Modeling of Effective Edge Surface Area of Montmorillonite Particles

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In the disposal process of radioactive-waste in geological repositories, montmorillonite clay is examined to be used for the buffer material in the engineered barrier system (EBS). It is known that montmorillonite particles will be dissolved by highly alkaline groundwater and its dissolution rate is greatly dependent on the compaction condition. One of the reasons for this phenomenon is physical masking, i.e., the edge of a sheet-like shaped montmorillonite particle is covered with the other particles and consequently the reactive edge surface area decreases effectively. In order to understand the mechanism of physical masking, we performed Monte Carlo simulation of infinitely thin disks which model montmorillonite particles and investigated the equilibrium structure under various density conditions. We used two kinds of potentials for the interaction between particles. One is the quadrupole potential which can accurately describe anisotropic interactions of clay particles. However, the quadrupole potential is not available on high density condition due to the limitation of the model. Therefore we performed the Monte Carlo analysis using the rigid body potential under various density conditions simultaneously and examined the consistency between the results by these two potentials. Figure 1 shows the numerical results of equilibrium structure of disks obtained from different potentials. Fig.1(a) and (b) display the equilibrium structure at low density by the quadrupole potential and the structure from low to high density conditions by the rigid body potential, respectively. As shown in Fig.1(a), the particles make aggregates with “house of cards” structures which can be observed in natural clay particles at low density. On the other hand, the equilibrium structure of particles at high density by the rigid body potential simulation (Fig.1b) shows a layered structure, which is similar to montmorillonite stacking reported elsewhere. It is found that the equilibrium structures between two potentials show different features at low density. However, they indicate the similar structures with increasing the density because the possible states of particles are limited due to the excluded volume effect. We calculated effective Edge Surface Area (ESA), i.e., the edge area which is not masked by other particles. In order to calculate effective ESA based on equilibrium structure from Monte Carlo simulation, we used a geometric masking model. The masked area was determined whether the vertical distance from the edge of all particles to the surface of other particles is less than arbitrary values *d*. In this study, the masking parameter d/s (s : diameter of platelet) is set to 0.01, 0.02, 0.04 and 0.08. Figure 2 shows the visualization results of the masked area (red part) for the equilibrium structure of particles obtained from Monte Carlo analysis by the respective potentials. They show that masked area increases with the increase of packing density. From these results, we calculated the effective ESA which is the area per unit mass unmasked by other particles quantitatively. The calculation results show that the effective ESA decreases monotonically as the density increases and the results by both potentials are smoothly connected at moderate density condition. The calculated effective ESA explains well why the dissolution rate of montmorillonite is extremely reduced at highly compacted conditions. Therefore, it strongly suggests that the dissolution rate reduction is mainly caused by physical masking. As the result, we can propose a mathematical model describing the relationship between the dry density and the effective ESA of montmorillonite particles. This research includes a part of the results of “Advancement of Processing and Disposal Technique for the Geological disposal of TRU Waste (FY2017)” under an entrustment from the Agency for Natural Resources and Energy (ANRE) in the

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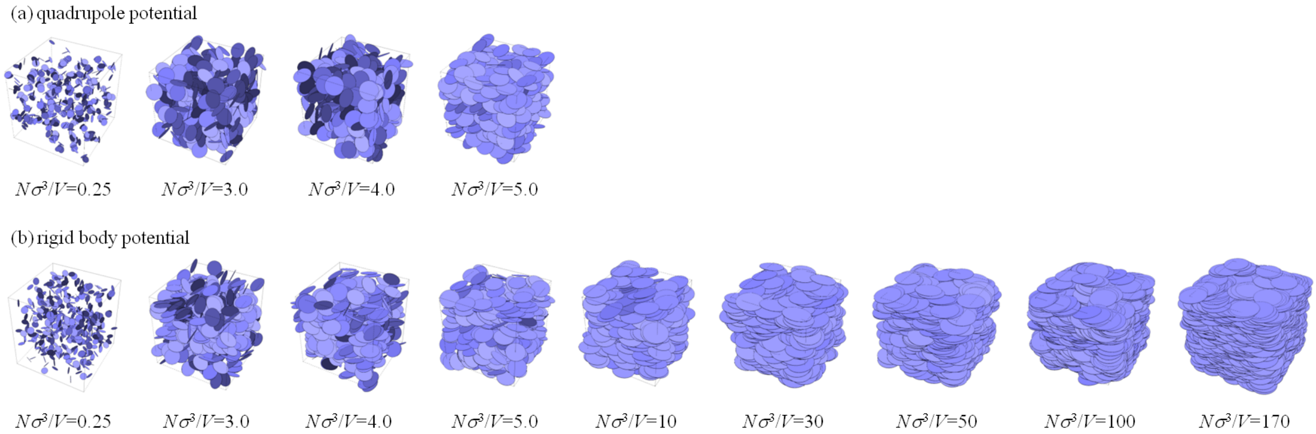

Fig. 1 Equilibrium structure of disks which model montmorillonite particles by Monte Carlo method using different potentials ($N\sigma^3/V$: nondimensional density)

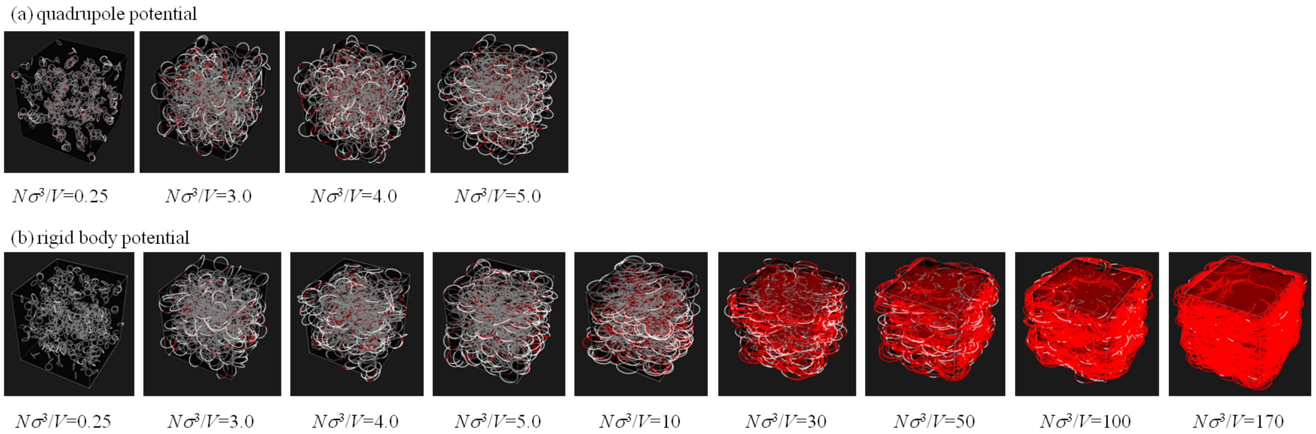

Fig. 2 Masked area (red part) and effective ESA calculated by masking model