Crystallization of aluminum droplet

Zdeněk Kožíšek, Robert Král, and Petra Zemenová

Institute of Physics of the Czech Academy of Sciences, Cukrovarnická 10, 162 00 Praha 6, Czech Republic

Aluminum (Al) melted droplet of mass $m = 9.9$ mg was undercooled with the cooling rates 2-20 K/min and the crystallization temperature $T_c \approx 642\ ^oC$ ($\Delta T \approx 18\ K$) was detected by the Differential Scanning Calorimetry (DSC) apparatus under non-isothermal conditions. Experimental data were analysed by the Johnson-Mehl-Avrami (JMA) model under assumption that nucleation and growth occur at T_c. It seems to be a reasonable presumption as the growth rate of Al is high (tens of meters per second). JMA model enables us to determine the kinetic Avrami coefficient $n = d + 1 \leq 2$ from experimental data. However, $n \leq 2$ shows that the JMA model fails as the dimensionality of growth $d < 1$ is unrealistic.

Similarly, we analysed the DSC data recorded at isothermal conditions with crystallization occurring at $\Delta T \approx 6\ K$ below the melting temperature T_m. Al liquid droplet was cooled down repeatedly to the temperature $T > T_c$ and kept at the isotherm sufficiently long time. Even at this low supercooling, the crystallization of droplet occured after a time delay τ. The time delay since the beginning of the isotherm T up to crystallization event changed occasionally: $\tau \approx 10 - 50$ minutes. Experimental data confirmed stochastic nature of crystal nucleation similarly to other systems [1].

We have numerically solved the kinetic equation of the crystal nucleation within standard model [2] to determine the time dependent number of nuclei as a function of their size. Small clusters have a tendency to diminish and only supercritical clusters (nuclei) grow independently. At crystallization, it is necessary to form a sufficent number of nuclei to initiate growth. The number of Al atoms in the liquid droplet decreases as the crystal phase is formed and this effect was taken into account. The stochastic data of the time delay τ are often analysed with stochastic method, when Poisson distribution of nuclei is considered. However, as a fit parameter the stationary nucleation rate is used and that is why this analysis fails due to non-stationary process. Standard nucleation model enables us to determine the interfacial energy limit, when the first nuclei are formed within the system. Experimental value of time delay of crystallization event is high in respect to high growth rate. Crystallization probably occurs by two step mechanism [3] when the structure of small sub-critical clusters differs from larger ones and thus kinetics of small clusters is slow.

This work was partially supported by Operational Programme Research, Development and Education financed by European Structural and Investment Funds and the Czech Ministry of Education, Youth and Sports (Project No. SOLID21 CZ.02.1.01/0.0/0.0/16_019/0000760).

[1] S. A. Kulkarni, Ss. S. Kadam, H. Meekes, A. I. Stankiewicz, Joop H. ter Horst: Cryst. Growth Des. 13 (2013) 2435.

[2] Z. Kožíšek: J. Cryst. Growth 522 (2019) 53.

[3] P. G. Vekilov: Nat. Mater. 11 (2012) 838.