Dynamics of Complex Two-phase Microstructure Selection in Peritectic Systems

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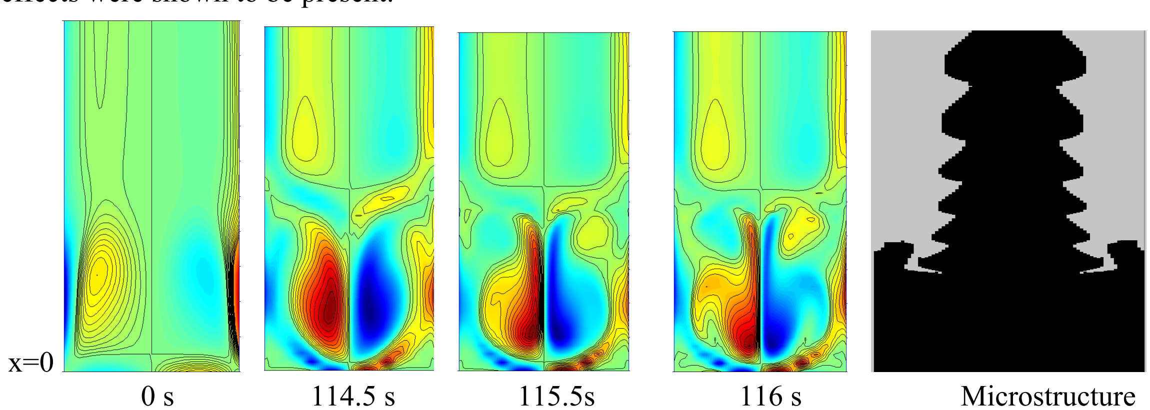
Abstract:

A major part of our understanding of microstructure evolution is based on steady-state growth of single-phase materials. More relevant microstructures in energy related materials, however, consist of two or more phases in which the microstructure evolves under highly non-steady state growth conditions. In addition, several competing phenomena must be considered simultaneously to establish the criteria for different mechanisms of microstructural selection. This complex problem was examined quantitatively through detailed analytical, numerical and experimental investigation of selection processes, as governed by nucleation undercooling, nucleation rates of the two phases, time-dependent complex fluid flow, and the time-dependent competitive growth of the two phases. Depending on the relative contributions of the above phenomena, significantly different microstructures were observed and predicted. Critical experiments were developed in which all the above factors influencing the microstructure formation were isolated to quantitatively assess the role of each phenomenon in the selection of a specific microstructure.

Recent Results:

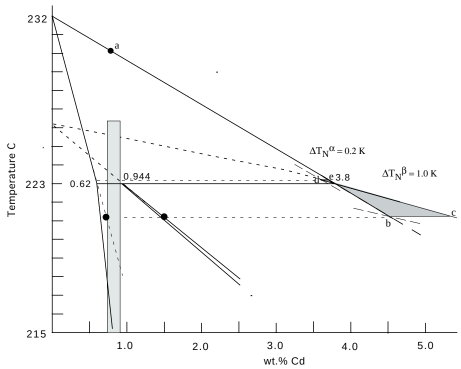
Detailed numerical modeling of diffusive and convective growth was carried out to determine the effect of fluid flow on microstructure selection in directional solidification. Several interesting flow transitions were predicted as a function of the Rayleigh Number, and these flow characteristics were coupled with the interface motion to predict their effects on the solidification microstructures. It was shown that in a system, in which an oscillatory or a chaotic fluid flow is present, as shown in the figure below, more complex patterns can evolve which are neither predicted by the diffusive models nor can they be directly related to the fluid flow behavior. When an oscillatory flow is present, its interaction with the solidification front was shown to give rise to an oscillatory structure in which a large oscillatory treelike structure of the primary phase is surrounded by the peritectic phase. Such a microstructure was experimentally observed by us previously in a sample diameter of 6 mm in which strong convection effects were shown to be present.



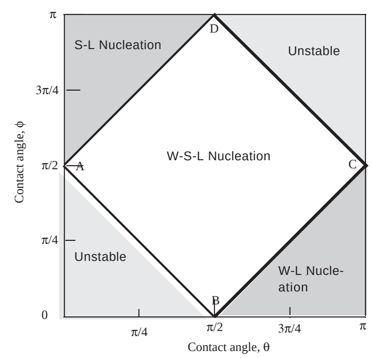
Spatial dynamics of the flow and its interaction with the solidification front to generate an oscillatory microstructure.

A novel experimental technique was developed to study microstructure evolution under diffusive growth conditions in directional solidification by employing specimen ampoules of very small diameter (0.2-1.0 mm). Using this technique, several new observations were made. Within a small hypo-peritectic composition range, discrete repeated bands of the primary and peritectic phases were observed, as previously predicted by the theoretical model developed at Ames Laboratory. By measuring the composition profile, the banding cycle was quantitatively established and the nucleation undercoolings for the primary and peritectic phases were determined. The nuclei were observed to form at the junction of the interface and the wall, so that the nucleation undercooling could be related to the contact angles with the interface and with the ampoule wall. A predictive model was developed for nucleation at the interface, wall, and the interface-wall junction. As shown in the figure below, a nucleation map was generated in the two contact angle (θ,ϕ) space, revealing the preferred nucleation mechanism with respect to the relative values of the two contact angles.

Banded structure



Banding cycle and nucleation undercooling



Nucleation map

Significance:

The strong effect of fluid flow on microstructure development in peritectic systems was quantitatively established for the first time, implying the ability to develop novel microstructures through proper design of fluid flow patterns. It was shown for the first time that accurate values of the nucleation undercoolings of the primary and peritectic phases could be obtained in peritectic systems through precise directional solidification experiments in which the nucleation site is known. A nucleation map for different heterogeneous sites was developed that would allow one to obtain a variety of microstructures by controlling the interface energy at the ampoule wall through the selection of different ampoule materials. The dynamics of competing growth of the two phases was shown to be critical in the evolution of final microstructures. A coupling of analytical and numerical models with experimental studies was shown to give quantitative insight into different physics that govern the evolution of complex two-phase microstructures.

Future Work:

The evolution of complex microstructures under different nucleation conditions will be examined. The process of nucleation is critical in giving rise to continuous or particulate bands. The selection of two-phase growth morphology will be quantitatively studied, with particular attention given to the situation where one of the phases becomes morphologically unstable, forming cells or dendrites enabling the study of nucleation in the intercellular region.

Interactions:

Significant interactions exist with Alain karma (Northeastern University) and W. Kurz (Ecole Polytechnique, Lausanne). The selection of microstructure in peritectic systems is also critical to the research in the magnetic materials group within Ames Laboratory.