Imaging of deuterium-labeled Mg-bearing hydration products by Isotope microscope

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For the engineered barrier that will be employed in geological disposal of radioactive waste generated from spent fuel reprocessing, the utilization of cement and bentonite are considered. Hence, the understanding of the influence of groundwater to the engineered barrier is critical in evaluating long-term stability of the designed function of the geological disposal. Especially, coastal areas are currently considered as a candidate site in Japan. Therefore, it is necessary to understand the influence of groundwater that has the composition of seawater that may reach the engineered barrier. Magnesium will be derived from seawater, and react with silica from cement and bentonite barrier. It is required to clarify the behavior of interaction between groundwater and barrier material. In previous studies, it is suggested that the magnesium silicate hydrate (M-S-H) is formed by the dissolution of cement by seawater derived groundwater, and by the alteration of bentonite due to the interaction with high alkaline cement pore water (Fernandez et al., 2009; Kosakowski and Berner, 2013). Although many researchers reported M-S-H is a low crystalline hydration product present as a metastable phase, the properties, conditions of formation and solubility of such M-S-H have not been investigated well.

Since M-S-H contains hydrogen in its structure (Nied et al., 2016), isotope labeling using deuterium atom instead of hydrogen atom may be possible. Utilization of heavy water in place of ordinal water as a reaction solution in the M-S-H synthesis could achieve the labeling, since ordinal water only contains 0.015% of deuterium. It may enable to clarify the hydration process by tracing M-S-H formation. Application of "Isotope microscope" to observe the synthesized products, would enable the image analysis that can make possible to distinguish the magnesium silicate hydrate formed with hydrogen and deuterium. Isotope microscope is the analyzer combining secondary ionization mass spectrometer (SIMS) and stacked CMOS active pixel sensor (SCAPS), which makes possible to image isotopes of the ppb scale in micro areas. By Isotope microscopic observation, hydrogen isotope imaging could be performed on micro scale hydration products incorporating deuterium, and the hydration reaction process can be observed from highly accurate imaging of deuterium distribution. This imaging method for hydration products have never been done for cementitious materials, so firstly we checked whether this method could be applied to the hydration reaction in a simple system.

In deuterium-labeling experiment of magnesium hydroxide ($Mg(OH)_2$), we dipped magnesium oxide in light water for 3days and in heavy water for latter 3days. By isotope microscope observation, we could get the deuterium distribution image and deuterium was concentrated in magnesium hydroxide on the surface of magnesium oxide particles. We used heavy water only during the latter three days and hydrogen isotope exchange from $Mg(OH)_2$ to $Mg(OD)_2$ is difficult to occur (Aimoto et al., 2014), so it is considered that deuterium-labeled magnesium hydroxide is generated during that period. It was possible to clarify the distribution of hydration products generated during heavy water use by this method. It is suggested that the deuterium labeling method can also be applied to hydration products in the Mg-Si system and more complicated systems, and it is expected that we can clarify the generation timing and generation location of hydration products including M-S-H.

In this presentation, we will present the observation results on deuterium-labeled magnesium hydroxide which is a starting material of M-S-H.

Keywords: hydration products, isotope microscope, hydrogen isotope