

MODELLING THE ROLE OF SURFACE MICROSTRUCTURE IN HETEROGENEOUS NUCLEATION OF ICE

O. H. Pakarinen, T. Ponkkonen and H. Vehkamäki

Department of Physics, P.O.B. 64, FIN-00014 University of Helsinki, Finland
email: olli.pakarinen@helsinki.fi

An accurate description of clouds in climate models requires solid knowledge about their properties, but formation of ice clouds, in particular, is not well understood. Ice crystals may form in the atmosphere either by homogeneous or heterogeneous ice nucleation. The latter process, initiated by an aerosol seed particle at relatively high temperatures, is not understood at the molecular level. We aim for revealing the molecular-level mechanisms of early stages of ice nucleation and understanding the experimentally observed differences between materials and nucleation modes.

We have developed an atomistic model of ice crystal formation using a continuum of methods from density functional theory (DFT) to large-scale classical molecular dynamics (MD). We are looking at ice nucleation in the presence of different surfaces that represent aerosol particles with varying lattice match with ice, surface corrugation and interactions, both for ideal test systems and for feldspars, AgI and black carbon that are active as heterogeneous ice nucleation seeds.

Simulations with the mW potential [1] show that surface geometry can largely explain the high nucleation activity of AgI, whereas the (001) surface of K-feldspar, considered an important ice nucleation agent in the atmosphere [2], shows very weak activity in mW simulations, without considering the real surface chemistry of the material, which we study with DFT.

We also study the importance of the effect of feldspar microstructure – the interface between alternating orthoclase and albite layers and defects due to lattice mismatch, as well as other crystal planes such as (100) found important in recent experiments [3]. In addition to studying nucleation dynamics with MD, we are looking at accurate energetics with DFT. Comparison of water layer adsorption on SiO₂ versus K-feldspar is helping us understand the difference in nucleation activity between these materials.

Contact nucleation mode simulations show nucleation activity at higher temperatures than immersion, in agreement with experiments, due to rotational freedom of water at the open surface.

Results of the work will help in understanding the experimentally observed differences in nucleation activity between different materials and nucleation modes.

[1] V. Molinero and E. B. Moore, [J. Phys. Chem. B 113 \(2009\) 4008](#).

[2] J. D. Atkinson et al., [Nature 498 \(2013\) 355](#).

[3] A. Kiselev, F. Bachmann, P. Pedevilla, S. J. Cox, A. Michaelides, D. Gerthsen and T. Leisner, [Science 355 \(2017\) 367](#).