The Interaction between Contacting Barrier Materials for Containment of Radioactive Wastes

Hao-Chun Chang[1,a], Chun-Yao Wang[1,b] and Wei-Hsing Huang[1,c]

[1] Department of Civil Engineering, National Central University, Taiwan

[a]caterpies@hotmail.com, [b]pikachu.jim@gmail.com, [c]t321655@ncu.edu.tw

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Abstract. Zhisin clay is used as raw clay material in this study. This clay is mixed with Taitung area argillite to produce the backfill material for potential application as barrier for the disposal of low-level radioactive wastes. The interaction between concrete barrier and the backfill material is simulated by an accelerated migration test to investigate the effect of contacting concrete on the expected functions of backfill material. The results show backfill material close to the contact with concrete exhibits significant change in the ratio of calcium/sodium exchange capacity, due to the release of calcium ions from the concrete material. Also, some decreases in swelling capacity in the backfill material near the concrete-backfill interface were noted.

Introduction

The disposal of low-level radioactive wastes requires multi-barrier facilities to contain the wastes from contamination. Typically, the engineered barrier is composed of a concrete vault backfilled with sand/bentonite mixture. The backfill material is a mixture of bentonite and sand/gravel produced from crushing the rocks excavated at the site. With a high swelling potential, bentonite is expected to serve the sealing function, while the crushed sand/gravel improves the workability and stability of the mixture. Due to the nature of radioactive wastes, the disposal site is designed for a service life of 300 years or more, which is much longer than typical engineering or earth works. With such a long service life, the site is subject to groundwater intrusion and geochemical evolution, making the near-field environment evolution of the disposal site a complex problem.

In the vicinity of the concrete vault in a disposal site, the high-alkali concrete environment can cause changes in the pore solution and alter the nature of backfill materials. Although the interaction between concrete barrier and the backfill material does not affect the two barriers immediately, the interaction is reacting continually for a long time. The physical characteristics of the two barriers can be changed by the long-term interaction. Takafumi and Yukikazy [1] used a migration technique to simulate the interaction between different types of concrete barrier and the backfill materials. It was found that the swelling capacity is reduced with the increase of accelerated migration test periods for the bentonites. The swelling capacity of the bentonite in contact with fly ash concrete is higher than that with OPC concrete. Therefore, the interaction between the concrete and the backfill material needs to be assessed, such that the barriers serve the expected functions for a pro-long time. In this research, an accelerated migration test was devised to understand the effects of leaching from the concrete on the characteristics of backfill material. The two barrier materials (concrete and backfill) were placed in contact and then an electric gradient applied to accelerate the move of cations between the two barriers. A direct current was used for a composite specimen with cylindrical section in which an electrical potential gradient was applied. The physical characteristics of bentonite are carefully examined so as to assure that the long-term contact of these two barriers does not cause severe degradation. The analysis includes swelling capacity and calcium and sodium exchange capacity (CEC) of the bentonite material.

Materials and methods

Materials. A locally available Zhisin clay originated from Taitung, Taiwan was used as raw clay material in this study. Zhisin clay is mixed with Taitung area argillite, which is originated from the rock around the disposal site to produce the backfill material. The chemical compositions of the clay is given in Table 1. The Taitung area argillite was crushed to a maximum size of 2.36 mm and minimum size of 150 μm. The mix proportion of concrete used in this study is given in Table 2. The compressive strength of the concrete at the age of 28 days was determined to be higher than 35 MPa. Hardened concrete specimens with a diameter of 70 mm were sliced at a thickness of 30 mm for the accelerated migration test.

Table 1
Chemical compositions in Zhisin clay.

Compositions	(%)
SiO_2	55.4
Al_2O_3	20.1
Fe_2O_3	5.5
CaO	2.6
Na_2O	0.7
MgO	1.7
K_2O	1.4

Table 2
Mix proportions of concrete (kg/m^3).

Mineral	(kg/m^3)
Cement	243
Slag	88
Fly ash	88
Silica fume	28
Water	210
Coarse aggregate	1003
Fine aggregate	658
Water reducing admixture	4.4

Migration test. In this research, a migration technique was applied to accelerate the move of calcium ions from the pore solution of concrete so as to investigate the alteration of backfill material in contact with the concrete. A direct current voltage of 15 Volts was used for a composite specimen with cylindrical section in which an electrical potential gradient was applied. Fig. 1 shows a schematic diagram of the accelerated migration test. The cathode was embedded in the compacted bentonite gravel mixture, while the anode was immersed in a saturated calcium solution. Migration tests were conducted in a temperature controlled room (25 ± 2℃). The current was meansured by multitester when the accelerated migration test was in progress.

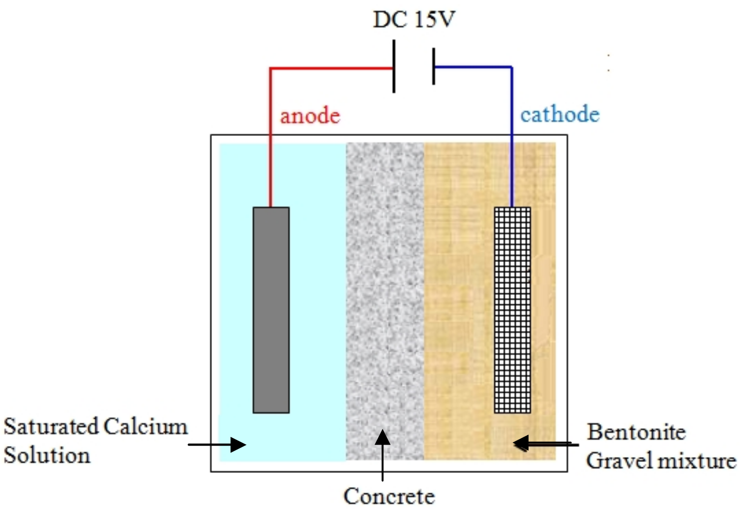

Fig. 1. Schematic view of accelerated migration test.

Analysis. At the end of the accelerated migration period, the composite cell was dismantled so that the specimens were ready for analysis. The compacted bentonite was sliced for the first layer to the seventh layer at different thicknesses. The thickness of the first to the fifth layer was 8 mm, and that of the sixth and the seventh layer was 15 mm for the sliced compacted bentonites.

In order to remove the sand in composite specimen, each slice of the backfill material was sieved such that only powder material with particle size less than 150 μm was used for testing of swelling capacity and exchange capacity of calcium and sodium (CEC). According to ASTM D5890, the powder samples of the bentonite were mixed with 100 mL distilled water and then the volume increase was measured after 24 hours. The swelling capacity was measured as an increase in the free volume of the bentonite. The CEC of Zhisin clay was measured by inductively coupled plasma (ICP) analysis.

Results and discussion

Cumulative electric charge on migration test. Fig. 2 shows the change of current with time in an accelerated migration test. The measured current became stable when the test periods lasted over 216 hours. In order to confirm the interaction has been completed between concrete barrier and the backfill materials, the accelerated migration test continued for 500 hours.

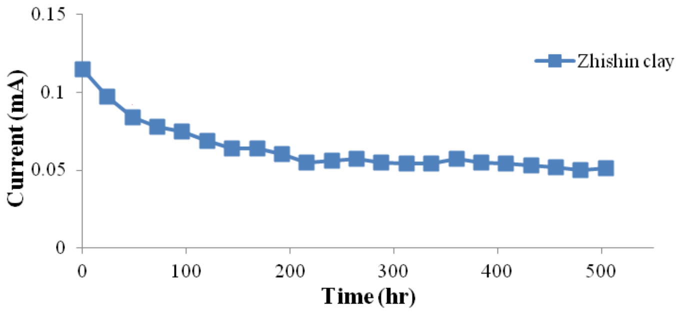

Fig. 2. Measured current during the accelerated migration test

The ratio of calcium to sodium content. Fig. 3 gives the exchangeable capacity of calcium and sodium in Zhisin clay along the depth of backfill materials. At the test period of 500 hours, the exchangeable capacity of calcium increased, while that of sodium remained at about the same.

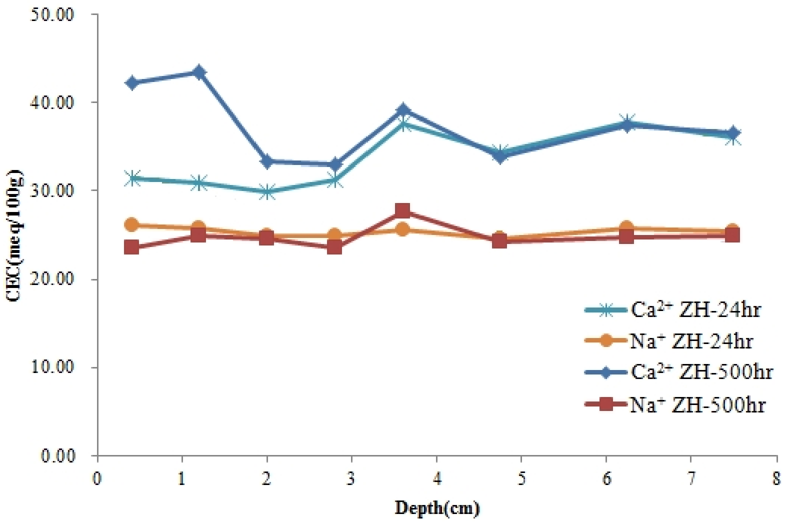

Fig. 3. The exchange capacity of calcium and sodium in Zhisin clay at different test durations

Fig. 4 shows the variation in the ratio of calcium to sodium exchangeable capacity in Zhisin clay. The interface between concrete and backfill is at depth 0 cm, and the depth in the horizontal axis represents the distance from the interface at which the exchangeable capacity was measured. In Fig. 4 the ratio of calcium to sodium content increases with the increase of accelerated migration test period. The ratio of calcium to sodium content at test periods of 500 hours for Zhisin clay shows an increase to 1.8. This indicates that there is an increase in the calcium content, especially in the layers close to the contacting interface. And the change in the ratio of calcium to sodium content in Zhisin clay occurs within 2.5 cm from the interface. As the distance from the interface increases, the change in the ratio of calcium to sodium content becomes less obvious.

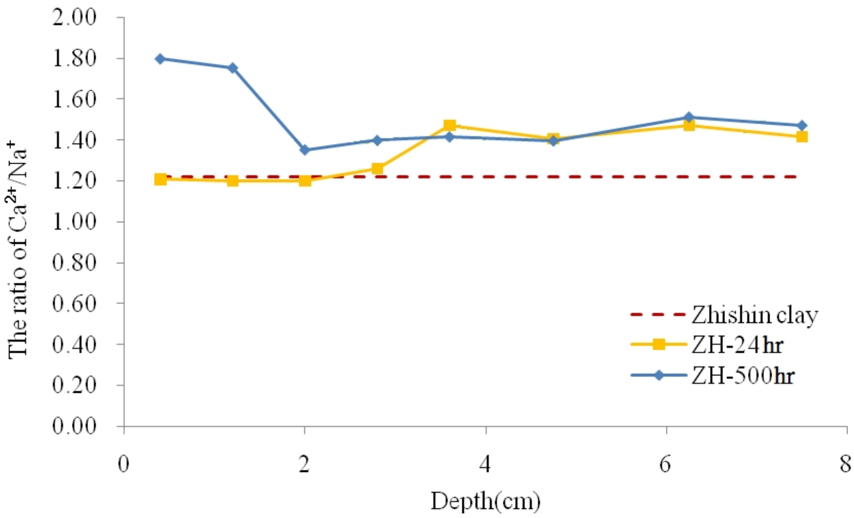

Fig. 4. Variation of the ratio of calcium/sodium content with distance from the interface.

Swelling capacity. The ions released from concrete tend to change the swelling capacity of bentonite. The change of the swelling capacity in the corresponding layer of the compacted backfill made with Zhisin clay is given in Fig. 5, the swelling potential of the raw bentonite material is shown in these figures in dash line.

It is observed that the swelling capacity is reduced with the increase of accelerated migration test period. In Fig. 5, the change of the swelling capacity in Zhisin clay occurs within a distance of 2.0 cm from the interface. This observation is most pronounced in the first layer at test period of 500 hours. The swelling capacity decreases to 6 mL/2g. This results from the accelerated migration of ions from the pore solution of concrete to backfill in an accelerated migration test. This ions react with montmorillonite, which is the main mineral component of bentonite, can be converted to non-swelling minerals such as zeolites, resulting in a decrease in swelling capacity [2]. And the shorter the distance from the interface, the more reduction in swelling capacity.

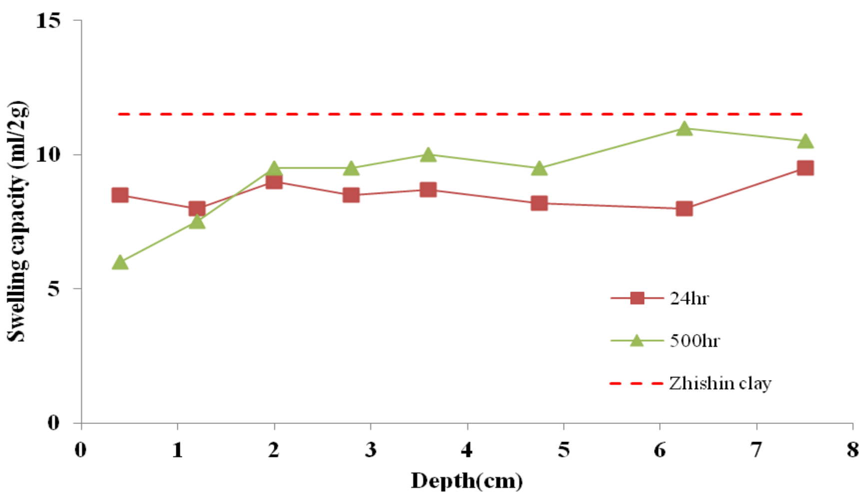

Fig. 5. Change of swelling capacity of Zhisin bentonite with depth.

Fig. 6 shows the relationship between the swelling capacity ratios (R_{EP}) and the ratio of cations using Eq. (1)

$$R_{EP} = \frac{EP}{EP_0} \qquad (1)$$

where EP is th swelling capacity at each layer and EP_0 is the mean value of the swelling capacity obtained from the pure Zhisin clay.The ratio of the swelling capacity reduces with the increase in the ratio of cations indicating that the swelling capacity has lowered when the relative content of calcium ions gets high. This tendency is more noticeable in the first and the second layers at test periods of 500 hours for Zhisin clay. The swelling capacity ratio of the layers were 0.52 and 0.65, respectively, while the corresponding ratio of cations are 1.80 and 1.75.

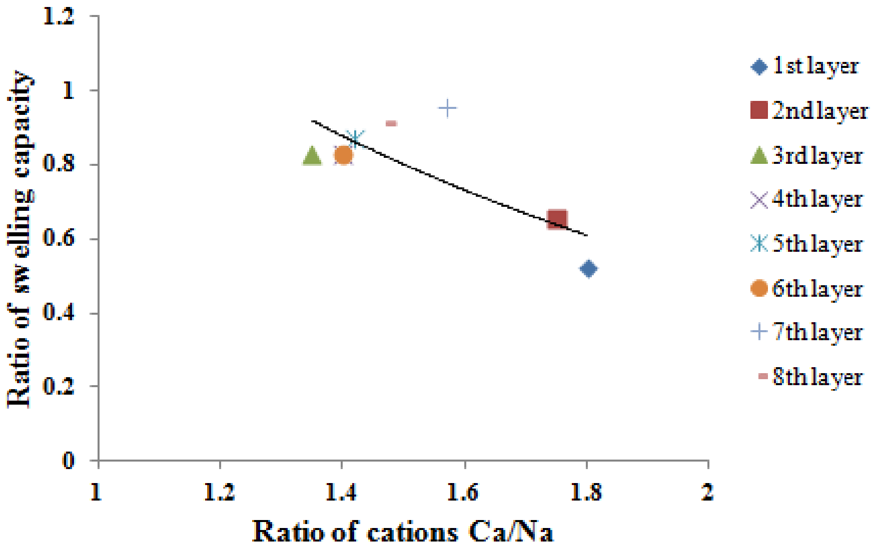

Fig. 6. Reduction of swelling capacity with increase in calcium ions.

Conclusions

In this research, a migration technique was applied to accelerate the migration of calcium ions from concrete to investigate the alteration of compacted Zhisin clay-sand mixture in contact with the concrete. The followings were concluded from the experimental results.

(1) The experimental results indicate that, the accelerated migration test could effectively simulate the long-term behavior of the interaction between concrete barrier and the backfill materials. At test periods of 500 hours, the interaction between concrete and backfill approaches completion.

(2) The migration of calcium from concrete results in reduction of swelling capacity of the contacting bentonite. And the shorter the distance from the interface, the more the ratio of calcium to sodium content increases. This alteration effect is more pronounced for bentonite material near the contact interface with concrete. The longer the distance from the interface, the effect of the migration of calcium on the bentonite decreases.

Acknowledgement

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References Acknowledgements

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