INFLUENCE OF SURFACE TENSION EFFECTS ON SOLIDIFICATION OF ALLOYS IN SPACE AND ON GROUND

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ABSTRACT

Solidification experiments of Al-Al$_3$Ni and Al-Bi alloys were carried out in space on board a Chinese recoverable satellite. An obvious double vortical radiant structure of Al-Al$_3$Ni eutectic and a homogeneous microstructure of Al-Bi monotectic were obtained. Combined fluid physics and metallography, the effect of surface tension gradient driven convection on the formation of radiant eutectic structure and the Marangoni migration of second-phase droplets in the molten alloy were analyzed.
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INTRODUCTION

Based on results from experiments in space, it was found that the convection driven by a surface tension gradient is one of the most important effects on producing materials in space, for example, a thermocapillary oscillatory convection may result in striations or segregation bands in crystal growth of semiconductors (Walter, 1987).. In order to explore the effect of thermocapillary convection on solidification of eutectic alloys and Marangoni migration of Bi droplets in Al-Bi monotectic alloys, Al-RE, Al-Al$_3$Ni eutectic alloys and Al-Bi alloys were carried out successfully in space on board Chinese recoverable satellites in 1990 and 1996 by the present authors (Zhang, 1993 and 1997). They discovered that the thermocapillary convection may be one of the important factors for forming the radiant eutectic structures under Microgravity conditions and they also analysed the Marangoni migration of Bi droplets in Al melt in space. Ratke et al (Ratke, 1996) considered that the microstructure of monotectic alloys results from the interaction among nucleation, growth, Stokes motion, Marangoni migration and the flow field, in which the Marangoni migration of the melted drop is important. The investigation of the effect of surface tension on solidification is the overlapping problem of materials science and fluid dynamics, which promotes the development of solidification theory and is useful for directing and improving the materials producing technique on the ground.

EXPERIMENT PROCEDURE

Al-10%Bi immiscible alloys with homogenous structure were manufactured by control casting technique. A sample of Al-Bi + Al was prepared by diffusion welding. And Al-Al$_3$Ni eutectic alloys were prepared in a vacuum unidirectional furnace on the ground Lab. Solidification experiments with a temperature gradient furnace under microgravity conditions in space were performed on board a Chinese recoverable satellite in which the microgravity level is superior to 10^{-5} g$_0$. The size of the samples were 8mm in diameter, 8mm in length for Al-Bi and Al, 12mm in length for Al-Al$_3$Ni. The samples were placed into graphite crucibles and

sealed in quartz tube in vacuum of 10^{-3}Pa. Figure 1(a) and (b) show the temperature-time curve in space and the position of thermocouple on ampoule.
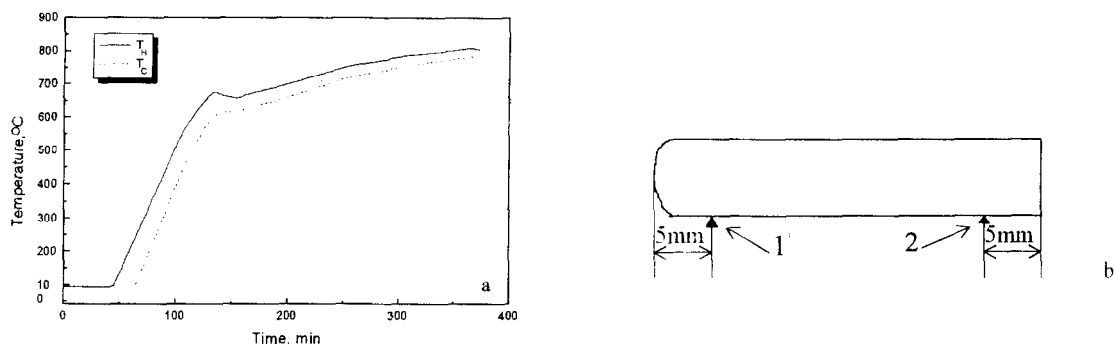

Fig.1 Temperature-time curve in space (a) and position of thermocouple on ampoule (b) in gradient furnace in Chinese recoverable satellite.

RESULTS AND DISCUSSION

Analysis of Radiant Eutectic Structure Obtained in Space

A radiant structure of Al-RE eutectic alloy was found in a 1990 space experiment (Zhang, 1993). In this experiment, only the bottom of the sample partly wetted and adhered to the graphite crucible, which means the other part of the space melt would not contact with the crucible. The difference thermal transport at the bottom and the other surface will yield a temperature gradient which might cause surface tension gradient driven convection (Marangoni or thermocapillary convection) in the Al-RE melt. This convection may induce the radiant structure. In order to ensure the analysis, re-melt and solidification of Al-Al₃Ni eutectic alloys were carried out in a space furnace with average temperature gradient of 5 °C/cm in the 1996 recoverable satellite. No any radiant structure was found in one of the Al-Al₃Ni eutectic sample(1[#]) with serious oxide surface, even the shape of the sample looks like a bullet with a bulboid head caused by surface tension effect, as shown in Figure 2(a) and (b). In another sample of Al-Al₃Ni alloy(2[#]) with metallic luster surface, a more obvious radiant structure with double vortices of Al-Al₃Ni eutectic was obtained as shown in Figure 3(a). For analyzing this phenomenon, the flow structure driven by surface tension in the melt was performed by using numerical calculation to solve basic equations of stream and vorticity functions with the thermophysical parameters of the alloys and the experiment data in space. The model adopted in the
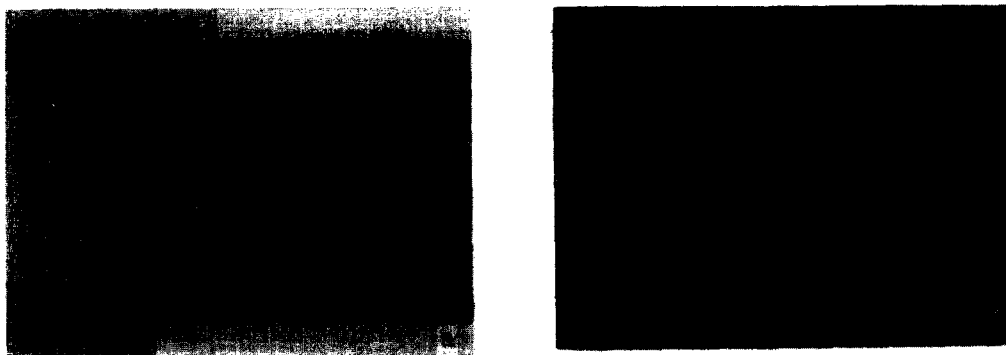

Fig.2 Macrograph (a) and microstructure of Al-Al₃Ni(1[#]) in longitudinal section (b) on board Chinese recoverable satellite.
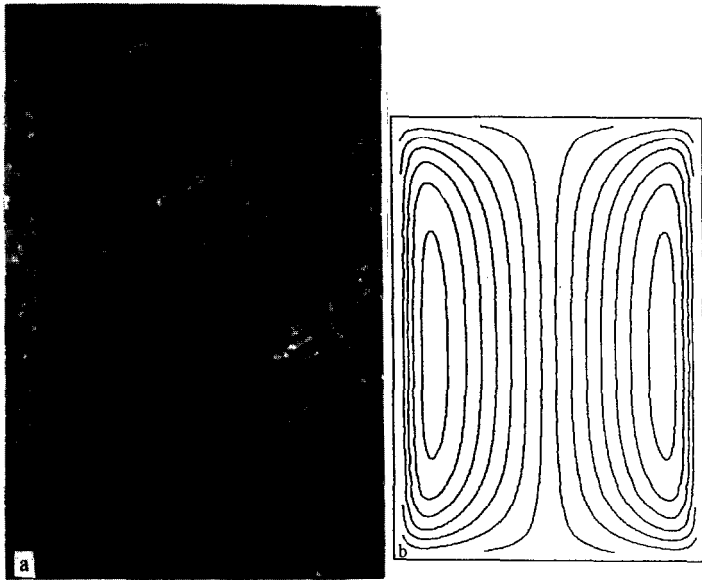
Fig. 3 Radiant eutectic structure with double vortices of
Al-Al$_3$Ni alloy(2[#]) obtained in space(a) and simulated
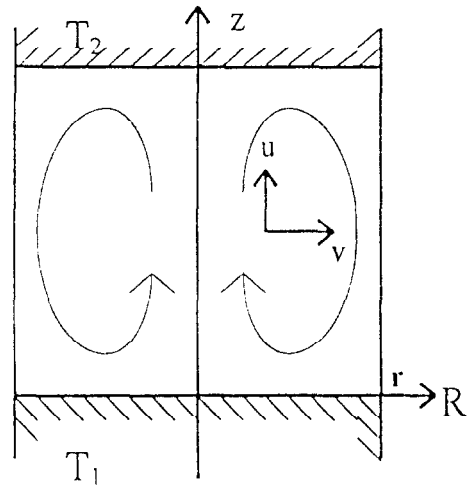thermocapillary convection pattern in the melt (b).

Fig.4 Schematic drawing of convection
by surface tension gradient

present paper is shown in Figure 4. The melt is limited in a smooth cylindrical container, not pasted on the side cylindrical wall of the crucible, that is, it is can slide along the side cylindrical wall. The temperature T$_2$ at the top of the cylinder is higher than T$_1$ at the bottom. The non-uniform distribution of temperature at cylindrical wall drives convection along the direction shown in Figure 4, when the applied temperature difference ∆T=T$_2$-T$_1$ is lower than the critical one, the three-dimensional, steady and axis-symmetric flow field and temperature distribution satisfy the following non-dimensional equations:

$$\frac{\partial \vec{U}}{\partial t} + (\vec{U} \cdot \nabla)\vec{U} = -\nabla(\Delta P) + \frac{1}{R_e}\nabla^2 \vec{U} \tag{1}$$

$$\frac{\partial T}{\partial t} + \vec{U} \cdot \nabla T = \frac{1}{M_a}\nabla^2 T \tag{2}$$

$$R_e = \frac{|\sigma_T|\Delta TL}{\rho v^2} \qquad M_a = \frac{|\sigma_T|\Delta TL}{\rho v \kappa} \tag{3}$$

with the following boundary conditions:

$$v=0, \quad \frac{\partial u}{\partial r} + \frac{\partial \Gamma}{\partial z} = 0, \quad T_r = T_1 + (T_2 - T_1)z \tag{4}$$

at the side cylindrical wall, and v=0, u=0, T=T$_1$ at the bottom, v=0, u=0, T=T$_2$ at the top, Where u and v are respectively the axial and radical components of the velocity vector $\vec{U}$, σ_T is the surface tension differentiation with respect to temperature, L represents the length of the melt region and ρ, v, κ are the density, kinematics viscosity and the thermal diffusion coefficients respectively. Figure 3 (b) is the simulated flow pattern of thermocapillary convection in Al-Al$_3$Ni melt. The pattern is similar to the radiant eutectic structure. When the solid-liquid (s-l) interface pushes upward, the flow effect still exists and has an influence on the temperature and solute redistribution as well as the configuration of solid-liquid interface. Under the temperature gradient of 5^{0}C/cm of the space furnace, the planar growth condition (G/R>1.786 × 10^5 °C for Al-Al$_3$Ni alloy) is not satisfied. According to the microstructure of Figure 3 (a), the convexity of the solid-liquid interface should be changed gradually during solidification and the smaller the pulling velocity, the more curved the solid-liquid interface (Xiong, 1993). Furthermore, the eutectic phases of Al-Al$_3$Ni alloy always directionally grow along a direction normal to the solid-liquid interface, so the formation of the radiant eutectic structure may related to the convex change of the s-l interface during the

solidification.

Marangoni Migration of Bi Droplets in Al-Bi Melt in Space

Marangoni migration of Bi-droplets in Al-melt was studied by re-melting an Al-10%Bi + Al sample in the gradient furnace under microgravity conditions in space. It was found that Bi-droplets moved in the temperature gradient of 5 °C/cm towards the hotter side of the sample. Figure 5(a) and (b) show the distribution of smaller Bi particles in Al-Bi region and larger Bi particles in Al region. Figure 5(c) shows the size distribution of particles with different diameters and the area content as a function of axial positions.

The Marangoni migration velocity V_m of droplets with mean diameter r in a constant temperature gradient can be given by the equation of Young, Goldstein and Block (Young, 1959):

$$V_m = \frac{2\dfrac{dT}{dx}\dfrac{d\sigma_{L1L2}}{dT}k}{(2k+k')(2\eta+3\eta')}r \tag{5}$$

Where η, η' κ, κ' are the viscosity and the thermal conductivity of the matrix liquid and the dispersion liquid, respectively. During solidification, the pushing velocity of s-l interface is an important factor, which influence the particle distribution. The pushing velocity of s-l interface under microgravity can be given as follows(Neumann and Goldstein, 1959):

$$V_s = \frac{2a\Delta\sigma_0 r^2}{3\eta(r+d)^2(2r+d)} \tag{6}$$

Where a is the atomic diameter, d is the distance between droplets and interface. $\Delta\sigma_0$ is the difference of the interface energy. The calculations of Marangoni migration velocities V_m and V_M of mean diameter particles and maximum diameter particles, and pushing velocity Vs of the s-l interface (in figure 5 (d)) show that the smaller Bi droplets were engulfed by the developing s-l interface and dispersed homogeneously in the Al-Bi matrix. The larger Bi droplets of which migration velocity V_m is larger than V_s moved under the temperature gradient ahead of the s-l interface and distributed into Al matrix.
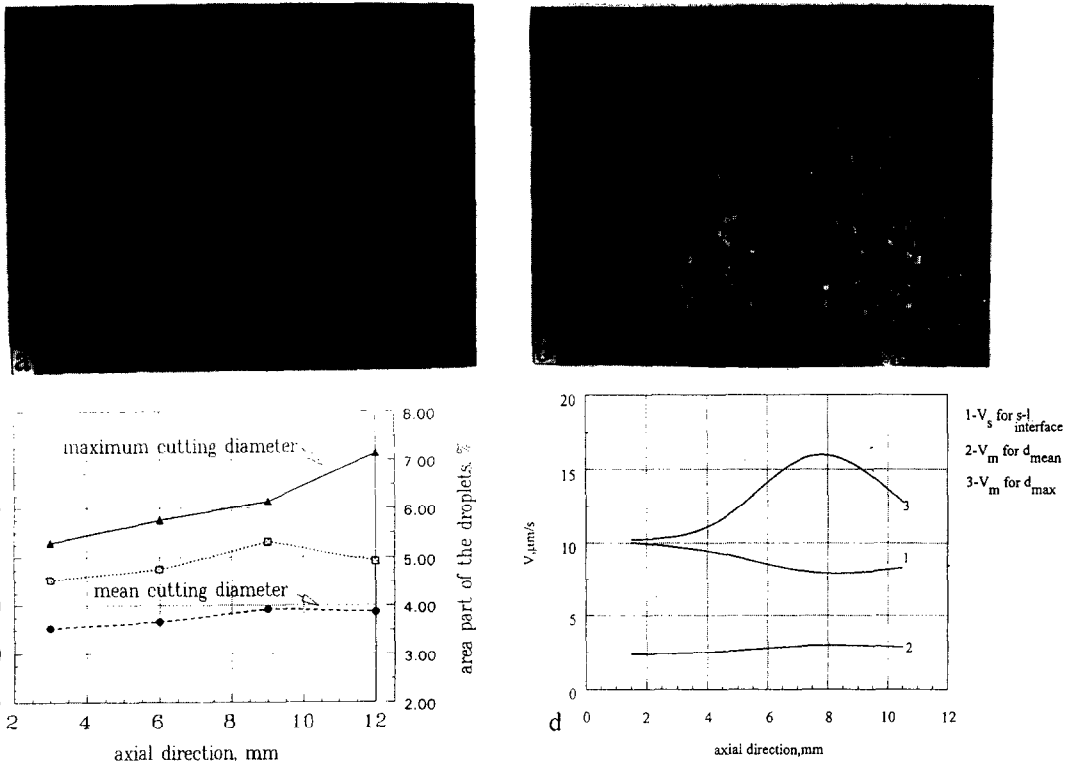

Fig.5 Distribution of Bi-particles in Al-Bi region (a) and in Al region (b) obtained in space, particle size distribution (c). Migration and pushing velocities of V_m, V_M and V_s (d) .

Application of Surface Tension Effect on Earth Solidification Process

As a result of microgravity investigation in space, the droplets of the second phase in the melt of immiscible alloys can be moved towards hotter melt by Marangoni migration, which can be used to overcome the gravity effects on earth materials processing. In the technique, the net velocity of the droplets will be dependent on its size. When the resultant velocity is zero, there will be a critical size of the droplets. By controlling the stability of the solidification process, an ingot with uniform distribution of the droplets can be obtained as the microstructure of Al-10Pb% alloy shown in Fig. 6(a), (b) and (c). The critical size r_0 of the secondary phase droplets can be estimated by the following equation (Xian, 1996):

$$ r_0 = \frac{3\eta \left|\frac{dT}{dx}\right| \left|\frac{d\sigma_{L1L2}}{dT}\right| \kappa}{g(2\kappa + \kappa')(\rho - \rho')(\eta + \eta')} \tag{7} $$

Solidification test on Al-Bi, Al-Pb and Al-Pb-Si alloys showed that the size of the Bi and Pb particles in Al matrix is roughly the same order of magnitude as that obtained by above equation.
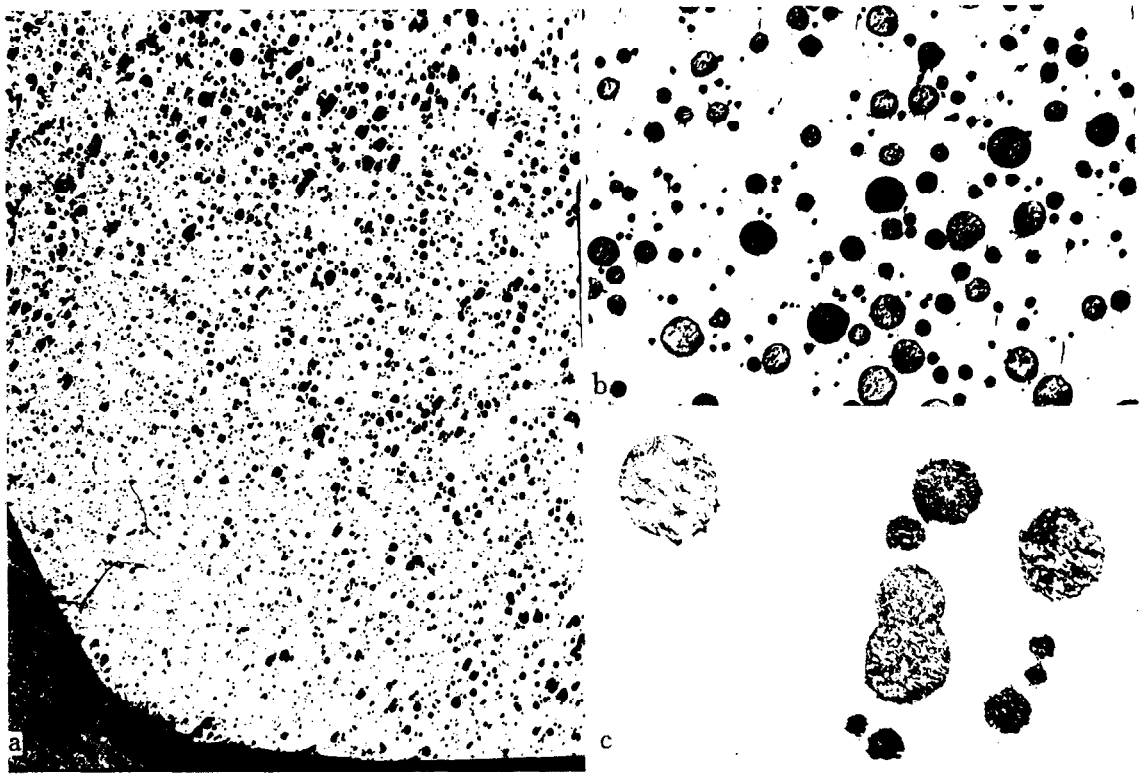

Fig. 6 Microstructure of the ingot of Al-10Pb alloy. (a)12.8 ×   (b)100 ×   (c) 400 ×

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