Numerical calculation of montmorillonite dissolution in consideration of the size distribution

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In recent years, how to dispose of large amounts of radioactive waste that were generated with the use of nuclear power plants has become an issue. In Japan, a geological disposal has been studied as one of the feasible disposal methods. In the geological disposal, the high-level radioactive waste in glass solidified body is placed in a steel container, then covered with a cushioning material mainly composed of clay minerals (montmorillonite) of bentonite, and it is disposed of the underground deeper than several hundred meters. Montmorillonite expands in contact with water and therefore it has a function to protect the radioactive waste from groundwater. However, during a long period spanning several thousand years, the reaction with the groundwater causes the dissolution of montmorillonite, so there is a risk that the radioactive material of the inside is flowing out by groundwater. Therefore, it is very important to predict the dissolution behavior of montmorillonite for the long-term stability of storage.
In the present study, we carried out the numerical calculation in order to examine the dissolution behavior of montmorillonite. We assumed that montmorillonite particles with various sizes are exposed to the alkaline solution under closed environment. The size change of each particle by dissolution and the concentration change of the solution were calculated. We considered that the dissolution rate depends on the degree of supersaturation of the solution, and the particle size (Gibbs-Thomson effect). When the solution is unsaturated, the dissolution rate is given by an empirical formula obtained by dissolution experiments [1]. When the solution becomes supersaturated, we assumed that the montmorillonite particles are turned to growth and the growth rate is given by the same form to the dissolution rate except of its sign. The degree of saturation of the solution was calculated by using the PHREEQC [2]. As the initial particle size distribution, we examined the following four cases: uniform, log-uniform, normal and log-normal distributions.
Let us describe the numerical results of the case that the initial size distribution is log-normal. At the beginning of the calculation, all sizes of particles are dissolved in the unsaturated solution. The particle dissolution increases the degree of supersaturation, and eventually the solution becomes supersaturated. The small particles continue to be dissolved by the Gibbs-Thomson effect, however, the larger particles are turned to growth. The period that is required for the larger particles begin to grow depended on the ratio of the total particle volume to the solution volume (solid-liquid volume ratio) at the beginning of the calculation. In the case of the solid-liquid volume ratio is 10^{-5}, the larger particles are turned to growth after about 7000 years. With increasing the solid-liquid volume ratio, it was found that the period is reduced to about 10 to 100 years. It was also found that the lower limit of the size above which the montmorillonite particles are turned to growth increases with increasing the solid-liquid volume ratio. Thus, by the numerical calculation of the dissolution process of the different-sized montmorillonite particles, we concluded that it is important to consider the Gibbs-Thomson effect for the long-term stability of storage.
References: [1] Cama et al. (2000), Geochem. Cosmochim. Acta 64, 2701. [2] PHREEQC -A Computer Program for Speciation, Batch-Reaction, One-Dimensional Transport, and Inverse Geochemical Calculations. http://wwwbrr.cr.usgs.gov/projects/GWC_coupled/phreeqc/.

