Critical Radius of Supercooled Water Droplets: On the Transition toward Dendritic Freezing

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The freezing of freely suspended supercooled water droplets with a diameter of bigger than a few micrometers splits into two rather different freezing stages, because the freezing enthalpy cannot completely be stored in the droplet in the first freezing run and has to be released to the environment during an about 1000 times longer time span. In the present work the distribution of the ice portion in the droplet directly after the dendritic freezing phase as well as the evolution of the ice and temperature distribution has been investigated in dependence of the most relevant parameters as droplet diameter, dendritic freezing velocity (which correlates with the supercooling) and supercooling temperature. On the experimental side acoustically levitated droplets in climate chambers have been investigated in combination with high-speed cameras to study the correlation between supercooling temperature and freezing speed. The obtained results have been used for finite element method (FEM) simulations of the dendritic freezing phase under consideration of the beginning second, much slower heat-transfer dominated freezing phase. A theoretical model covering 30 layers and 5 shells of the droplet has been developed which allows us to describe the evolution of both freezing phases at the same time. The simulated results are in good agreement with experimental as well as with calculated results exploiting the heat balance equation. The most striking result of this work is the critical radius of the droplet which describes the transition of one-stage freezing of the supercooled water droplet toward the thermodynamically forced dendritical two-stage freezing in which the droplet cannot sufficiently get rid of the formation heat anymore. Depending on the parameters named above this critical radius was found to be in the range of 0.1 to 10 micrometers by FEM simulations.

Further, in our presentation we adopt the hypercooling temperature as a concept which is common in the materials sciences to water ice research. The hypercooling temperature is defined by the highest temperature at which the freezing enthalpy can completely be stored within the freezing system. This means that the liquid droplet can completely freeze in one run without heat release to the environment. We emphasize that the heat capacity of the supercooled water strongly depends on the temperature, and also the freezing enthalpy is temperature dependent. This leads to a considerably higher hypercooling temperature compared to the value found in the literature.