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Use of a migration technique to study alteration of compacted sand–bentonite mixture in contact with concrete

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ABSTRACT

In this research, a migration technique was applied to accelerate the migration of calcium ions from the pore solution of concrete so as to investigate the alteration of compacted bentonite in contact with the concrete. A 15 V of direct current was used for a composite specimen with 100 mm in diameter and 100 mm in length in which the electrical potential gradient was equal to 167 V/m. This composite consists of compacted bentonite (Na-type) sand mixture and concrete that is adjacent to each other. After a target cumulative electric charge of 13×10^3 C was reached, the specimen was removed for analysis. The results of electron probe micro analyzer (EPMA) showed that the concentration of calcium became higher in the compacted bentonite in the vicinity of the interface with the concrete. This observation was supported by the results of thermogravimetry/differential thermal analysis (TG–DTA) that provided distinct evidences of reduced quantities of $Ca(OH)_2$ in the concrete in the vicinity of the interface with the compacted bentonite. Therefore, calcium ions migrated from the concrete to the compacted bentonite under the given electric field. The results of the X-ray diffraction (XRD) curves indicated possible transformation of the characteristic of the compacted bentonite. However depending on the mix proportions of concrete employed, altered characterization of the compacted bentonite was different. It was found that the swelling capacity of the compacted bentonite in contact with normal concrete was decreased to 64% of its initial swelling capacity as the ratio of calcium content to sodium content (Ca/Na) increased up to 3.7. On the other hand the swelling capacity of the compacted bentonite in contact with fly ash concrete was 85% of its initial swelling capacity with a ratio (Ca/Na) of 2.3. Accordingly the use of the electrical migration technique enables quantitative evaluation of the reduced swelling capacity of the compacted bentonite in accordance with different types of hardened concrete.

1. Introduction

Long term stability of a radioactive waste repository is indispensable during the intended operation period. At present, for a sub-surface low level waste disposal facility in Japan geological repository with a given engineered barrier system is planned to construct at 50–100 m underground where horizontal tunnels of about 10 m in diameter are constructed (Tsuji and Sugiyama, 2005). It is considered that the engineered barrier system consists of a multi-layer system with a compacted bentonite sand mixture and cement-based materials (Tsuji and Sugiyama, 2005; Niwase et al., 2006). The compacted bentonite sand mixture is used as a buffer material for its low-water permeability, self-sealing characteristics due to its swelling capacity and the adsorption of radionuclides. These characteristics can be reinforced by the use of bentonite of which primary exchangeable cation is sodium ion (Na-type bentonite). On the other hand, several types of cement based materials are being considered in their application to engineered barrier system. Recent candidate for the barrier system under consideration is a multi-layer barrier system where cement-based materials will be used to sandwich a bentonite layer (Niwase et al., 2006). Cement based materials are expected to play an important role as a low diffusion layer at one side. For these materials concrete with dense microstructure as well as low alkalinity will be employed. In addition filling materials at another side are made of cement based materials. As a result, compacted bentonite will be continuously in contact with cement based materials.

It has been known that highly alkaline environments induced by the pore solution in concrete result in the alteration of the characteristic of benonite (Savage et al., 1992; JNC, 2005). In particular the characteristic of the Na-type bentonite is changed by cation exchange under calcium bearing solution (Tatematsu et al., 1979). In this way advantage of the Na-type bentonite in use for the engineered barrier system in Japan may be hampered by the presence of the concrete.

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In the past laboratory experiments synthetic pore solutions have been mostly used to study the possible alteration of the Na-type bentonite under highly alkaline conditions (Kubo et al., 1998; Kurosawa et al., 2002; Karnland et al., 2007). In this kind of immersion test, the liquid–solid ratio, the chemistry of the alkaline solution, the state of bentonite under immersion (e.g. in the state of fine particles or compacted mass) and conditions for the agitation in the liquid–solid mixture are considered to be some parameters to have influences on the results. In addition by considering current multilayer systems being planned for the LLW disposal facility in Japan it is expected that the alteration of compacted bentonite results from the diffusion of calcium ions through the interface with hardened concrete. Therefore, one likes to employ alternative experimental methods using compacted bentonite and hardened concrete to simulate the actual condition. However since it is concerned that the period of experiment will be lengthy using conventional diffusion tests (Sugiyama et al., 2008), a migration technique is considered as an alternative to accelerate the transport of calcium ions in concrete. For cement-based materials, electrical acceleration tests have been applied to study the deterioration due to the dissolution of portlandite and calcium silica hydrates (Saito et al., 1992). Other migration techniques were employed to determine the diffusion and dispersion coefficients of sodium ions and helium in compacted bentonite, respectively (Higashihara et al., 2004). However a migration technique has never been applied to a composite specimen of compacted bentonite and hardened concrete that are connected each other. Preliminary studies to investigate the advantage of using a migration technique have indicated that the calcium ions in the pore solution of concrete transported to the compacted bentonite sand mixture under a given electric potential gradient (Sugiyama et al., 2005).

This paper demonstrates the effective use of a laboratory experiment using a migration technique to study the alteration of compacted bentonite sand mixture in contact with different types of concrete. A composite specimen of hardened concrete and compacted bentonite that were connected each other was used. In order to investigate the effect of different concrete mixes on the swelling capacity of the Na-type bentonite fly ash concrete (FA concrete) was studied as well as normal concrete with ordinary Portland cement (OPC). It is generally expected that FA concrete provides the pore solution of low pH due to the pozzolanic reaction between the portlandite in the hydrated cement product and fly ash particles (Mahotra and Ramezanianpour, 1994). This means that the volume of the portlandite that is a primarily dissolved product in the hydrated cement system is reduced. After a given prescribed test periods, concrete and compacted bentonite were examined by chemical analysis, X-ray diffraction (XRD), thermogravimetry/differential thermal analysis (TG/DTA) and electron probe micro analyzer (EPMA).

2. Experimental methods

2.1. Preparation of specimen

2.1.1. Hardened concrete

Two types of concretes were investigated in this research, namely normal concrete (OPC concrete) and fly ash concrete (FA concrete). Chemical compositions of ordinary Portland cement (OPC) that was used for both concrete types are given in Table 1. OPC is specified in the Japanese Industrial Standard (JIS R5210). Fly ash used in this research is the Type II fly ash in accordance with JIS A 6201. The density of the fly ash was 2.35 g/cm^3 and the specific surface area was 3660 cm^2/g. The ignition loss of the fly ash was 1.6%.

Table 1
Chemical compositions for cement and bentonite (%).

	Cement[a]	Bentonite[b]
SiO_2	20.8	70.2
Al_2O_3	5.3	14.2
Fe_2O_3	2.9	2.5
CaO	64.6	2.0
MgO	1.3	2.2
Na_2O	0.3	2.5
K_2O	0.4	0.2
Ig-loss	1.9	5.2

[a] Ordinary Portland cement (JIS R5210).
[b] Na-type (Kunigel-V1).

Table 2 shows the mix proportions for OPC and FA concrete. The water to binder (cement plus fly ash) ratio was constant at 55%. For FA concrete OPC was partially replaced by fly ash by 30% in mass. Land sand was used for the fine aggregate with the density of 2.62 g/cm^3 while crushed stone was used for the coarse aggregate with the density of 2.89 g/cm^3, respectively. The maximum size of the coarse aggregate was 20 mm. The compressive strengths at the age of 28 days were 39.4 and 30.1 N/mm^2 for OPC and FA concrete, respectively. The strength of FA concrete would increase with time due to the pozzolanic reaction.

Concrete was placed in a rectangular steel mould with 730 mm in length, 150 mm in height and 50 mm in width as shown in Fig. 1. According to a standard curing method concretes specimens were sealed with a wet cloth and placed in a temperature controlled room at 20 ± 2 °C so as not to allow drying for a period of 28 days. Then concrete cores were taken so that cylinder shaped concrete specimens with 100 mm in diameter and 25 mm in height were obtained (Fig. 1). Before preparing a composite specimen with compacted bentonite sand mixture, the cylindrical concrete specimen was subjected to vacuum saturation.

2.1.2. Compacted bentonite and sand mixture

Chemical composition of the bentonite is given in Table 1. A Na-type bentonite (Kunigel-V1) was used in this research. According to current material design for a buffer material in Japan (JNC, 2000, 2005) sand was mixed with the Na-type bentonite. Sand inclusion can enhance the characteristic of compacted bentonite. In this research land sand was used with the maximum particle size adjusted below 1.2 mm using a given sieve size. Bentonite was mixed with sand by using an Omni-mixer according to a bentonite to sand mass ratio of 7–3. Distilled water was added as the mixing water so as to reach a water content of 15% in mass. In this research, the dry density of the compacted bentonite sand mixture was set to 1.9 g/cm^3. To obtain the target dry density, preliminary compaction tests were conducted with changing water contents and compaction energy (Sugiyama et al., 2005). Then, 15% of water content was selected. During the mixing, the water content was adjusted using a sprayer. After the mixing, the bentonite sand mixture was kept in a polyethylene bag in order to make its moisture content uniform for 1 day. By this treatment the formation of cracks was avoided after compaction.

2.1.3. Preparation of composite specimens

The bentonite sand mixture was placed and compacted on hardened concrete as the base of the composite specimen as shown in Fig. 2. Note that the interface of the concrete in contact with the compacted bentonite was chosen in such way that the steel mould surface touched its interface during the placement and curing of the concrete. In this way real interfacial zone in the multilayer system was simulated. The bentonite sand mixture was compacted by dropping a rammer weighting 2.5 kg from 300 mm above. The first layer was compacted with a compaction of 200 times/20 mm in