

Ionic Diffusivity and Pore Structure of Hardened Cement Paste Exposed to High Temperature Environment for Long Period

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1 Introduction

The structure of C-S-H can change due to the hydration and polymerization reactions under the influence of temperature. Consequently, changes in the microstructure influences the ionic diffusivity in the hardened cement. Evaluation of ionic diffusivity is also a key issue for the design and assessment of a radioactive waste disposal system with a cement-based barrier.

Especially, the influence of high temperature on the performance of a cement-based barrier should be thoroughly examined to build a disposal system with high reliability and safety. The temperature of the action depends on the actual concentration of exothermic radionuclides such as cobalt-60 in nuclear waste, assumed to be around 60°C below the design temperature, to prevent excessive influence of temperature on the disposal system. The duration of the exposure to the high temperature can extend over several decades or hundred years. However, until now, few studies have reported on the influence of long-term high temperature on the ionic diffusivity of hardened cement. Therefore, it is necessary to gain deeper understanding on the behavior of the hardened cement under the high temperature.

The purpose of this study is to investigate the ionic diffusivity of hardened cement exposed to high temperatures over a long period. From the results of the investigation, we discuss the effectiveness of a prime candidate material for the disposal system.

2 Experiments

Two specimens, OPC and LF30, of cement paste were prepared with water binder mass ratio of 0.45 were prepared. The OPC specimen was made of ordinary Portland cement. The LF30 was made of low-heat Portland cement, fly ash, and limestone filler. LF30 is a prime candidate mix for the nuclear waste disposal system. Specimens were stored in a saturated atmosphere at 20°C for one year. After the curing, the specimens were immersed in lime-saturated water at 20, 40, 50, 60, and 80 °C for one year.

Adsorption tests with N₂ gas and water vapor after exposure were carried out to investigate the change in microstructure of hardened cement paste, then pore size distributions were calculated by BJH method and the BET surface areas were obtained. The effective diffusion coefficient of Cl⁻ ions of the specimens after exposure was also measured by the steady-state penetration-type diffusion test.

3 Results

- In the case of OPC, the pore size distribution determined from N₂ gas desorption isotherms using the BJH method shows obvious temperature-dependent coarsening of the microstructure of hardened cement. In the case of LF30, temperature had a negligible influence on the microstructure, except at 80°C. (Figure 1).
- BET surface area of OPC calculated from N₂ gas and water vapor adsorption isotherms decreased with temperature. In contrast, the BET surface area of LF30 had low sensitivity to temperature. (Figure 2)
- The Cl⁻ ionic diffusion coefficient of OPC increased with temperature. The coefficient of LF30 is nearly four orders of magnitude lower than that of OPC below 50°C. However, the value slightly increased at 60°C and significantly at 80°C. (Figure 3).

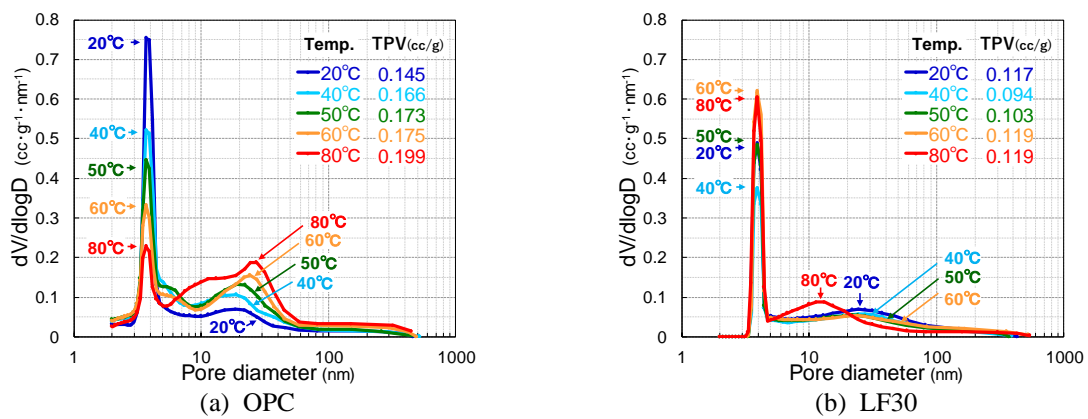


Figure 1. Pore size distributions calculated from N₂ gas desorption isotherms.

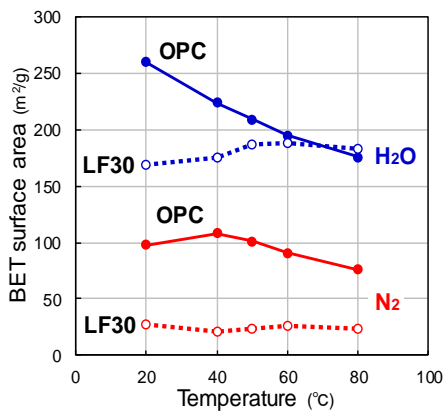


Figure 2. BET surface areas obtained from N₂ gas and water vapor adsorption isotherms.

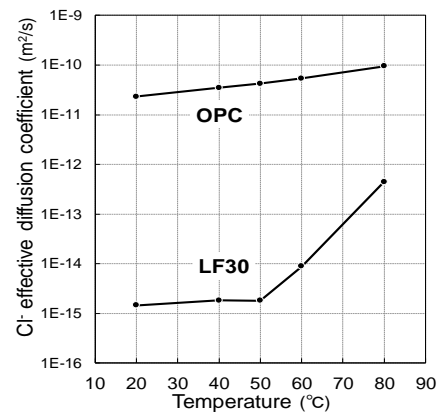


Figure 3. Effective diffusion coefficients of Cl⁻ ions.

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References

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