Dissolution Characteristics of Cement Paste Using Quick Setting Admixture for shotcrete

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Abstract

For the purpose of clarifying the dissolution characteristics of shotcrete used for tunnel lining of radioactive waste repositories, the cement was mixed with a quick setting admixture, and immersion tests were conducted. The quick setting paste was ground after hardening and contacted with pure water in a few months of testing. As a result, it was confirmed that more ettringite was produced in the cement paste with the quick setting admixture, which consisted mainly of calcium sulfoaluminate, than in the cement paste without the quick setting admixture. Immersion of the cement paste in water was also confirmed to result in the dissolution of Ca(OH)2 and monosulfate and further formation of ettringite. Furthermore, it was clarified that Calcium ions, Sulfate ions, and Aluminium ions are being consumed for the production of ettringite, and these ions do not readily leach to surrounding water. Based on these test results, a dissolution equilibrium model for shotcrete was developed in order to evaluate the durability of the barrier system or pH plume for the underground repositories.

1. Introduction

Hydration products in cementitious materials can dissolve, and the dissolved ions are leaching to the surrounding water, but owing to the slowness of this process, this is rarely considered a problem in ordinary structures. However, leaching is known as a deterioration mechanism of tunnels (Galana et al. 2019). On the other hand, in the case of radioactive waste disposal facilities for which the evaluation of extremely long-term stability is required, it has been shown that high alkaline components leached from cementitious materials can cause dissolution of minerals in the nearby bentonite and changes in the pore structure of the rock (Gaboreau et al. 2011), and therefore proper evaluation of leaching behavior is important. A site selection process for geological repositories for spent fuel, such as high-level waste (HLW), long-lived intermediate-level waste (L/ILW), and low-level radioactive waste (LLW) is underway all over the world (Alonso et al. 2010; Akiyama et al. 2011; Berner et al. 2013; Savage 2018; etc.). The safety of the HLW, L/ILW, and LLW repositories shall be ensured by a multi-barrier concept and distancing from living area. Shotcrete is essential material in most tunnels around the world.

Research on the dissolution of cement hydrates in ordinary cement has been carried out for a long time (Tayler 1964; Greenberg and Chang 1965; Atkinson et al. 1987; etc.). Given that shotcrete, which is used as a support material for underground spaces of radioactive waste disposal facilities, is in contact with the surrounding rock, shotcrete is considered to be a material that is relatively susceptible to alteration due to contact with groundwater among the cementitious materials used at disposal facilities (Luna 2007). In particular, high-strength shotcrete, which is used as a support material for large cross-section tunnels and deep underground spaces due to its excellent rapid setting and strength development, is mainly composed of calcium sulfoaluminate, and there is concern that sulfate ions may affect the chemical alteration of shotcrete itself and surrounding cementitious materials.

Regarding the chemical alteration of cementitious materials, it is generally understood that the main dissolved component is Ca. As mentioned above, chemical alteration due to dissolution of cementitious materials takes a long time, but as an empirical method, the dissolution behavior can be evaluated in a few months by conducting dissolution tests using finely crushed cementitious materials.

In this study, dissolution tests of cement paste with a quick setting admixture using calcium sulfoaluminate as the main component were conducted to evaluate the dissolution characteristics of shotcrete by analyzing the mineral composition of the residue and the ion concentration of the soaking water after immersion.

2. Test method

2.1 Constituent materials and mix proportions

The materials used and mix proportions of the paste are shown in Tables 1 and 2. The quick setting admixture is
composed mainly of calcium sulfoaluminate. The mix proportions of the cement paste were derived from the mix proportions of high-strength shotcrete, and the specified strength of the shotcrete is 36 N/mm².

### 2.2 Chemical composition of constituent materials and their analysis methods

The chemical compositions of the cement and quick setting admixture were analyzed. The respective samples were prepared by the glass bead method, and quantitative analysis was performed using X-ray fluorescence. The content of each element of cement was analyzed by using a calibration curve for the cement. The content of each element of the quick setting admixture was analyzed by using a calibration curve for gypsum. We couldn’t analyze the content of Si, Al, Fe, and Ti because it was included only Ca, S, Na, K, P for the calibration curve for gypsum. So the content of Si, Al, and Fe for the quick setting admixture was analyzed by applying the method for chemical analysis of gypsum (JIS R 9101), and the content of Ti and insoluble residue was analyzed by applying the methods for chemical analysis of cement (JIS R 5202). The chemical compositions of the cement and quick setting admixture are shown in Table 3. Since the quick setting admixture is composed mainly of calcium sulfoaluminate, it is characterized by large amounts of SO₃ and Al₂O₃. Table 4 shows the content of minerals in the cement paste calculated based on the chemical compositions shown in Table 3 and the mix proportions shown in Table 2.

### 2.3 Cement paste preparation method

The fresh properties of base cement paste are shown in the left of Fig. 1. As soon as the quick setting admixture used in this study is added to the base cement paste, the cement paste changed its fresh properties, as shown in the right of Fig. 1. Therefore, in this test, after the base cement paste was prepared, mixing was completed after stirring the cement paste for 10 seconds following the addition of the quick setting admixture. Next, the cement paste was poured into the mold that the size of φ50 × 100 mm vibrated with a table vibrator to ensure thorough filling. The mixing was carried out at the temperature of 20°C in the room, and the temperature of mixed mortar was measured as 23.1°C. The specimens were then sealed and cured in a thermohygrostatic chamber at 40°C and 60% RH until the age of 56 days so that the hydration proceeds sufficiently.

### 2.4 Preparation and water immersion of samples

After curing, the cement paste was crushed in a glove box filled with nitrogen gas to prevent reaction with the CO₂ in the atmosphere, and the particle size was adjusted to 1.2 to 2.5 mm. This cement paste was immersed in ion-exchanged water of the temperature of

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**Table 1 Materials used.**

| Material                  | Symbol | Remarks                                      |
|---------------------------|--------|----------------------------------------------|
| Water                     | W      | Ion-exchanged water                          |
| Cement                    | OPC    | Ordinary Portland cement, density of 3.16 g/cm³ |
| Quick setting admixture   | AC     | Quick setting admixture (calcium sulfoaluminate-type), density of 2.80 g/cm³ |
| High-performance water reducing admixture | SP | High-performance water reducing admixture (polyethylene glycol-type) |

**Table 2 Mix proportions of cement paste.**

| W/C (%) | Unit content (kg/m³) |
|---------|----------------------|
| W       | OPC                  |
| 45      | 540                  |
|         | 1202                 |

**Table 3 Chemical compositions of cement and quick setting admixture (mass %).**

| Material   | CaO | MgO | SO₂ | Na₂O | K₂O | SiO₂ | Al₂O₃ | Fe₂O₃ | TiO₂ | P₂O₅ | MnO | Ig.loss | Insol | Total |
|------------|-----|-----|-----|------|-----|------|-------|-------|------|------|-----|--------|-------|-------|
| OPC        | 63.61 | 2.40 | 2.02 | 0.19 | 0.33 | 20.65 | 5.18  | 2.94  | 0.28 | 0.22 | 0.02 | 17.7   |       | 100.0 |
| Quick setting admixture | -    | -   | -   | -    | -   | -    | 1.74  | 0.23  | 0.58 | -    | -    | 1.37   | -     | 99.7  |

**Table 4 Mineral content of cement paste with quick setting admixture.**

| W/C (%) | Unit content (kg/m³) | Mineral content (mmol/kg-paste) |
|---------|----------------------|---------------------------------|
| W       | OPC                  | AC | Na | K  | Ca | Al | Si | SO₂ |
| 45      | 540                  | 120 | 86 | 46 | 7817 | 945 | 2240 | 388 |

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Fig. 1 Fresh properties of cement paste.
20°C. Since the dissolution characteristics of minerals may differ depending on the amount of water in contact with the cement paste, dissolution tests were conducted using the ratio of the weight of the soaking water to the weight of the specimen to be immersed as a parameter. The test cases are shown in Table 5. In the cases where the liquid-solid ratio was set to 10 and 100, the mineral composition of the solid phase was evaluated at 14, 28, 56 and 196 days after the start of immersion to clarify the dissolution process of hydrates. The concentration of ions leached into the liquid phase used for immersion was also measured. In the cases where the liquid-solid ratio was 1, 5, 7, 20, 100, 2000, and 3000, the concentration of ions leached in the liquid phase was performed by the Rietveld analysis software, quantitative analysis of the mineral composition of the solid phase was analyzed using a powder X-ray diffractometer. The test cases are shown in Table 5. Focusing on the mineral composition before immersion, it can be confirmed that portlandite, ettringite, and monosulfate are produced. Usually, in the case of Ordinary Portland Cement, the generated ettringite is converted into monosulfate and hardly any ettringite remains (Hoshino et al. 2006). The peaks which show the presence of ettringite can be confirmed in the XRD charts.

Next, the change over time in the mineral composition due to immersion in water is described. Regardless of the solid-liquid ratio, a change in the peak, which may be due to the generation of ettringite, occurred. At the liquid-solid ratio of 100, the peak of portlandite clearly decreased with the passage of immersion time. Moreover, as the immersion period progressed, a broad peak was generated at 2θ of about 12°. The mineral at the top of the peak is monocarbonate, and it is thought that aluminate-based minerals are increasing. Further, at a liquid-solidification ratio of 100, the peak of C-S-H became prominent with the lapse of the immersion period.

Based on these results, it is thought that portlandite and monosulfate (3CaO · Al₂O₃ · CaSO₄ · 12H₂O) are leached from the paste when the cement paste containing the quick setting admixture is in contact with water for a long period of time, and ettringite and aluminate minerals are formed, and C-S-H is also newly formed in the region of high liquid-solid ratio.

3. Test results

3.1 Dissolution process of cement paste in water

(1) Changes in mineral composition of solid phase

The XRD patterns of the cement paste before immersion and after 14, 28, 56, and 196 days of immersion at the liquid-solid ratios of 10 and 100 are shown in Figs. 2 and 3. Focusing on the mineral composition before immersion, it can be confirmed that portlandite, ettringite, and monosulfate are produced. Usually, in the case of Ordinary Portland Cement, the generated ettringite is converted into monosulfate and hardly any ettringite remains (Hoshino et al. 2006). The peaks which show the presence of ettringite can be confirmed in the XRD charts.

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(2) Changes in ion concentration of the liquid phase

In this section, the results are summarized with a focus on the changes in ion concentration of the liquid phase over time. The changes in pH and Ca ion concentration over time at each solid/liquid ratio are shown in Fig. 4. The pH was constant at 12.8 for the liquid-solid ratio of 10 and 12.5 for the liquid-solid ratio of 100. The Ca ion concentration in the liquid phase was stable at about 15
mmol/l regardless of the liquid-solid ratio. The pH of the saturated Ca(OH)\textsubscript{2} solution is about 12.5, and the liquid phase is considered to be nearly saturated with portlandite for both liquid-solid ratios of 10 or 100.

Next, the changes in Na ion and K ion concentrations over time are shown in Fig. 5. Regardless of the liquid-solid ratio, the concentrations of Na and K ions in the liquid phase were almost constant from 14 days of immersion onward. Further, regardless of the liquid-solid ratio, Na ions were dissolved about twice as much as K ions, which is proportional to the mineral ratio of Na and K as shown in Table 4. Moreover, the concentrations of Na ions and K ions dissolved at the liquid-solid ratio of 100 were about 1/10 of those at the liquid-solid ratio of 10. Based on the above, it is considered that all of the leachable Na ions and K ions in the cement paste were leached into the solution at an early stage.

Finally, the changes in Al ion and sulfate ion concentrations over time at each solid-liquid ratio are shown in Fig. 6. Regardless of the liquid-solid ratio, although sulfate ions were dissolved during the early stage of immersion, subsequently, they were not detected in the liquid phase. This is thought to be due to the reaction of sulfate ions with some other ions, causing return to the
solid phase. Considering the fact that only trace amounts of Al ions were detected, along with the results of Figs. 2 and 3, these ions were probably consumed in the ettringite formation reaction.

### 3.2 Ion leaching characteristics according to the amount of acting water

The relationships between the liquid-solid ratio and the various ion concentrations after 70 days of immersion are shown in Figs. 7 and 8. According to Fig. 7, the pH decreased as the liquid-solid ratio increased. Furthermore, it can be seen that the slope of the decrease in pH changes at a liquid-solid ratio of 100. According to Fig. 8 on the left, in the region of small liquid-solid ratio, the concentrations of Na ions and K ions are high, and that of Ca ions is low. This is thought to be due to the high solubility of NaOH and KOH, which resulted in the prior dissolution of Na and K ions, thus limiting the dissolution of Ca ions. On the other hand, given the low content of Na ions and K ions as shown in Table 4, it is considered that all Na ions and K ions are leached out when the liquid-solid ratio becomes large, and then Ca ions are leached out from portlandite, which has the next highest solubility. When the liquid-solid ratio is 100 or more, the Ca ion concentration in the liquid phase decreases. It means that the pH fell below 12.5 due to the dissolution of portlandite. The decrease in pH at a liquid-solid ratio of 100 or higher is due to the dilution of hydroxide ions from portlandite. This is thought to indicate that the concentration of Ca ions in the liquid phase is not maintained and decreases because portlandite is completely dissolved and the dissolution of C-S-H, which has low solubility, becomes dominant (Buil et al. 1992).

According to the figure on the right in Fig. 8, sulfate ions and Al ions hardly dissolve regardless of the liquid-solid ratio. This is thought to be due to the fact that these ions were used for the formation of ettringite and aluminosilicate minerals, as shown in the results of solid-phase analysis at the liquid-solid ratios of 10 and 100. Although Si ions hardly leached out when the solid-liquid ratio was low, the concentration of Si ions in the liquid phase increased when the solid-liquid ratio was 1000 or higher. In the region where the liquid-solid ratio is 1000 or higher, dissolution and alteration of amorphous materials such as C-S-H and C-A-S-H are considered to have occurred.

![Fig. 7 Relationship between liquid-solid ratio and pH in the liquid phase.](image1)

![Fig. 5 Relationship between immersion period and Na⁺ and K⁺ concentrations in the liquid phase.](image2)

![Fig. 6 Relationship between immersion period and Al³⁺ and SO₄²⁻ concentrations in the liquid phase.](image3)
3.3 Dissolution characteristics of shotcrete

Based on a previous study by one of the authors and others (Yokozeki et al. 2004), the changes in the ions leached into the liquid phase and the mineral composition of the solid phase when water acts on a cement paste simulating shotcrete were summarized by focusing on the ratio of the Ca ion concentration leached into the liquid phase to the Ca element concentration remaining in the solid phase. According to previous studies by one of the authors and others (Yokozeki et al. 2004; Nakarai et al. 2006; Buil et al. 1992), the relationship between the concentration of Ca ions in the solid and liquid phases can be divided into two regions: one due to the dissolution of portlandite, and the other due to the dissolution of C-S-H after the portlandite is completely dissolved. In the region where the dissolution of portlandite is dominant, the Ca ion concentration in the liquid phase is maintained at a high concentration, and the Ca ions in the solid phase gradually decrease. In contrast, in the region where C-S-H dissolution dominates, Ca ions are less likely to leach into the liquid phase because of the low solubility of C-S-H, and Ca ions in the solid phase decrease with low concentration in the liquid phase. These processes were also sorted out and described in a comprehensive experiment by Berner et al. (1990).

In this study, the solid phase Ca concentration ratio \( \frac{C_{\text{p}}} {C_{\text{p}0}} \) was calculated from the amount of Ca present in the solid phase before immersion \( (C_{\text{p}0}) \), which was calculated based on Table 4, and the Ca ion concentration leached into the liquid phase after the dissolution test \( (C) \). Figure 9 shows the relationship between the Ca ion concentration in the liquid phase and the ratio of Ca concentration remaining in the solid phase in the cement paste simulating shotcrete. The model in the figure was set as follows based on one of the authors’ previous research (Yokozeki et al. 2004). In addition, Fig. 9 shows the data of Ordinary Portland Cement paste obtained in the author’s previous research (Yokozeki et al. 2004).

When \( A_{\text{ap1}} < C_{P}/C_{P0} < 1.0 \), \( C = C_{0} \).
When \( C_{P}/C_{P0} < A_{\text{ap1}} \), \( C_{P}/C_{P0} = A_{\text{ap1}}(C/C_{0})^{1/n} \) \( (C_{1} < C < C_{0}) \), \( C = C_{1} \) (when \( C \leq C_{1} \)).

Here, \( C_{P} \) : Solid phase Ca concentration per unit volume of paste (mmol/l)
\( C \) : Ca ion concentration in the liquid phase (mmol/l)
\( C_{P0} \) : Initial solid phase Ca concentration per unit volume of paste (mmol/l)
\( A_{\text{ap1}} \) : Coefficient related to the composition ratio of \( \text{Ca(OH)}_{2} \) and C-S-H (-)
\( n \) : Coefficient related to dissolution equilibrium of C-S-H (-)
\( C_{0} \) : Liquid phase Ca ion concentration at the time of \( \text{Ca(OH)}_{2} \) dissolution (mmol/l)
It can be calculated by the solubility product \( K_{sp} \) about experiments of OPC
\( C_{1} \) : Liquid phase Ca ion concentration at the completion of C-S-H dissolution (mmol/l)

Of the above coefficients, \( A_{\text{ap1}}, n, C_{0}, \) and \( C_{1} \) need to be set so as to match the experimental results. In a previous study by the authors (Yokozeki et al. 2004), the respective values of Ordinary Portland Cement paste at

![Fig. 8 Relationship between liquid-solid ratio and concentration of ions dissolved in the liquid phase.](image)

![Fig. 9 Solid-liquid equilibrium model for calcium of shotcrete.](image)

Table 6: Coefficients in solid-liquid equilibrium model.

| Ref.          | Cement | A<sub>ap1</sub> | n  | C<sub>0</sub> | C<sub>1</sub> |
|---------------|--------|----------------|----|--------------|--------------|
| Yokozeki et al. 2004 | OPC    | 0.82           | 4.5| 21.75        | 1.5          |
| This study    | Shotcrete | 0.82           | 20 | 21.75        | 1.5          |
the temperature of 20°C were calculated as shown in Table 6. In the cement paste using the quick setting admixture, the value of “n” was only different from the values of Ordinary Portland Cement paste. The difference indicates that the mineral composition of the cement paste using a quick setting admixture is less prone than that of Ordinary Portland Cement paste to leach from the solid phase into the liquid phase. As described above regarding changes in mineral composition, ettringite is formed along with the dissolution of portlandite. The cement paste, due to the formation of ettringite, has very low solubility (Warren et al. 1994).

Further, in the present study, there is a region where the Ca ion concentration in the liquid phase is lower than that of C3, in the range where the liquid-solid ratio is small, and the Cp/Cp0 value is close to 1.0. As shown in Fig. 8, this is the region where Na ions and K ions are leached into the liquid phase first, indicating that Ca ion leaching is suppressed in this region. In other words, in the region where the effect of water is small, Ca ion leaching into the liquid phase is considered to be suppressed to a greater extent compared to the dissolution equilibrium model shown in Fig. 9. To further refine the model, it is necessary to consider the effect of suppressing Ca ion leaching due to the influence of Na ions and K ions.

4. Conclusions

In this study, dissolution tests of cement paste with a quick setting admixture using calcium sulfoaluminate as the main component were conducted to evaluate the dissolution characteristics of shotcrete by analyzing the mineral composition of the residue (solid phase) and the ion concentration of the soaking water (liquid phase) after immersion. As a result, it was confirmed that, during the curing process, more ettringite was produced in the cement paste with the quick setting admixture, which consisted mainly of calcium sulfoaluminate, than in the cement paste without the quick setting admixture. Soaking of the cement paste in water was also confirmed to result in the dissolution of Ca(OH)₂ and monosulfate and further formation of ettringite from the solid phase into the liquid phase. As described above regarding changes in mineral composition, ettringite is formed along with the dissolution of portlandite. The cement paste, due to the formation of ettringite, has very low solubility (Warren et al. 1994).

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