

Mineral size distribution modeling during dissolution of montmorillonite

YAMAGUCHI, Kohei^{1*}; MIURA, Hitoshi²; SATOH, Hisao¹

¹Mitsubishi Materials Co., ²Nagoya City University

In the geological disposal of radioactive waste, the waste is sealed by cement-based materials and bentonite-based material to prevent leakage into environment. The bentonite-based material protect the radioactive waste from the groundwater flow around the geological disposal area, so its low permeability should be maintained for a long term. The low permeability could be achieved by the swelling of montmorillonite in the bentonite-based material. However, montmorillonite will dissolve by a reaction with high-alkaline pore water, spoiling the low permeability of the bentonite-based material. In addition, precipitation of secondary minerals such as zeolite will promote the dissolution of montmorillonite through changes in composition of the pore water. In order to assess the long-term permeability of the bentonite-based material, it is necessary that the dissolution of montmorillonite and crystallization of secondary minerals are comprehended over a long time of several tens of thousands years.

In the pore water, there are numerous montmorillonite particles of various sizes. When montmorillonite of various sizes co-exists in the same solution, the smaller particle dissolves faster than the larger one because of the Thomson-Gibbs effect. The mean size of montmorillonite will increase gradually, leading to a delay of further dissolution. In addition, an evolution of size distribution is also important for the crystallization process of the secondary minerals, e.g., zeolite. Since zeolite is not present in the initial solution, the crystallization process is described in the nucleation and subsequent growth. Evolution of the size distribution of zeolite affects the dissolution of montmorillonite through changes in solution composition. This implies that the evolution of the size distribution of montmorillonite and zeolite should be considered to assess the long-term behavior of the permeability of the bentonite-based materials. However, in the previous chemical equilibrium calculations, the evolution of the size distribution has not been considered.

In this report, we numerically modeled the time evolution of the size distribution of montmorillonite due to dissolution according to a theoretical model described in Yao et al. (1993). The crystallization of zeolite was neglected as a first step. We consider the dissolution of montmorillonite in a closed system. The evolutions of the size distribution, bulk concentration of solution, and mean radius of montmorillonite were successfully calculated.

The model given in this report is a model in a closed system. On the other hand, the geological disposal environment is not a closed system because there is an actual mass transfer due to the flow of groundwater and diffusion. To couple the local mineral dissolution/crystallization and the global mass transfer, some chemical reaction-mass transfer calculation codes have been developed. However, these codes assumed chemical equilibrium, so the evolution of the size distribution of minerals did not considered. The evolution of the size distribution of minerals would significantly affect the long-term behavior of the permeability of the bentonite-based materials. Therefore, it is important to compare the calculation results of the model with the evolution of the size distribution and chemical equilibrium calculation result.

Keywords: montmorillonite, dissolution, Mineral size distribution