1–18 Understanding the Cesium Adsorption Behavior in Concrete

— Accurate Simulation of Cement Hydrate Using Machine Learning Molecular Dynamics —

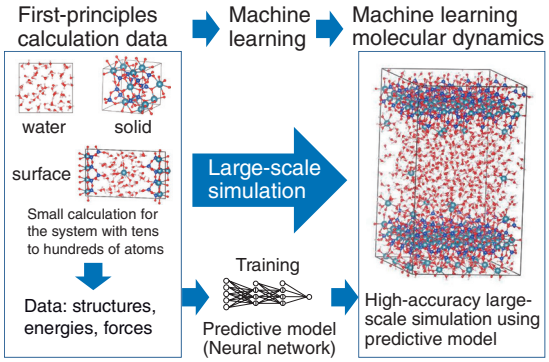

Fig.1-37 Schematic representation of machine learning molecular dynamics
By performing a large number of FP calculations for various structures (solid, water, surface, etc.) with small system size, the FP data set (about 70000 structures) is created, and a predictive model reproducing the energy and forces of the FP data is constructed using machine learning. Molecular dynamics based on this predictive model enables us to conduct a high accuracy large-scale simulation.

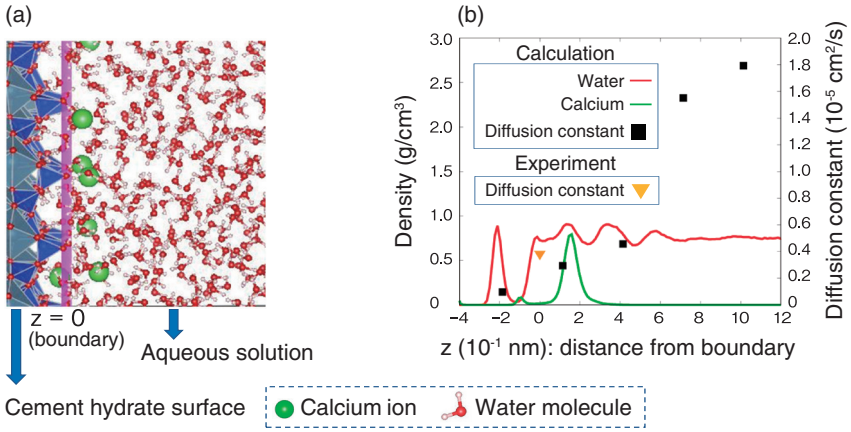

Fig.1-38 Distribution and transport properties of water and ions at the water-surface interface of cement hydrate
(a) Structure of CH surface and water interface obtained by MLMD. The boundary between the CH surface and aqueous solution is set at z=0. (b) Computed density distribution of water and calcium ions near the interface, and the diffusion constants of water. It can be found that the diffusion constants of water become significantly lower than that of bulk water, and the ions are localized near the CH surface.

At TEPCO's Fukushima Daiichi NPS, due to damage to the reactor pressure vessel, a huge amount of radioactive cesium has been adsorbed into the concrete of the reactor containment vessel. For the disposal and volume reduction of large amounts of contaminated concrete obtained by decommissioning the nuclear power plants, understanding the adsorption behavior of cesium in concrete becomes important.

It is known that cesium ions are strongly adsorbed into cement hydrate (CH) in concrete, a very porous substance. Cesium ions are presumed to diffuse with water in the porous CH and adsorb onto the surface of CH. Atomic simulation is an essential tool to obtain detailed information about the form and intensity of cesium adsorption on CH. However, large-scale simulation taking into account the chemical reaction and water transport in the pores of CH is required, which is difficult to achieve with high accuracy first principles (FP) calculations due to its high computational effort.

In this study, we used machine learning molecular dynamics (MLMD) to overcome the computational cost of FP calculation. In this method, using a large amount of FP calculation data for various CH structures with tens to hundreds of atoms, a predictive model (machine learning force field: MLFF) reproducing the results of the FP calculation is created using a machine learning technique (see Fig.1-37). Molecular dynamics simulation using MLFF is called MLMD, which enables us to conduct simulation with almost FP accuracy and much lower computational effort than an FP calculation.

As a first step toward elucidating the adsorption behavior of cesium on CH, we constructed the MLFF for the basic constituent elements of CH: silicon, calcium, hydrogen, and oxygen atoms. Using the constructed MLFF, we conducted a large-scale MLMD simulation for the CH-water interface, including thousands of atoms, to investigate the transport and adsorption properties of water and calcium ions. Our simulation successfully reproduced the experimental values of the diffusion constant of water near the CH surface as shown in Fig.1-38.

In the past, accurate simulations using FP calculations have been difficult for complex systems such as CH. We have shown that MLMD enables us to perform the simulation of CH with almost FP accuracy. Future work will aim to extend the MLFF of cesium to clarify the adsorption behavior of radioactive cesium in concrete.

(Keita Kobayashi)

Reference

Kobayashi, K. et al., Machine Learning Potentials for Tobermorite Minerals, Computational Materials Science, vol.188, 2021, 110173, 14p.