Effect of water confinement on heterogeneous ice nucleation

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Understanding the formation of ice is of great importance to many fields of science. Sufficiently pure water droplets can remain in the supercooled liquid phase to nearly -40 ºC. Crystallization of ice in the atmosphere therefore typically occurs in the presence of ice nucleating particles (INPs), such as mineral dust or organic particles. These can trigger heterogeneous ice nucleation at clearly higher temperatures. Therefore, a better understanding of how the various types of aerosol particles present in the atmosphere affect ice nucleation (IN) in clouds would be an important advance in the field of atmospheric science.

Experiments have shown in great detail what is the IN activity of different types of compounds, and recently also clarified the importance of small surface features such as surface defects, which function as active sites for ice nucleation. On most mineral dust particles, there may be only a few active sites for ice nucleation, typically around defects or pits (Holden et al., 2019). Simulations also showed enhanced ice nucleation efficiency in confined geometry such as wedges or pits (Bi, Cao and Li, 2017).

We systematically study the effect of water confining defects with different surface geometries; pyramidal pits, wedge-shaped cracks and slits with water confined between two parallel walls, using molecular dynamics simulations with both all-atom and monatomic water models, and show that that these defects enhance ice nucleation both at large supercooling and at very low supercooling.

Results of simulations on pyramidal pits on Si (100) surfaces, realizable experimentally, show a clear (ΔT > 10 ºC) enhancement of ice nucleation compared to the very weakly IN active flat Si (100) or Si (111) surfaces. To show that water confinement can enhance IN also at very low supercooling, at temperatures above -10 ºC, we constructed wedge shaped structures with β-Agl (0001) surface as one of the two side walls, and slit systems by positioning two β-Agl (0001) slabs to mirror each other to cancel the dipole field from the polar surfaces. Depending on the wedge angle or the relation of the width of the gap between two slabs in the slit systems with the thickness of ice bilayers, ice nucleation can be clearly enhanced or hindered. We also clarify the different mechanisms behind IN enhancement at different geometries.

Understanding the enhanced activity at surface features may enable characterization of ice nucleation active sites on some atmospheric particles, creation of IN active sites at otherwise poorly active materials such as silicon, and also enable enhancing very active IN materials such as
AgI, to nucleate ice at nearly zero supercooling.

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