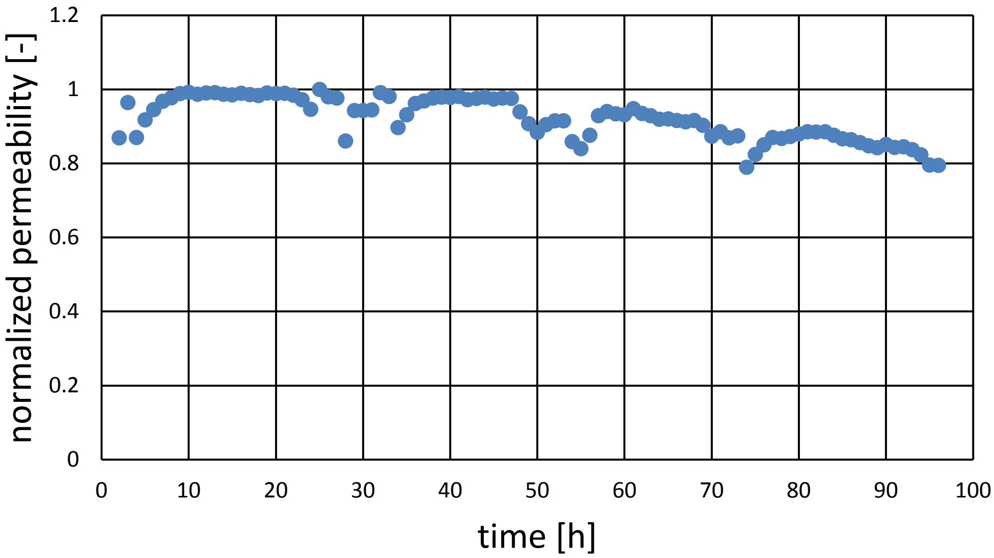

Fig. 5. Change of permeability [5]. (Flow rate = 1 ml/h)

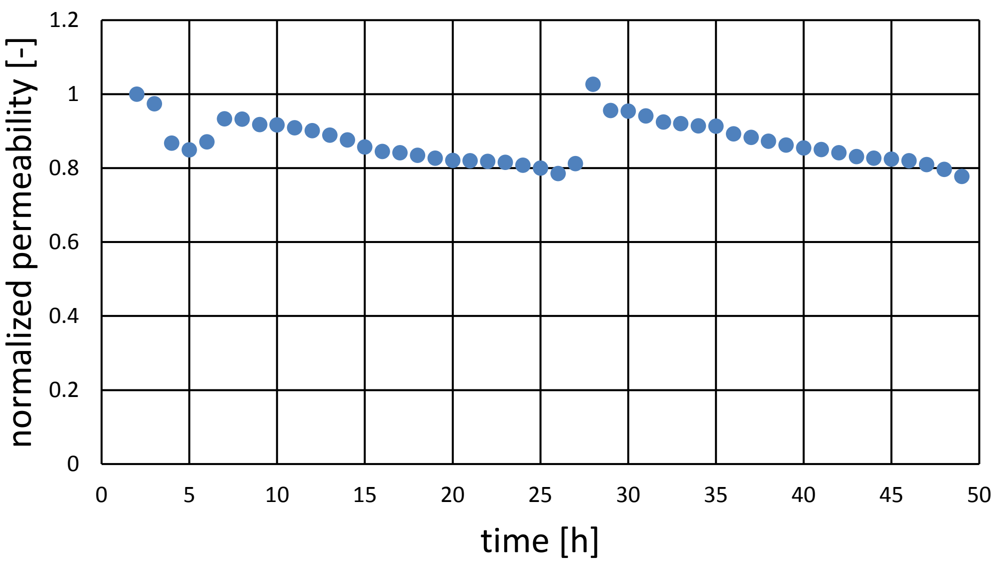

Fig. 6. Change of permeability [5]. (Flow rate = 2 ml/h)

Figure 7 summarizes the experimental results in Figs. 3-6 for the comparison of the permeability changes. The permeability at a steady state under the condition of 0.06 ml/h obviously became smaller than those under other conditions, and reached around one-tenth of the initial one. On the other hand, the decrease in the permeability did not depend on the flow rates in the range from 0.2 to 2.0 ml/h [5]. These results mean that most of CSH formed in the micro flow cell contributes the clogging of flow path because CSH hardly elutes under the condition of 0.06 ml/h. Besides, the permeability under the condition of 0.06 ml/h gradually decreased as a whole, while the permeability periodically increased and decreased. Such periodic changes in the permeability suggest that the clogging underwent with local deposition/moving of CSH repeated in flow paths even under the condition of such lower flow rate. Considering that the flow rate of the fracture in the granite rock is estimated to be about 0.1 ml/h by JNC report [9], these results in this study explain that, also under the flow condition of natural ground water, the clogging of flow-path will be facilitated with the deposition of CSH caused by the highly alkaline and Ca-rich condition around the repository. Such a dynamic behavior would form a natural barrier, bringing the retardation effect against the migration of radionuclides released from waste body.

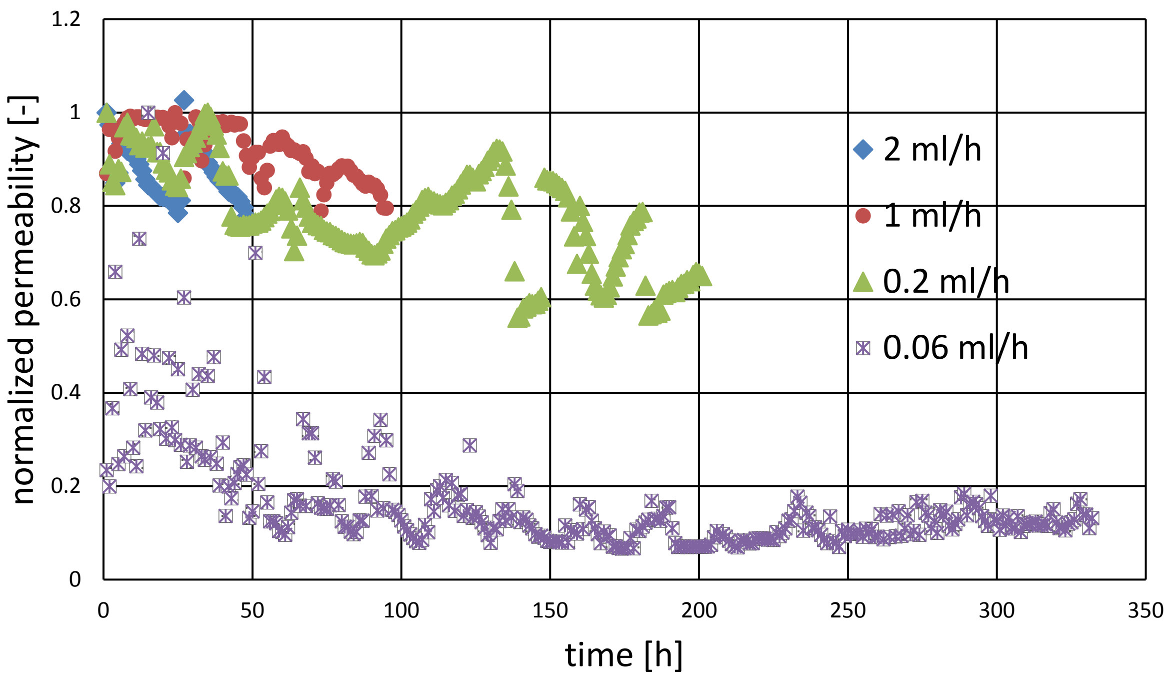

Fig. 7. Change of permeability. (summarized Figure 3-6)

CONCLUSIONS

This study examined the influence of flow rate on the clogging effect with the formation of CSH by using micro flow cell. From the experimental results, although the decrease in the permeability did not depend on the flow rates under the condition of the higher flow rate, the permeability under the condition of the lower flow rate (0.06 ml/h) significantly became smaller than those under other conditions. These results suggest that most CSH formed will contribute the clogging effect of flow path under the condition of the lower flow rate because most of CSH remain in flow path without flowing out. In addition, under the condition of 0.06 ml/h, the permeability gradually decreased with the periodical increase and decrease as well as those of the higher flow rates. This means that the clogging effect in flow path is occurred by repeating the local deposition of CSH and re-distribution with moving of CSH particles even if little CSH flows out of flow path. These results in this study show that under the highly alkaline and Ca-rich condition around the repository, the clogging of flow-path by the formation of CSH will contribute the retardation effect of radionuclide migration.

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