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CRYSTALLIZATION OF RAPIDLY QUENCHED METAL DROPLETS WITH HIGH MELTING POINT

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Abstract: The crystallization process of rapidly quenched liquid metal droplets has been studied by using photographic techniques. Cooling rates of these droplets have been obtained and they have shown apreciable supercooling before a sharply phase transition occurs which is contrary to classical nucleation theory. The instability of the liquid phase is investigated within the framework of an adiabatic nucleation model.

The mechanism by which a crystalline solid forms in the freezing of a pure liquid is controlled essentially by the nucleation rate of small crystallites in the supercooled liquid phase. However, the nucleus itself cannot be studied directly and this limits experimental information to the measuring of the limit temperature T_L to which the liquid can be supercooled before crystallizing.

Small droplets has been used in the study of the solidification mechanism to minimize the probability of nucleation due to the presence of foreign nuclei inside the liquid. By using this technique, this limit temperature T_L has been measured for a variety of liquid metals .

We have applied the technique of dendritic crystal growth to obtain dendriticc crystals from rapidly quenched metal droplets with high melting point. This technique improved with an stereophotographic apparatus has permited to obtain the cooling rate of these droplets and, then, to determine the amount of supercooling these metals can sustain.

This procedure was described previously.(1,2) A metal wire is overloaded by a high current inside a chamber and the sample is liquefied. Liquid metal droplets are produced and these radiating spheres are then fall down in the chamber, being cooled during the time of flight.

Calibrated curves of color density ratio on photographic film vs temperature and position of free falling droplet vs time enable us to plot the temperature of droplet at given position vs time as is represented in Figure 1. It should be noted that the limit temperature T_L to the supercooling the liquid droplet undergoes is just the minimum temperature before the sudden increase of temperature at which the solid phase appears.

This curve also has the following characteristics: (i) the temperature at which the solid appears is not a plateau-like but immediately falls; (ii) the cooling rate after this limit temperature corresponds to a radiative cooling droplet; and (iii) the temperature attainable by the solid phase is lower than that predicted upon release of the latent heat during the nucleation process.

As is suggested by Nelson(3) the characteristic (iii) merely indicates that only a fraction of the mass of the supercooled droplet could crystallize rapidly, the remainder of the solidification being rate-limited by the process of heat loss from the outer surface of the droplet. However, this picture estimates that only after a time of the order of 30 msec or larger the Ta droplet of Figure 1 should present a radiative cooling behavior.

The present results sugest that nucleation phenomenon may not be regarded as an isothermal one as considered by the classical nucleation theory. It seems the solid can nucleate at higher temperature than the liquid phase but this temperature may be lower than the associated melting tempeature, the resulting evolution towards equilibrium ocurring very fast, as predicted from the adiabatic theory of nucleation.(4)

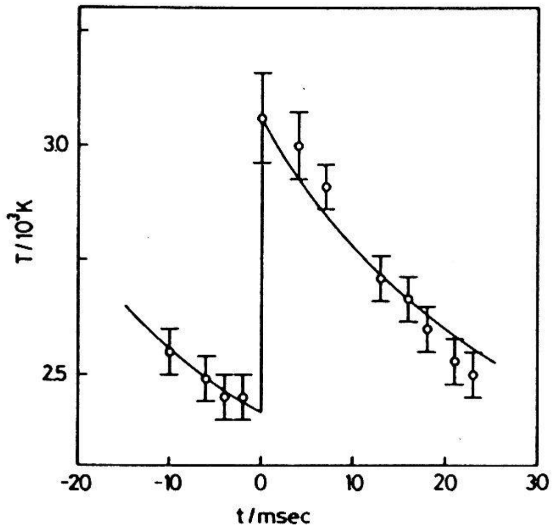

Fig.1 Typical cooling rate to 400μm Ta droplets

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