

Remelting-induced anomalous eutectic formation during solidification of deeply undercooled eutectic alloy melts



X.X. Wei^a, X. Lin^b, W. Xu^c, Q.S. Huang^a, M. Ferry^c, J.F. Li^{a,*}, Y.H. Zhou^a

^aState Key Laboratory of Metal Matrix Composites, School of Materials Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, PR China

^bState Key Laboratory of Solidification Processing, School of Materials Science and Engineering, Northwestern Polytechnical University, Xi'an 710072, PR China

^cAustralian Research Council Centre of Excellence for Design in Light Metals, and School of Materials Science and Engineering, The University of New South Wales, Sydney, NSW 2052, Australia

ARTICLE INFO

Article history:

Received 13 April 2015

Accepted 11 May 2015

Keywords:

Eutectic solidification

Undercooling

Anomalous eutectics

Remelting

Crystal orientation

ABSTRACT

Anomalous eutectics are argued to form due to the remelting of the primary solid during solidification of deeply undercooled eutectic alloy melts. As an indicator of the tendency for anomalous eutectic formation, the remelted fraction of the primary eutectic was analyzed systematically based on the eutectic dendrite growth theory. For eutectic alloys with either larger equilibrium solute distribution coefficients or gentle liquidus slopes, the primary eutectic is highly supersaturated with solute and more prone to remelting. When the eutectic composition is set to different values, (e.g. the eutectic point is closer to one phase of the eutectic), the two eutectic phases under rapid growth change their compositions simultaneously, but their remelted fractions during temperature recalescence do not vary significantly. Three representative binary eutectic alloys Ag–39.9 at.%Cu, Ni–19.6 at.%P and Pd–16.0 at.%P – their eutectic products are solid solution–solid solution, solid solution–stoichiometric intermetallic compound and stoichiometric intermetallic compound–stoichiometric intermetallic compound, respectively, were solidified at large undercooling. Anomalous eutectics were observed in the first two eutectic alloys whose eutectic structure consists of at least one solid solution phase, whereby the stoichiometric intermetallic compound phase remained highly oriented whereas the solid solution phase had a near random distribution of orientations. For the Pd–16.0 at.%P alloy, however, the primary eutectic retained its original morphology as the final solidification structure regardless of the degree of undercooling. All these experimental results validate the argument that anomalous eutectics result from the remelting of the primary solid.

© 2015 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.

1. Introduction

Eutectic structures composed of at least two phases can exhibit a wide variety of morphologies as a function of the alloy composition and the solidification condition. An issue that has been discussed intensively over the past few decades is the transition from lamellar or fibrous eutectics at low undercooling to anomalous eutectics at large undercooling [1–8]. Several ideals have been proposed to explain the anomalous eutectic formation. Based on the finding that particulates of Cu in the Ag–Cu anomalous eutectic were distributed discontinuously in the Ag matrix, Powell and Hogan [9] argued that repeated nucleation of Cu took place in the undercooled eutectic alloy melt, resulting in the formation of

an anomalous eutectic. Later, Kattamis and Flemings [1] solidified a Ni–Sn eutectic alloy at large undercooling and carried out serial sectioning by consecutively polishing and etching the sample to generate a three dimensional distribution of the phases. They found that each eutectic phase was spatially interconnected, which led them to propose that an α -Ni supersaturated solid solution of eutectic composition is first formed during rapid solidification, but subsequently decomposes to the equilibrium α -Ni and β -Ni₃Sn phases. Considering that the two eutectic phases might be significantly different from each other in terms of their growth kinetics, Jones [2], Wei [4], Li [6] and their various coworkers pointed out that one of the phases might grow much faster than the other in a substantially undercooled eutectic melt, due to which the coupled eutectic interface no longer remains stable and an anomalous eutectic is formed. Distinct from those researchers, whereby anomalous eutectic formation was related to the failure of coupled eutectic growth, Goetzinger et al. [10] assumed that

* Corresponding author. Tel./fax: +86 21 54748530.

E-mail address: jfli@sjtu.edu.cn (J.F. Li).

the very thin eutectic lamellae formed in rapid solidification of Ni–Si, Co–Sb and Ni–Al–Ti alloys were unstable due to their high interfacial energy and would fragment into an anomalous eutectic structure.

Li et al. [11,12] also investigated the solidification of an undercooled Ni–Sn eutectic alloy and found that coupled eutectic growth gave way to decoupled growth only when the undercooling exceeded 130 K, despite the fact that anomalous eutectics started to form at an undercooling as low as 25 K. These workers concluded that both coupled and decoupled growth of the two eutectic phases could result in anomalous eutectics. Recently, Yang et al. [13] and Clopet et al. [14] carefully measured the crystal growth velocity in Ni–Sn and Ag–Cu eutectic alloy melts as a function of undercooling and examined the crystal orientation of each phase in the anomalous eutectics, respectively. Their investigations led to the same conclusion as Li and co-workers.

Along with the rapid solidification of undercooled alloy melts, significant temperature recalescence is known to occur. The primary solid supersaturated with solute may be partially remelted due to superheating, thereby disintegrating into an anomalous eutectic structure [11–14]. Without the help of the remelted liquid, it cannot be expected that the primary solid will breakup and lose its initial morphology, as solid solution diffusion is very slow [10]. Depending on the types of alloying elements, eutectic alloys can form a range of constituent phases that differ from each other in terms of their solidification and remelting behaviors and, therefore, should exhibit different tendencies to form anomalous eutectics. However, very little systematic work has been carried out to determine what influences anomalous eutectic formation. In the present work, the extent of remelting of the primary solid and its dependence on the characteristic parameters of eutectic phase diagrams were first analyzed theoretically on the assumption that coupled eutectic growth takes place during the rapid solidification of undercooled eutectic alloy melts. To test the theory, three representative eutectic alloys with phases composed of (i) solid solution–solid solution, (ii) solid solution–stoichiometric intermetallic compound, and (iii) stoichiometric intermetallic compound–stoichiometric intermetallic compound, were solidified at different degrees of undercooling and their structures analyzed by optical microscopy and electron backscatter diffraction (EBSD).

2. Theoretical description

2.1. Characterization of degree of remelting

Below the equilibrium liquidus temperature, T_L , an alloy melt crystallizes to form a solid, which is accompanied by a decrease in free energy. Generally, crystallization commences by heterogeneous nucleation at a temperature very near to T_L due to the abundant nucleation sites in the melt. As those most effective nucleation sites are removed from the melt, there is a considerable increase in undercooling prior to nucleation. In this case, driven by the accumulated large free energy difference between the liquid and crystallizing phases, the nuclei that form will grow rapidly in the deeply undercooled melt. To dissipate the latent heat of crystallization into the liquid effectively, the solidification interface breaks into branches [15–17]. Fig. 1 illustrates the temperature distribution along a growing eutectic branch and the undercooling constitution at its tip. Obviously, the temperature in the solid is the lowest at the tip. The solid formed at this position is therefore the richest in solute. As the branch grows forward (equivalent to going back from the tip), the temperature of the solid behind the tip is elevated because latent heat is continually released while the branch is thickening, and the solid crystallizing onto the side of the branch changes its composition toward the equilibrium value at the eutectic temperature. This means that the primary solid that

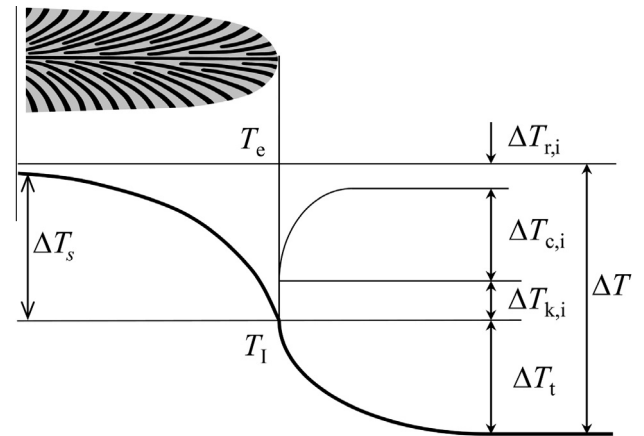


Fig. 1. Schematic illustration of the temperature distribution along a growing eutectic branch and the undercooling constitution at the tip, where T_e is the eutectic temperature, T_l tip temperature, ΔT melt undercooling, ΔT_s temperature rise in the solid, and $\Delta T_{c,i}$, $\Delta T_{r,i}$, $\Delta T_{k,i}$ and ΔT_t the solute undercooling, curvature undercooling, kinetic undercooling and thermal undercooling, respectively, at the branch tip. Note that the solute undercooling, curvature undercooling and kinetic undercooling are generally different from eutectic phase α to β .

comprises the central part of the branch becomes superheated and will be partially remelted since temperature recalescence is completed within a very short time, and it is impossible for the primary solid to adjust its composition immediately by solid-state solute diffusion. One can understand the reason for this behavior from the experimental work of Dutta and Rettenmayr [18]. Putting an Al solid in contact with an Al–Mg liquid, they found that remelting would take place so long as the solid and liquid compositions at the interface were out of equilibrium, and an increase in the liquid solute supersaturation would lead to an increased driving force for remelting.

Below the equilibrium eutectic temperature, both eutectic and single phase structures are permitted to form from a thermodynamic viewpoint. What will form, according to the competitive principle, depends on their growth velocities, whereby the phase with a higher growth velocity will form preferentially. Generally, if the melt undercooling is not too large, coupled eutectic growth first occurs during rapid solidification. At large undercooling, one of the eutectic phases may grow singly as the primary solid [11]. Regardless of whether the primary solid consists of a eutectic or single phase, it may all be partially remelted owing to temperature recalescence and, correspondingly, result in the formation of an anomalous eutectic.

Depending on the interface anisotropy strength and solidification conditions, solidification interfaces in undercooled alloy melts may be of dendritic or seaweed morphology [19,20]. For metallic systems, dendritic growth is often observed, while seaweed growth only occurs under special conditions [21]. Due to space limitations, this paper mainly considers the anomalous eutectic formation when eutectic dendrites grow primarily in an undercooled alloy melt.

To evaluate the remelted fraction of the primary eutectic, we first need to calculate the compositions of the two eutectic phases at the dendrite tip. The model developed by Li and Zhou [22], unlike the previous work where only solute diffusion and curvature effects are considered, also takes into account the effects of thermal diffusion and interface kinetics, and therefore, is suitable for calculating the eutectic growth in undercooled melts. Following the treatment of Trivedi et al. [23], this model provides mathematical solutions for two types of phase diagram: (i) the metastable phase diagram is cigar-shaped such that, below the eutectic temperature, the solidus and liquidus are parallel, and

Download English Version:

<https://daneshyari.com/en/article/7879701>

Download Persian Version:

<https://daneshyari.com/article/7879701>

[Daneshyari.com](https://daneshyari.com)