Investigation of the Relationship between Undercooling and Solidification Velocity

Summary of Research
and
Final Report

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Task Objective

This work was aimed at reconciling the differences between experimental measurements of the theoretical predictions of the solidification velocity as a function of undercooling. The theory proposed by Boettinger, Coriell and Trivedi (the BCT theory) has been one of the most widely used models for describing the nature of the solidification of undercooled metals and alloys. However, for undercoolings greater than about 5% of the absolute melting temperature, there is considerable discrepancy between theory and experiment. At these large undercoolings, experimental results exhibit a much lessened dependency of solidification velocity on undercooling than is predicted by theory. Furthermore, unpredicted plateaus in the solidification velocity as a function of undercooling are observed.

Task Description

Containerless processing is inherently necessary to obtain accurate measures of the solidification velocity as a function of undercooling. The absence of a container promotes high degrees of undercooling in a melt and allows the application of direct imaging techniques to obtain accurate measures of solidification velocity as a function of undercooling. For this work, electromagnetic levitation and electrostatic levitation were chosen as the appropriate techniques. Electromagnetic levitation techniques were already extensively used on a variety of alloys. However, electromagnetic levitation is highly perturbing, producing liquid flows of the order of tens of centimeters per second. As a result, it was not entirely clear that experimental results are unaffected. Since electrostatic levitation is a more benign technique, experiments were duplicated and compared to electromagnetic levitation. A basis for low earth orbit experimentation, the most benign experiments, was evaluated upon a comparison of results. At every step, experimental results were examined within the theoretical framework.

Task Significance

The understanding of solidification kinetics is fundamental to the synthesis of materials. The understanding of the speed of solidification, along with nucleation frequencies, are missing links for modelers who are focused on the development of innovations in solidification processing. Basically, knowing the manner in which to control and predict the speed of solidification is the key to knowing how to influence the bulk microstructure and, as a result, the properties and performance of the material through solidification processing.

Summary of Research

Electrostatic Levitation Experiments

A comparison is being conducted between electromagnetic levitation (EML) and electrostatic levitation (ESL) experiments for the following nickel-based alloys: Ni-4at.%Sn, Ni-5at.%Ti and Ni-5at.%Cu. Since these two experimental techniques cause different streamline flows in the sample[1], this comparison is desired in order to determine what effect turbulence has on solidification kinetics. Experiments on Ni-4at.%Sn were conducted at NASA MSFC's Electrostatic Levitation Facility to cover the entire range of achievable undercoolings.

The results from the ESL experiments were identical to the results from EML experiments. From these experiments we can definitively state that the fluid streamlines from EML have no effect on the measurement of solidification velocities above 2 meters per second.

Effect of Residual Gases on Solidification

The solidification of pure nickel in both EML and in containers has been studied very extensively over the past 40+ years, and deviations from current rapid solidification theory[2] are usually observed at high undercoolings. It has been hypothesized that oxygen or other residual gas contaminant may play a role in these deviations[3-5]. Grain refinement mechanisms[6] may also play a role since the deviation from rapid solidification theory occurs at roughly the same undercooling as the onset of grain refinement[7].

Previous experiments at Vanderbilt showed that the type of cooling gas used in EML influenced the solidification velocity behavior[4]. However, residual gas concentrations were not measured after processing. During FY03, extensive experimentation on pure nickel in EML was conducted using various cooling gases (UHP He, He-15vol%H_2 and He-15vol%CO) to study the effect of absorbed gases on solidification velocity. Processed samples were examined for residual gas concentrations (oxygen, nitrogen and hydrogen) by LECO® combustion analysis. Initial results indicate that hydrogen is absorbed in quantities around the eutectic concentration of 360 ppm H and oxygen is maintained below 100 ppm O when a helium-hydrogen mixture is used as the cooling gas. While hydrogen appears to be an effective oxygen reducing agent in EML, its absorption into the liquid nickel sample affects the kinetics of solidification. Nitrogen is not absorbed in any detectible quantity by liquid nickel. Carbon monoxide also acts as an effective oxygen reducing agent but is known to adsorb on nickel surfaces without dissociating. Solidification velocity results were significantly reduced in the presence of CO. Solidification velocity measurements under a UHP He environment showed significant data scatter, but measured oxygen quantities in processed samples show no strong correlation to solidification velocity. Accurate measurement of oxygen in processed samples may be problematic since oxygen adsorption may occur even after processing. Extensive post processing analysis has shown that very little helium is absorbed nickel melts. From experiments, modeling, and analysis we have concluded that hydrogen is responsible for the scatter in solidification velocity results on nickel at high undercooling.

Modeling of rapid solidification processes

Extensive modeling was completed on experimental results in accordance with the current rapid solidification theory. The theory is known to disagree with experimental results for certain alloy systems, namely those with a sufficiently high equilibrium partition coefficient (k_E). The reason for the disagreement stems from the theory's inability to correctly predict experimentally observed "plateaus" in the solidification velocity data at intermediate undercoolings. The theory does not predict a plateau for alloys with a value of k_E greater than about 0.1, but a plateau is always experimentally observed regardless of the value of k_E[8].

These solidification velocity plateaus are accompanied by a transition from dendritic to cellular solidification morphology and a rapid increase in solute trapping. Analysis is being conducted on an alternate selection criterion proposed by Sekerka known as the optimum stability conjecture (OSC)[9]. There is some experimental evidence[10] that dendritic growth at low undercoolings is governed by a variable stability parameter that decreases with increasing thermal Peclet number. OSC, in conjunction with the Ivantsov solution of the current theory, was found to agree with experimental data on nickel-based alloys in EML.

Our experimentation on nickel-based alloys in EML at low undercoolings show that for high k_E alloys, the OSC is required to obtain good fits of experiment with theory.

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