



Diffusional constrained crystal nucleation during peritectic phase transitions

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Abstract

The casting behaviour, microstructure formation and resulting mechanical properties of materials that undergo a peritectic phase transition during solidification are largely determined by the kinetics of this high-temperature phase transformation during the production process. However, current nucleation models do not accurately predict the nucleation behaviour and phase transition kinetics in many polycrystalline materials. Utilizing a newly developed experimental technique, we have performed in situ observations to study the nucleation behaviour of a newly forming intermediate phase by using the peritectic phase transition in Fe–C and Fe–Ni alloys as examples. In our experiments as well as by using thermodynamic and kinetic arguments we demonstrate that nucleation of a new intermediate (peritectic) phase can be constrained in the presence of solute diffusion fields that form during primary solidification due to an increase in the Gibbs free energy barrier for nucleation. We found a strong correlation between the magnitude of these diffusion fields and the resulting nucleation undercooling required for the formation of a new phase. Our study casts new light on, and clarifies for the first time, the much-debated underpinning reason for the occurrence of massive phase transformations occurring during solidification processing at large nucleation undercoolings. These new insights contribute to the improvement of nucleation theory and allow more accurate predictions on nucleation events and, in turn, physical properties of materials that undergo phase transitions in the course of materials production processes.

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1. Introduction

The formation of crystalline solids from solution is fundamental to many natural and industrial processes. The crystallization process begins with nucleation, which plays a central role in determining the structure and size distribution of the crystals [1]. These crystallization processes are important in a variety of fields such as medical engineering, nanoparticle production, protein crystallization, and the solidification of metals and alloys. Hence, it is critically important to understand the fundamental scientific principles underpinning this initial, often limiting, step

in the phase transformation process. Only with such understanding can we exercise control over and optimize solidification structures of significance to our industries that work towards the enhancement of the quality of human life.

In the classical picture, nucleation is a stochastic process in which compositional fluctuations induce the formation of clusters that are unstable with respect to dissolution. Clusters increase in both size and free energy due to the creation of an interface until a threshold is crossed, whereupon the free energy reduction due to forming the new phase overcomes the penalty for creating an interface, and growth proceeds spontaneously. However, recent observations have clearly suggested that this classical theory of nucleation often fails to predict nucleation

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behaviour during phase transitions [2,3], such as the peritectic phase transition. Materials that undergo the peritectic phase transition include technologically important materials such as steel, copper and aluminium alloys, nickel-based superalloys and titanium aluminides as well as magnetic and electronic materials for the superconductor industry [4–6]. For instance, the production of large single-crystal (RE)–Ba–Cu–O superconductors is based on the formation of the RE-123 phase through a peritectic reaction [7] at high undercoolings in order to improve the kinetics of the reaction [8]. The pivotal role nucleation can play in peritectic systems can be illustrated with reference to the Al–Ti system. The primary intermetallic Al₃Ti phase that forms in the course of the peritectic transition can act as a nucleant for aluminium grains [9–12] and these nucleation events are fruitfully utilized to achieve grain refinement during casting or welding.

The peritectic phase transition occurring in the Fe–C system at 1768 K is arguably the most important peritectic from an industrial point of view since much of the more than 1.5 billion tonnes of steel produced annually undergoes this phase transition. Moreover, this peritectic reaction and subsequent peritectic transformations have been implicated in a myriad of difficulties encountered in the production process that hamper production rates and result in inferior product quality [13]. In the Fe–C system, the liquid and primary solid δ -ferrite phases produce a second γ -austenite solid phase ($\delta + L \rightarrow \gamma$) with a different crystal structure than δ . Under equilibrium conditions, nucleation of γ occurs at δ grain boundaries that are in contact with liquid at a temperature just below the equilibrium peritectic temperature, T_P^E . Following the nucleation event, the γ -phase grows along the liquid/ δ interface (termed the peritectic reaction) and subsequently further into the δ and into the remaining liquid (the peritectic transformations). However, at high cooling rates the δ to γ phase transition is suppressed and a transformation occurs by a massive-type of phase transformation [14–16]. The mechanism by which this massive-type of transition occurs has never been satisfactorily explained notwithstanding the fact that it is this phase transition that seems to be the root cause of the quality defects encountered in the continuous casting of steel [16]. Jacot et al. [17] recently showed that solute-trapping-free massive transformation can occur above the limit of absolute stability, i.e. at high velocities of the transformation front without the need of high nucleation undercoolings. However, linked to the occurrence of the massive-type of transformation in peritectic systems is the observation that significant undercooling is required to induce this massive-type of transformation, which in turn suggests a mechanism linked to a nucleation constraint. The occurrence of high nucleation undercooling in peritectic systems and the resulting massive transformation of δ to γ has frequently been reported [14–15,18–20], but a satisfactory explanation for these nucleation constraints is still lacking. The application of classical nucleation theory (CNT) [21] to

explain the high nucleation undercooling in peritectic systems has had limited success since the experimentally determined temperature at which γ nucleation occurs (undercooling below the equilibrium peritectic temperature) is much lower than can be explained by this theory [18,19]. Fundamental understanding of the nucleation process in peritectic systems is still in its infancy and the present study is aimed at providing new insights with respect to constrained nucleation events.

2. Experimental

Progress in the study of nucleation behaviour in metallic systems during peritectic solidification has been inhibited by the difficulties encountered in conducting experiments at the elevated temperatures at which they occur. However, the development of techniques for the in situ study of high-temperature phase transformations, such as transmission X-ray observation, the Bridgman furnace and high-temperature laser-scanning confocal microscopy provides the ability to capture solidification processes in real time and high resolution as well as to observe and measure the morphology and kinetics of phase transformations. We employed a concentric solidification technique [22–25] within a high-temperature laser-scanning confocal microscope to study in situ the nucleation behaviour and transformation kinetics of the peritectic phase transition in Fe–C and Fe–Ni alloys as examples of the nucleation behaviour of a newly forming intermediate phase. Specimens (9.8 mm diameter and 250 μ m thick) are placed beneath a quartz viewpoint in a gold-plated, ellipsoidal infrared heating furnace under an ultrahigh-purity argon atmosphere. A 1.5 kW halogen lamp located at one focal point of the ellipsoidal cavity heats the specimen positioned at the other focal point. Capitalizing on a radial thermal gradient across the specimen, a

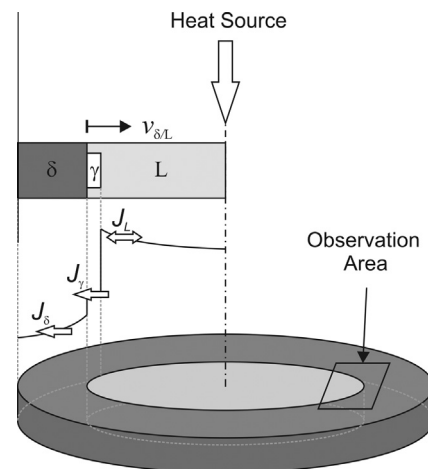


Fig. 1. Schematic illustration of the concentric solidification technique for HTLSCM. The sketch on the left hand side shows the concentration distribution and solute diffusion fluxes J_i in the vicinity of a γ cluster at the δ/L interface.

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