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Effect of electric current pulses on solidification of immiscible alloys

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ABSTRACT

The effect of electric current pulses (ECPs) on the solidification of immiscible alloys is investigated. The results demonstrate that ECPs mainly affect the microstructure formation through changing the nucleation behaviors of the precipitated phase droplets (PPDs). When the PPDs have a higher electrical conductivity compared to the matrix, ECPs enhance the nucleation rate and promote the formation of a well-dispersed microstructure. Otherwise, ECPs cause a decrease in the nucleation rate and promote the formation of a phase segregated microstructure.

1. Introduction

In nature, many alloys show a phase diagram characterized by the appearance of a miscibility gap in the liquid state. These alloys have great potential applications in industry [1,2]. For instance, Cu–Cr is the most important contactor materials [1], Cu–Pb or Al–Bi alloys are potential materials for advanced bearings in automotive applications [2]. Unfortunately, when an immiscible alloy is cooled into the miscibility gap, liquid–liquid decomposition occurs and serious phase segregation takes place [3].

Microstructures are at the center of materials science and engineering. Microstructure control is essential for any materials, especially metals and alloys [4]. Electric current pulses (ECPs) have great potentials in controlling the solidification microstructure of alloys [5]. Many research studies have been carried out to investigate the effect of ECPs on the solidification of pure metals [6], eutectic alloys [7] and solid solution alloys [8] in the past decades, They demonstrate that the application of ECPs during the solidification of metal and alloys can cause a refinement of the microstructure and a significant improvement in the mechanical properties of products. But up to date, little work was done on how ECPs affect the solidification of immiscible alloys [9]. The microstructure evolution under the effect of the ECPs remains an unsolved scientific problem. The objective of this letter is to investigate the effect of ECPs on the microstructure formation of immiscible alloys.

2. Experimental

Continuous solidification experiments were carried out with Bi–10%Cu–10%Sn (wt%) and Cu–25%Bi–25%Sn (wt%) alloys. The experimental procedures are as follows: The alloy was first melted and heated up to 1173 K in an alumina crucible. The melt was held at temperature of 1173 K for 40 min to form a single phase liquid. ECPs were then applied to the melt (ECPs pass through the melt from the top to the bottom of the sample, the frequency and the duration of each ECP are 50 Hz and 6 μs, respectively) and the crucible was withdrawn into a bath of liquid Ga–In–Sn alloy. The sample has a cylindrical shape of the size of 120 mm (length) × 6 mm (diameter). Microstructural investigations were carried out by using a scanning electron microscope (SEM) in a back-scattered imaging mode. The sizes of the dispersed particles were determined by the quantitative metallographic analysis.

3. Results and discussion

Fig. 1 shows the microstructures of the Bi–10%Cu–10%Sn samples solidified at the rate of 8mm/s with or without ECPs treatment. EDS analyses revealed that the dark phase is (Cu,Sn)-rich phase and the white phase is the Bi-rich matrix phase. The liquid phase separation took place and the solidification microstructure with serious phase segregation formed for the alloys solidified without ECPs. The alloy solidified under the effect of ECPs shows a microstructure consists of the dispersed particles of (Cu,Sn) phase and the Bi-rich matrix. These results indicate that ECPs promote the Bi–10%Cu–10%Sn alloy to form a well-dispersed microstructure.

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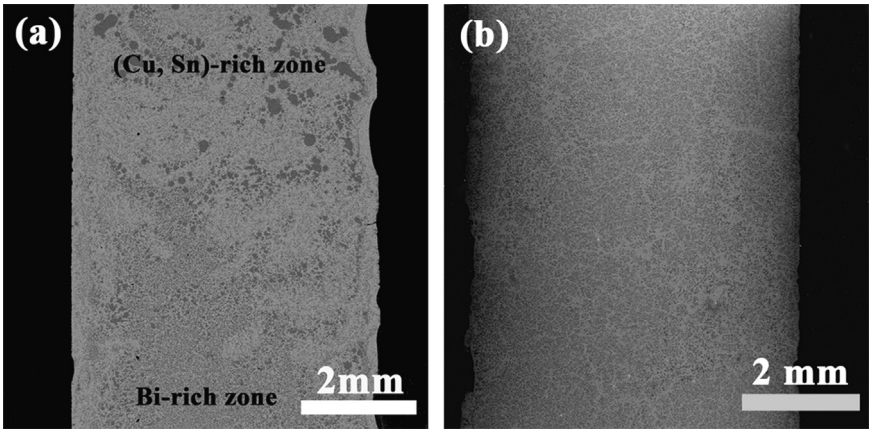

Fig. 1. Microstructures of Bi–10%Cu–10%Sn alloy solidified continuously at the rate of 8 mm/s: (a) without ECPs, (b) with ECPs, the peak value of the pulse current density is 20,000 A/cm^2.

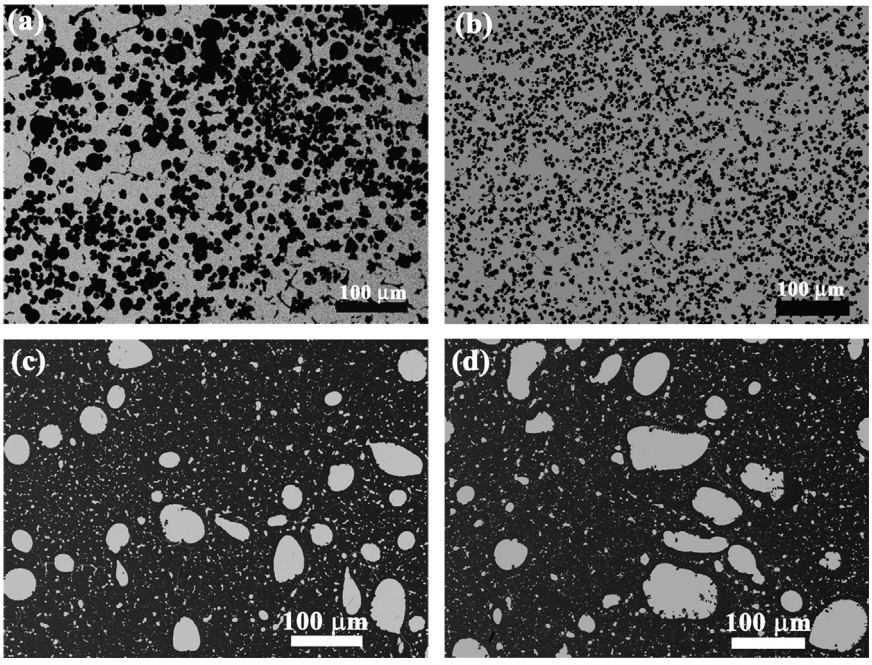

Fig. 2. Microstructures of Bi–10%Cu–10%Sn alloy (a, b) and Cu–25%Bi–25%Sn alloy (c, d) solidified continuously at the rate of 10 mm/s; (a) and (c) without ECPs; (b) and (d) with ECPs, the peak values of the pulse current density are both 30,000 A/cm^2.

Fig. 2 shows the microstructures of the Bi–10%Cu–10%Sn alloy samples and the Cu–25%Bi–25%Sn alloy samples solidified at the rate of 10 mm/s with or without ECPs treatment. Many dispersed particles are located at the grain boundaries and triple junctions, indicating that the liquid phase partially wet the solid phase in the final stage of solidification [10–12]. Fig. 3 shows the average diameter and the volume fraction of the dispersed particles. The experimental results demonstrate that, for Bi–10%Cu–10%Sn alloy, ECPs cause a decrease in the average size of the dispersed particles (see Fig. 3a) and promote the formation of a well dispersed microstructure (see Fig. 2a and b). While for Cu–25%Bi–25%Sn, ECPs cause an increase in the average size of the dispersed particles (see Fig. 3a) and are against the formation of a dispersed microstructure (see Fig. 2c and d).

ECPs may affect the microstructure formations through changing the temperature field, the spatial motions of the PPDs and the nucleation behaviors of PPDs. A model is proposed to clarify the microstructure formation process as well as the effect mechanisms of ECPs. Let fdR be the number of the PPDs per unit volume in a radius range between R and $R+dR$. Taking into account the concurrent action of the nucleation, growth and spatial motions of the PPDs, this function satisfies Eq. (1) during a liquid–liquid phase transformation [3]:

$$\frac{\partial f}{\partial t}+\nabla[(\boldsymbol{V}_0+\boldsymbol{V}+\boldsymbol{u_M}+\boldsymbol{u_S}+\boldsymbol{u_E})f]+\frac{\partial}{\partial R}(vf)=\frac{\partial I}{\partial R}|_{R=R^*} \tag{1}$$

where $\boldsymbol{V}_0$ is the moving velocity of the sample, $\boldsymbol{V}$ is the convective flow velocity of the melt, $\boldsymbol{u_S}$ and $\boldsymbol{u_M}$ are the Stokes and Marangoni velocities of the PPDs, $\boldsymbol{u_E}$ is the moving velocity of the PPDs due to an electric current [3]. R^* is the critical nucleus radius. ν is the growth rate of the PPDs. I is the nucleation rate of the PPDs and it can be calculated by using the classical nucleation theory [13]. The free energy change due to the nucleation of the PPDs under the effect of an electric current is given by [14]

$$\Delta G=\Delta G_0+\mu\frac{\sigma_M-\sigma_D}{\sigma_D+2\sigma_M}\left[\frac{3}{2}\ln\left(\frac{R_S}{R^*}\right)-\frac{65}{48}-\frac{5}{48}\frac{\sigma_M-\sigma_D}{\sigma_D+2\sigma_M}\right]R_S^2J^2\Delta V \tag{2}$$

where ΔG_0 is the change of free energy in a current-free system. σ_M and σ_D are the conductivities of the matrix and droplets, respectively. μ is the permeability of the melt which approximates