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Effect of Pozzolan on Cement-bentonite Interaction in Nuclear Waste Management

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ABSTRACT

In radioactive waste disposal facility, sodium-type bentonite will be used as an artificial engineered barrier material. However, during the tens of thousands of years, bentonite may be deteriorated and dissolved by alkaline water with the leached ions from the surrounding cement-based materials. It decreases swelling performance of the bentonite. In this paper, we propose a method to solve this problem by the mixing of pozzolanic materials such as fly ash and silica fume to the bentonite. These materials are expected to consume the leached calcium ions and reduce the deterioration of the bentonite. In this study, swelling performance of the bentonite specimen was measured using a one-dimensional swelling-deformation test under constant vertical pressure. As a result, it was found that the mixing of pozzolanic materials can reduce the deterioration of the bentonite in alkaline water from the viewpoint of swelling performance.

Keywords. Bentoite, swelling, engineered barrier, nuclear waste management

INTRODUCTION

In radioactive waste disposal facilities, sodium bentonite having high swelling capacity will be used as an artificial barrier material (JSCE, 2008). It is expected to delay the infiltration of groundwater and to fill cracks of the surrounding host rock. However, during the tens of thousands of years, the bentonite can be deteriorated by the alkaline water with the leached ions from the surrounding cement-based materials. It decreases swelling capacity of the bentonite. To reduce the decrease in the swelling capacity by the alkaline water, the authors have proposed two kinds of admixtures. One is the mixing of pozzolanic materials such as silica fume and fly ash to the sodium bentonite. These pozzolanic materials are expected to consume the leached calcium ions from the cement-based materials and to reduce the deterioration of the bentonite. The other proposal is the mixing of sodium carbonate to the bentonite. Here, calcite precipitation between the bentonite-based materials and cement-based materials was expected.

In this study, we investigated the effect of the mixing of fly ash and silica fume to bentonite on the swelling characteristics of the bentonite. To investigate the swelling characteristics, we used swelling-deformation tests proposed by Komine and Ogata (1994, 1999). Then, we measured swelling deformation of the compacted bentonites which were immersed in the distilled water or the saturated aqueous solution of calcium hydroxide.

OUTLINE OF EXPERIMENTS

Specimens. Table 1 shows the outline of specimens. In this study, we used fly ash (FA) or silica fume (SF) as an admixture. Then, we prepared three kinds of specimens; pure bentonite without admixtures, bentonite mixed with fly ash, and bentonite mixed with silica fume. In the experiment, the commercial bentonite, called Kunigel-V1, was used. The fly ash included 64.7% of SiO_2 and its specific surface area was 0.38 m^2/g. The mixing ratios of fly ash to dry bentonite were 5.0, 10.0, 15.0, and 30.0 mass%. The silica fume included 94.9% of SiO_2 and its specific surface area was 17.5 m^2/g. The mixing ratios of silica fume to dry bentonite were 0.5, 1.0, 2.0, 5.0, and 8.0 mass%. The water content of bentonite was set to 21 % by considering the value designed for a subsurface radioactive waste repository for low level wastes in Japan (JSCE, 2008). In this paper, the names of specimens indicated the composition of the materials, the mixing ratio of admixture, and the type of solution (W: distilled water, CH: saturated calcium hydroxide solution).

Table 1. Specimens

Name	Material composition (%)			Type of solution
	Bentonite	Fly ash (FA)	Silica fume (SF)	
Be1.3-W	100.0	0	0	Distilled water (W)
Be1.3FA-15-W	85.0	15.0	0	
Be1.3FA-30-W	70.0	30.0	0	
Be1.5SF-0.5-W	99.5	0	0.5	
Be1.5SF-2-W	98.0	0	2.0	
Be1.5SF-3-W	97.0	0	3.0	
Be1.5SF-5-W	95.0	0	5.0	
Be1.5SF-8-W	92.0	0	8.0	
Be1.3-CH	100.0	0	0	Saturated calcium hydroxide solution (CH)
Be1.3FA-5-CH	95.0	5.0	0	
Be1.3FA-10-CH	90.0	10.0	0	
Be1.3FA-15-CH	85.0	15.0	0	
Be1.3FA-30-CH	70.0	30.0	0	
Be1.5SF-0.5-CH	99.5	0	0.5	
Be1.5SF-1-CH	99.0	0	1.0	
Be1.5SF-2-CH	98.0	0	2.0	
Be1.5SF-3-CH	97.0	0	3.0	
Be1.5SF-5-CH	95.0	0	5.0	
Be1.5SF-8-CH	92.0	0	8.0	

In the mixing of materials, firstly, air-dried bentonite and admixture were mixed at low speed for 30 seconds using an Omni mixer. During the mixing, water contents of these mixtures were also adjusted by spraying distilled water. After that, they were mixed at high-speed for 60 seconds. Then, they were kept in a polyethylene bag for 24 hours. In the next day, these mixtures were mixed at high-speed for 60 seconds again, and we measured water contents of these mixtures.

In this study, cylindrical specimens having diameters of 60mm and heights of 5mm were prepared. The height of the specimens became small to save time for water absorption because the compacted bentonite specimens have low conductivities. The dry density was set to 1.3g/cm^3, which was relatively low density for the bentonite barrier in the nuclear waste management, to accelerate the degradation by the alkaline water. The sample was statically compacted by applying vertical pressure for several hours until the height of specimens became 5mm.

Swelling-deformation test. Figure 1 shows the schematic drawing of the test apparatus used in this study. Swelling characteristics of the bentonite specimens were evaluated by using the one-dimensional swelling-deformation test under a constant vertical pressure proposed by Komine and Ogata (1994, 1999). Iron plate was fixed to the top of the piston applying the constant vertical pressure because the targeted pressure in this study was small. We used a filter paper, of which the diameter is 60mm and the thickness is 0.19mm, and a porous metal, with 60mm diameter and 5mm thickness, as filters. The filters were placed below the top cap and on the pedestal. They were used for mitigating the outflow of bentonite particles from specimens.

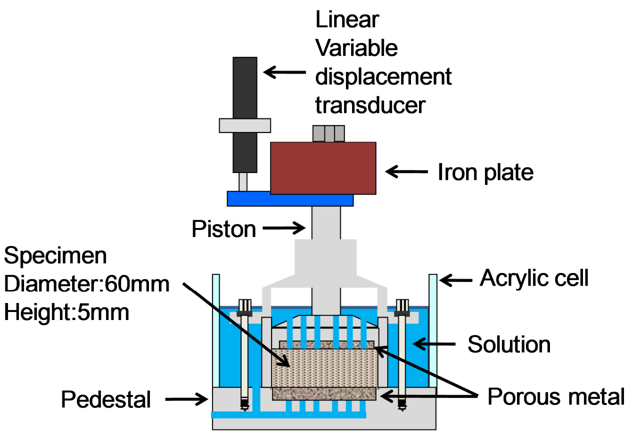


Figure 1. Schematic drawing of test apparatus

After applying the prescribed vertical pressure (10kPa) to the specimen, aqueous solutions were supplied to the specimens. Then, the relationship between the axial swelling deformation and the time required from the start of water supply was measured.

The solutions supplied to the specimens were two kinds of aqueous solutions. The first solution was the distilled water to measure basic performance. The second solutions were the saturated aqueous solutions of calcium hydroxide to discuss the effects of calcium ions leached from cement-based materials. To keep saturated concentration during measurement, additional calcium hydroxide powders were included in the solution.

The temperature during the experiments was set to 40 °C in order to promote pozzolanic reaction in the experiments. The solution was supplied from one side of specimens in this study. Measurement period was basically decided to be 7 days.

Evaluation method. The swelling strain measured in this study is defined by the following equation.

$$\varepsilon_S = \frac{\Delta S}{H_0} \times 100 \quad (1)$$

where ε_S is swelling strain (%), ΔS is swelling deformation (mm), and H_0 is initial specimen height (5mm).

It is known that the curves of swelling strain versus time can be approximated by the hyperbola in Eq.(2) (Komine and Ogata, 1994, 1999). Then, the maximum swelling strain ε_{smax}(%) is obtained from the asymptotic line of the hyperbola, and can be calculated by Eq.(3)

$$\varepsilon_s(\text{time}) = \frac{\text{time}}{a + b\text{time}} \quad (2)$$

$$\varepsilon_{smax} = \lim_{\text{time}\to\infty} \varepsilon_s(\text{time}) = \frac{1}{b} \quad (3)$$

where "time" is the duration (in day) from the start of water supply, ε_s(time) is swelling strain at "time" , and a and b are constants determined by fitting procedures.

RESULTS AND DISCUSSIONS

Effect of solution on pure bentonite. Figure 2 shows the time histories of swelling strains of the pure bentonite specimens measured in the swelling-deformation tests. The swelling strains were gradually increased with the supply of the solutions. Regarding to the influence of type of solutions, the strain of Be-CH was smaller than that of Be-W. The calculated maximum strains of the specimens Be-W and Be-CH were 223.6% and 115.7%, respectively. The decrease in the swelling capacity of Be-CH was caused by the degradation of bentonite associated with ion exchange with the supplied calcium ions.

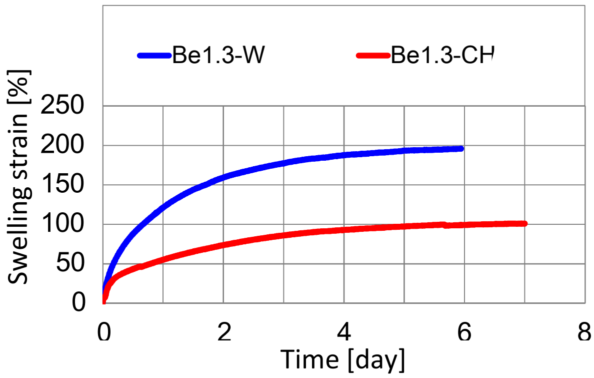


Figure 2. Time histories of swelling strains of pure bentonite specimens

Effect of mixing of fly ash. Figure 3 shows the time histories of swelling strains of the bentonite specimens mixed with fly ash. **Figure 4** shows the relation between the calculated maximum swelling strain and the mixing ratio of fly ash. Here, the solid and broken lines in the figure are the referential maximum swelling strains calculated from the maximum swelling strain of the pure bentonite specimens and the contents of bentonite in the mixed specimens. They assumed the maximum swelling strains proportional to the amount of pure bentonite in the specimens.

The measured swelling strains of the specimens in the distilled water were gradually decreased with the increase in the mixing of fly ash. Since the maximum swelling strain of Be1.3FA-15-W was plotted on the referential line, the decrease was explained by the decrease in the amount of the pure bentonite. On the other hand, the maximum swelling strain of Be1.3FA-30-W was plotted below the referential line. The solidification by the pozzolanic reaction of fly ash in the water might decrease in the swelling capacity.

The swelling strains of the specimens in the saturated calcium hydroxide solution were firstly increased with the increase in the mixing of fly ash and then decreased. The maximum swelling strain of Be1.3FA-10-CH showed highest in the specimens with fly ash in the calcium hydroxide solution because the consumption of the supplied calcium hydroxide by the pozzolanic reaction of fly ash could reduce the degradation of the bentonite. On the other hand, the swelling strain of Be1.3-FA-30-CH showed very small value. It means that the mixing of the large amount of fly ash did not reduce the deterioration of the bentonite due to the solidification by the pozzolanic reaction.

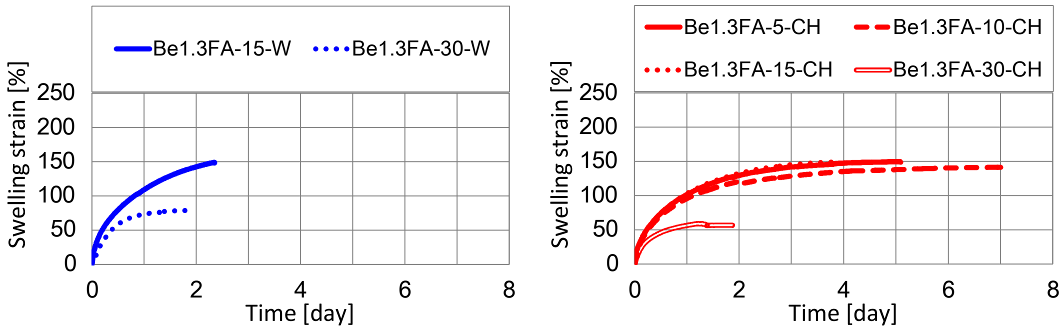


Figure 3. Time histories of swelling strains of bentonite specimens with FA (Left: in distilled water, Right: in saturated $Ca(OH)_2$ solution)

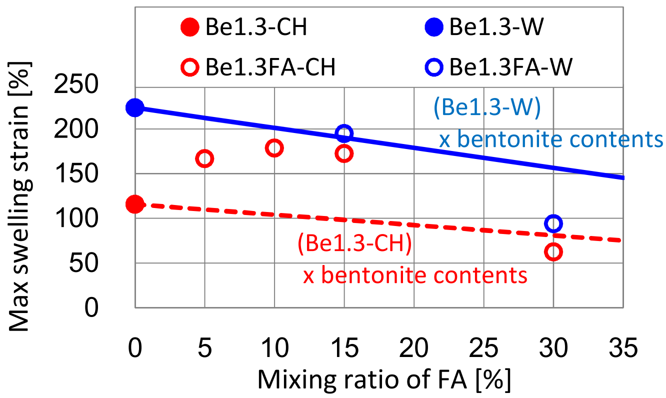
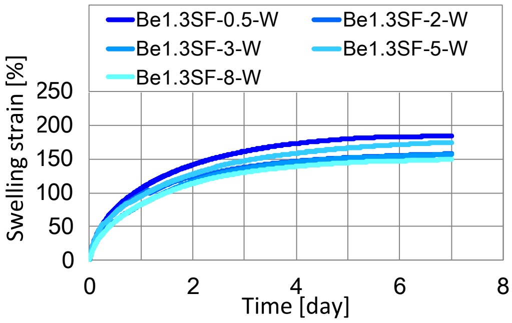


Figure 4. Relation between maximum swelling strain and mixing ratio of FA

Effect of mixing of silica fume. **Figure 5** shows the time histories of swelling strains of the bentonite specimens mixed with silica fume. **Figure 6** shows the relation between the calculated maximum swelling strain and the mixing ratio of silica fume. Here, the solid and broken lines in the figure are also the referential maximum swelling strains calculated from the maximum swelling strain of the pure bentonite specimens and the bentonite contents.

The maximum swelling strain of the specimens mixed with silica fume in the distilled water was smaller than the referential line. It means that the maximum swelling strain was decreased by the mixing of silica fume when the distilled water was supplied. We supposed that it was caused by solidification with the pozzolanic reaction of silica fume in the water same as BeFA-30-W and it was more significant on the silica fume having relatively high reactivity.

The maximum swelling strain of the specimens in the saturated calcium hydroxide solution became larger in the case of the mixing ration between 0.5 and 5.0 % than the referential line and showed relatively sharp peak at BeSF-1.0-CH. It means that the mixing of silica fume can strongly reduce the deterioration of bentonite by alkaline solution. When we mixing ratio of the silica fume became more than 1.0 %, the maximum swelling strain gently decreased as mixing ratio of silica fume increase. It was also caused by the decrease in the bentonite proportion in the specimen and solidification associated with the pozzolanic reaction of silica fume in the water.



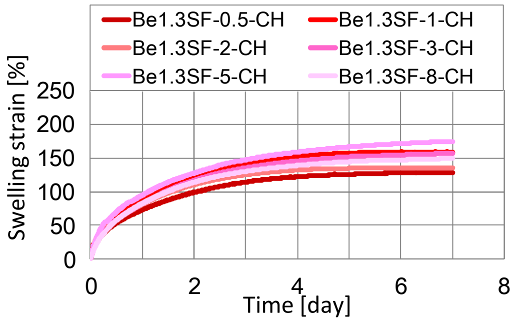


Figure 5. Time histories of swelling strains of bentonite specimens with SF (Left: in distilled water, Right: in saturated $Ca(OH)_2$ solution)

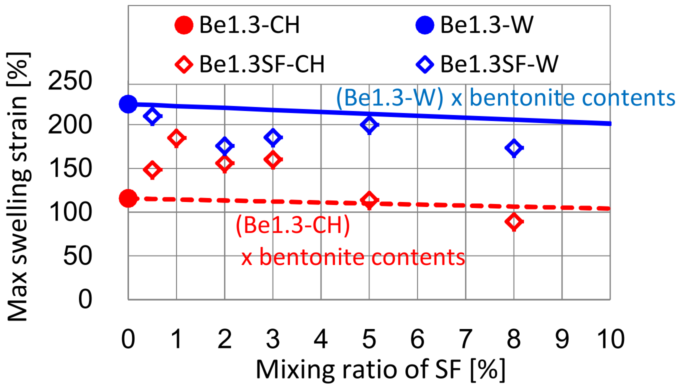


Figure 6. Relation between maximum swelling strain and mixing ratio of SF

CONCLUSIONS

In this study, we investigated the effect of admixtures of pozzolanic materials such as fly ash and silica fume to the bentonite in order to develop new materials reducing the deterioration of the sodium bentonite by alkaline water. The performance was evaluated based on the swelling deformation of specimens measured in the distilled water and the saturated calcium hydroxide solution. As a result, it was found that the mixing of pozzolanic materials can reduce the deterioration of the bentonite in alkaline water from the viewpoint of swelling performance. Especially, the mixing of the small amount of silica fume showed large reduction of the deterioration.

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REFERENCES

Japan Society of Civil Engineers (JSCE). (2008). “The method for setting nuclide migration assessment parameter used in groundwater migration scenario in safety assessment of sub-surface disposal.” (in Japanese)

Komine, H. and Ogata, H. (1994). “Experimental study on swelling characteristics of compacted bentonaite.” Canadian Geotechnical Journal, Vol.31, No.4, pp.478-490.

Komine, H. and Ogata, H. (1999). “Experimental study on swelling characteristics of sand-bentonite mixture for nuclear waste disposal.” Soils and Foundations, Vol.39, No.2, pp.83-97.