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Molecular dynamics simulations of homogeneous CO2 nucleation

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The condensation of carbon dioxide (CO2) is a topic of general interest in view of global decarbonization targets, e.g. in low-temperature CO2 capture technologies promoting the phase transition of CO2 gas is the crucial step. Homogeneous nucleation of a mixture of CO2 and argon gas in a supersonic nozzle has been studied at temperatures from 78 to 92 K, and CO2 partial pressures between 70 and 800 Pa. The consistency between the current data and measurements at higher temperature suggests the critical clusters remain liquid-like even at these low temperatures.

Here we present large-scale atomistic molecular dynamics (MD) simulations of homogenous CO2 nucleation from the vapour phase at temperatures from 75 to 105 K. The MD approach is an unbiased method to study the nucleation process, including the phase and structure of even the smallest clusters. We used argon carrier gas as a heat bath for the CO2 molecules to avoid unphysical removal of latent heat.

Simulations confirm that despite strong undercooling, nucleation proceeds through liquid-like clusters. Also, by applying standard steady-state cluster growth kinetics, we are able to calculate the cluster formation free energies from the MD simulations. The results suggest a curvature correction to the classical liquid drop model used in the classical nucleation theory. The correction depends only on the bulk liquid properties, and hence the simulation-based correction can be applied to predict the nucleation rates of real CO2.

The simulation-based theory is able to capture the magnitude and the temperature-dependency of the nucleation rate rather well, whereas both standard CNT and its self-consistent version (SCNT) underestimate the rate by several orders of magnitude. Here we have corrected the theoretical values with the non-isothermal factor, which is about 0.01-0.1 for the studied system.