Structure and dynamics of water at feldspar surfaces from machine learning augmented molecular simulation

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Abstract:

Over the last decade, significant evidence has been gathered that points to feldspar minerals as the most important ice nucleating particles in our planet's atmosphere [1]. There have been efforts to understand the formation of ice at feldspar surfaces using experimental tools such as optical microscopy [2], electron microscopy [3], and atomic force microscopy (AFM) [4], as well as computational tools such as density-functional theory (DFT) calculations [5], and molecular dynamics (MD) simulations [6, 7]. In spite of these efforts, the microscopic characteristics of the nucleation site and the atomic-scale mechanism of ice nucleation at feldspar surfaces have not yet been fully elucidated.

Here, we develop a machine-learning potential (MLP) to model the interactions at the water/microcline feldspar interface, using a dataset generated from density-functional theory (DFT) calculations based on the SCAN exchange-correlation functional [8]. The MLP accurately reproduces the energies and forces obtained from DFT calculations. We then performed MD simulations, driven by the MLP, to investigate the structural and dynamic properties of water on various fully hydroxylated terminations of the (100), (010), and (001) surfaces of microcline feldspar. With these simulations, we explore the effects of ions and surface structure on ice nucleation. Our results provide important insights into the effects of solid-liquid interface on the structure and dynamics of liquid water. These insights contribute to unravelling the microscopic mechanism of ice nucleation on feldspar.

References

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Figures

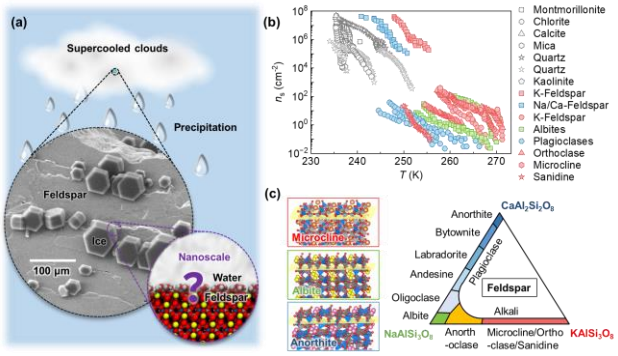

Figure 1. Background of ice nucleation on feldspar surfaces: Feldspar exhibits the highest efficiency for ice formation, yet the underlying mechanism remains unknown.

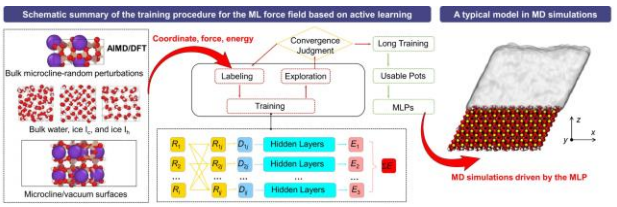

Figure 2. Schematic summary of the training procedure for the machine-learning potential based on active learning.