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Super-hydrated kaolinite under high pressure: A machine learning molecular dynamics study

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Numerical simulation is a practical tool to investigate the physical and chemical properties of clay minerals. Molecular dynamics (MD) is a powerful method based on nanoscale dynamics. Primary MD methods are classical molecular dynamics (CMD) and *ab initio* molecular dynamics (AIMD). CMD is based on a highly simplified physical model of atoms, i.e., atoms are approximated as point particles of classical mechanics, and the interaction is formulated by simple functions. AIMD employs quantum mechanical calculations for electrons, although nuclei are approximated as point particles. The computational costs of the former and the latter are low and high, respectively. The calculation accuracies using CMD and AIMD are low and high, respectively. Therefore, CMD is mainly used for relatively large-scale simulations for a rough estimation of the physical and chemical properties of clay minerals, and AIMD is used for the simulations of small systems to evaluate these properties accurately. Recently, machine learning molecular dynamics (MLMD) was proposed. This new method realizes low computational cost and high accuracy due to artificial neural networks trained using a vast number of *ab initio* calculations results to reproduce the potential energy surface of *ab initio* calculations.

Kaolinite is one of the simplest phyllosilicates. Recently, the MLMD simulations of kaolinite atmospheric pressure have succeeded [1]. MLMD reproduced the experimental result of phonons of the hydroxy group in the low-frequency region, which CMD and AIMD could not reproduce. Recently, an interesting structural change with respect to pressure has been reported: kaolinite with a thin water layer between the layers, which is called “super-hydrated kaolinite” [2]. The thin water layer is an interesting research target of the mineral-fluid interface under extreme conditions. MLMD simulations of super-hydrated kaolinite were performed, and the lattice parameters of super-hydrated kaolinite observed by the experiment [2] were accurately reproduced by the simulations. The vibrational properties of the hydroxyl group, the diffusion of water molecules in the interlayers, and other static and dynamic properties will be given at the presentation.

References

[1] K. Kobayashi, A. Yamaguchi, M. Okumura, Appl. Clay Sci. **228**, 106596 (2022)

[2] H. Hwang *et al*., Nat. Geosci. **10**, 947 (2017).