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Evolution kinetics of microgravity facilitated spherical macrosegregation within immiscible alloys

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ABSTRACT

The evolution kinetics of microgravity facilitated spherical macrosegregation within model Fe-Cu immiscible alloys has been systematically investigated by drop tube experiments and 3D phase-field simulations. In microgravity environment, liquid phase separation of Fe-Cu alloys induced the appearance of spherical macrosegregation patterns with various core-shell structures. The formation probability and evolution characteristics of these phase-separated core-shell morphologies depended on the volume fraction of surface active Cu-rich liquid together with the cooling rate. As the cooling rate decreased, the duration time of phase separation extended and the formed dispersed structures showed a tendency to form as core-shell structures influenced by the effects of Marangoni convection and surface segregation. Meanwhile, core grew larger while the shell became thinner. The occurrence probability of spherical macrosegregation firstly increased and then decreased with the rise in the copper concentration, and core-shell morphologies changed from three-layer to two-layer which was accompanied by core shrinkage and shell thickening.

1. Introduction

The properties of immiscible alloys are strongly dependent on their macrosegregation characteristics [1–6]. By modulating the liquid phase separation processes in these immiscible alloys, various macrosegregation patterns have been obtained [7–14]. During conventional solidification, Stokes sedimentation dominates the liquid phase separation process and facilitates the formation of a layered macrosegregation structure [10–14]. As the solidification process of immiscible alloys is performed in microgravity, the gravity effects on the phase separation are considerably weakened [3,5,7], and the Marangoni convection [10,15,16] becomes the key factor to control the movement of the two coexisting liquids. This induces the appearance of two typical macrosegregation patterns, namely a core-shell structure and a dispersed structure [3,17–23].

To date, the formation mechanisms of these macrosegregation patterns have been sufficiently discussed based on both experiments and simulations [3,5,7,10–14,17–19,23]. During the phase separation, the liquid phase with a larger density sinks while the other liquid phase floats upward under the effect of Stokes motion [3,5,7,10,23]. Owing to the surface segregation, the liquid phase with a lower surface energy tends to move toward the sample surface and forms a surface segregation layer [5]. In contrast, Marangoni convection contributes to the migration of minor phase globules from the low-temperature regions to the relatively higher-temperature regions [3]. We believe that the cooling rate, the volume fraction variation and the surface free energy with respect to the two-coexisting immiscible liquids greatly influence the liquid phase separation process and even alter the final macrosegregation configuration, especially within the microgravity environment.

The attention to this research is not very significant due to the limited opportunity for, and high cost of, a space experiment. Because of that, at present the evolution kinetics of microgravity facilitated spherical macrosegregation for immiscible alloys is not completely understood within the broad ranges of both alloy compositions and cooling rates. After selecting three model alloys with compositions $Fe_{65}Cu_{35}$, $Fe_{50}Cu_{50}$ and $Fe_{35}Cu_{65}$ as shown in Fig. 1(a) [24], we set the purpose of this work as to study in a three-dimensional regime the phase separation kinetics and

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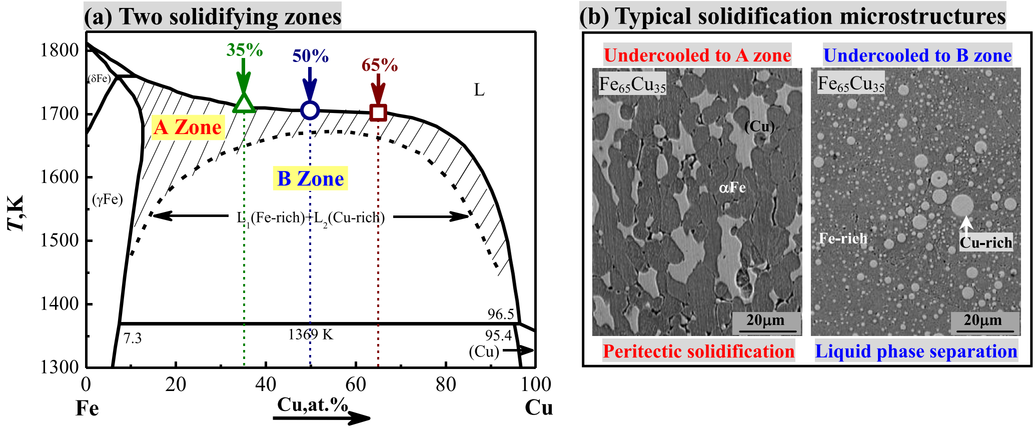

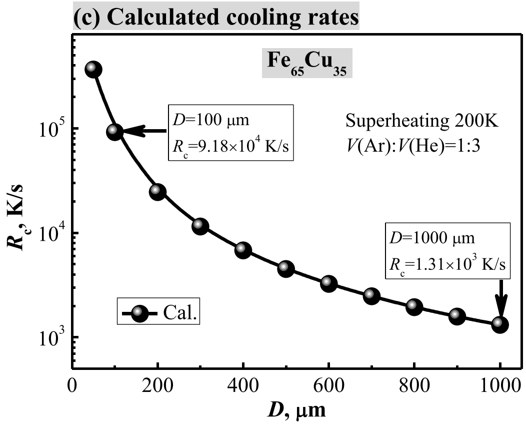

Fig. 1. Solidification mechanisms of Fe-Cu alloy droplets inside a drop tube: (a) two zones of solidification mechanism selection, (b) typical solidification microstructures, (c) calculated cooling rate R_c versus droplet diameter D.

microstructure evolution mechanisms in the reduced-gravity experiments inside a drop tube [25] and through the phase-field simulations [3].

2. Methodology

2.1. Drop tube experiment

Drop tube experiments of liquid $Fe_{65}Cu_{35}$, $Fe_{50}Cu_{50}$ and $Fe_{35}Cu_{65}$ alloys were conducted to clarify the effects of alloy compositions and cooling rates on the liquid phase separation kinetics under microgravity conditions. These master alloys were prepared from high purity elements of iron (99.999%) and copper (99.99%) in a high vacuum arc-melting furnace. Each sample with a mass of approximately 3 g was placed inside a quartz tube with a 16 mm diameter, a 150 mm length and a small orifice at the bottom. After loading, the quartz tube was installed on the top of a high vacuum chamber in the drop tube facility. Before the experiments, the vacuum chamber was evacuated to 3.0×10^{-5} Pa and backfilled with He cooling gas. Subsequently, induction heating was used to melt the alloy sample, and the molten alloy was dispersed into many alloy droplets from the orifice of the quartz tube by the jetting gas. As a consequence, these alloy droplets fell and rapidly solidified in the containerless and reduced-gravity environment. The solidified alloy droplets were mounted, polished and etched by a solution composed of 5 g $FeCl_3$ + 5 mL HCl + 20 mL H_2O. The microstructures were analysed with a Zeiss Axiovert 200 MAT optical microscope and a FEI Sirion electron microscope. The phase constitution was investigated by an Oxford INCA Energy 300 energy-dispersive spectrometer.

2.2. Phase-field simulation

The metastable phase separation and microstructure evolution for liquid Fe-Cu immiscible alloys were explored theoretically using a three-dimensional phase-field model. This model considers the phase separation influenced by the effects of surface segregation and Marangoni convection [3,16]. In this model, the free energy function of the Fe-Cu alloy droplet was simulated by

$$F = F_b + F_g + F_s \tag{1}$$

where F is the total Gibbs free energy, and F_b, F_g and F_s represent the bulk free energy, the free energy of the concentration gradient and the surface free energy [3], respectively. Furthermore, the value of this free energy function varies with the spatial coordinates **r**, the evolutionary time t, and the mole concentration C. The dimensionless concentration-field governing equation can be expressed as

$$\frac{\partial C}{\partial \tau} = \nabla \cdot \left(C(1-C) \nabla \frac{\delta F}{\delta C} \right) - \nabla \cdot (\nu C) + \nabla \cdot \xi \tag{2}$$

where C is the mole concentration of Cu, τ is the evolution time, v is the local velocity and ξ is the random Gaussian white noise. The definitions of physical symbols can be found elsewhere [3]. The