

A) Objective

The objective of this paper was to study the transition of (nano)-droplet evaporation from high to low Knudsen numbers and also subcritical to supercritical conditions. A Lennard-Jones Argon system is used to simulate the droplet and vapor system via classical Molecular Dynamics simulation. The simulations were run in NVE ensembles, with a special pressure control technique. The Knudsen number (in this study) is the ratio of the vapor mean free path λ to the droplet diameter D . The (ambient) pressure was varied from 0.1-1.5 times the critical pressure for Ar, and the temperature was varied from 2.3 to 6.1 times the critical temperature.

B) Important Results

The authors find that their results agree well with kinetic theory, which predicts that the droplet diameter will decrease linearly with time. This is no surprise, since they are modeling the system in this regime. For moderate pressures, the authors find good agreement with the D^2 law for a temperature of 500K. This is the only temperature where the author's find this agreement, where they attribute the disagreement with hydrodynamic theory with low Knudsen (non-continuum) conditions (which seems reasonable). For high ambient pressure, the authors observe a transition to supercritical droplet evaporation. While this ruins their procedure for estimating the Knudsen number (due to lack of surface tension defining a spherical droplet radius), these conditions also warrant a continuum analysis, since the Knudsen number should be much larger in the supercritical fluid. Here the authors observe that the droplet never enters into a steady state, where the droplet surface temperature would be constant (pseudowet-bulb temperature).

C) Concerns

The 2 phases used in the simulation were obtained by 2 separate single phase simulations, where a spherical sample was included from both samples, a spherical liquid droplet and a box containing vapor and a spherical exclusion. Why was this method used? Presumably because it would require a very long simulation and a much larger number of atoms to start with a single large droplet, begin evaporation to form the vapor, and then collect data?

To maintain a fixed ambient pressure, particles were randomly inserted or deleted when the ambient density was too low or high, respectively. The authors note that their method does not affect the system's dynamics. Was it not possible to control the ambient region via a proper barostat control? Or was it not possible to couple the ambient region to some kind of reservoir? Computational constraints might have limited these options.

Aside from these thoughts, it seems that the authors took great care to address these issues with tests of their own, so it seems to be a well performed study which I would recommend for publication.