Homogeneous versus heterogeneous nucleation in aluminum droplet at low supercooling: nucleation rates and the total number of nuclei

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Crystallization processes determine the quality of the product obtained and understanding the molecular mechanisms that occur during crystallization remains a scientific challenge. As a model system, we have chosen aluminum (Al), whose crystallization temperature $T_C \approx 642\ ^oC$ (corresponding to an undercooling $\Delta T_C \approx 18.3\ ^oC$) and can be well detected by differential scanning calorimetry (DSC) under non-isothermal conditions [1]. On the contrary, the crystallization event was repeatedly detected by the isothermal DSC after several tens of minutes at the undercooling $\Delta T \approx 6.8\ ^oC$. The standard kinetic model of homogeneous and heterogeneous nucleation was solved numerically to determine the size distribution of the nuclei, the nucleation rate, and the total number of nuclei. In our previous analysis of Al crystallization via homogeneous nucleation, we have determined [1] the most probable values of the interfacial energy and the kinetic barrier of nucleation. Heterogeneous nucleation of Al on the concave droplet surface is modeled for the wetting angle $\vartheta = 90\,^\circ$C when the nucleation barrier of heterogeneous nucleation $W^{het} \approx 1/2$ of the homogeneous nucleation barrier W^{hom} and thus the probability of heterogeneous nucleation would be much higher than homogeneous one. On the other hand, the number of nucleation centers, where cluster formation starts, is much higher for homogeneous nucleation. As a result, the two nucleation processes are comparable in the Al sample of the mass 9.91 mg. Homogeneous nucleation starts at shorter times, but heterogeneous nucleation prevails in longer times.

[1] Z. Kožíšek, R. Král, P. Zemenová: *Homogeneous nucleation and crystallization model of Aluminum droplet based on isothermal DSC analysis*, J. Therm. Anal. Calorim. 147 (2022) 13089–13098. https://dx.doi.org/10.1007/s10973-022-11497-z