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Eutectic Growth Mode of Al-Si Alloy Solidified Under High Pressure

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Abstract. The Al-Si eutectic alloy solidified under normal pressure was prepared by vacuum Ar-arc furnace. The alloy solidified under high pressure was prepared by hexahedral anvil equipment. The two heavily etched samples were observed by scanning electron microscope. The experiments show that the eutectic silicon phase solidified under high pressure presents curved and near parallel lamellar crystal, and there is definite spacing between the layers, which was very different from that of solidified under normal pressure. So the eutectic growth mode of Al-Si alloy solidified under high pressure was put forward, which indicate that the growth mode of Si have been changed gradually from facet to non-facet under high pressure.

Introduction

Aluminum-silicon (Al-Si) alloy have the advantages of being lightweight, superior cooling and tribological performance, which make them suitable alternative to replace components made of ferrous alloys. Both hypo- and hypereutectic Al-Si alloys have shown promise as engine blocks material due to their adequate wear resistance and higher strength to weight ratio[1-3]. High-pressure die-casting (HPDC) is the most common shaped casting method for this material[4-6]. Pressure is an essential thermodynamic parameter which can influence the solidification process[7]. Solidification under high pressure can affect and control the nucleation and growth in alloys during solidification, which can change the distribution of second phase, uniformity of composition, and obtain non-equilibrium microstructure[8]. To obtain the suitable properties, the controlling of the microstructure is very important. In this present work, the eutectic growth mode of Al-Si alloy solidified under high pressure was put forward, which indicate that the growth mode of Si have been changed gradually from facet to non-facet under high pressure.

Experimental Procedure

The Al-Si alloys were prepared by using pure aluminum (99.9%) and pure silicon (99.99%) in water cooling copper crucible in vacuum Ar-arc furnace. The smelting process was repeated 2-3 times to guarantee the alloys uniformity. And the eutectic composition Al-13at.%Si sample was obtained. Then the alloy samples with size of 5×5×5mm packed with BN solvent were heated to the temperature of 1500℃ for 8-10 min to make samples melt fully under high pressure of 5.0GPa on the hexahedral anvil equipment. The power was cut off and sample was cooled with the cooling velocity about 10 K/s to room temperature. The scanning electron microscope (SEM) was used to observe the microstructure, and X-ray diffraction to analyze the phase construction of Al-Si alloys.

Results and Discussion

The results of X-ray diffraction (XRD) of these alloys show that the phases are α(Al) and β(Si) solid solution and no new phase is observed. The SEM micrograph of the eutectic structure of Al-13at.%Si alloy solidified under normal pressure is shown as Fig.1. The sample was processed by heavy etching to remove the α(Al) phase, leading to only β(Si) phase in (α+β) eutectic region being reserved. It can be found that the microstructure is complicated netlike structure composed of acicular eutectic Si phase branches, and there is no definite spacing between the branches.

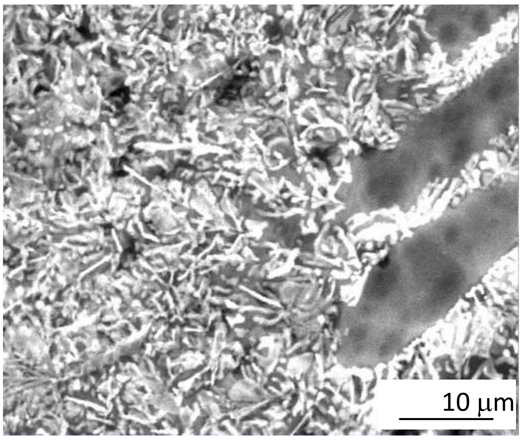

Fig.1 Scanning electron micrograph of Al-13at.%Si alloy solidified under normal pressure

Because the β(Si) phase is non-metal phase, so the solid-liquid interface of β(Si) phase is facet. In the process of eutectic growth, the Al phase and Si phase interact with each other, leading to the growth direction of Si phase changing constantly. So the solidified structure is intercross, and eutectic cell is difficult to be found[9]. If the Si crystal did not connect to each other, the Si crystal should not be reserved due to no supporting while the Al was etched off. The fact that the Si reserved connecting to each other to form the netlike structure indicate that the growth of the Si phase should not fall behind the Al phase during eutectic solidification of Al-Si alloy under normal pressure.

From above experimental result, it can be found that the eutectic structure of Al-Si alloy solidified under normal pressure show typical anomalous eutectic structure. This is because that Al-Si alloy is typical metal-nonmetal alloy, which have different mechanism of growth between metal and nonmetal. The interface between solid and liquid for mental aluminum is atomically rough, on which the atom can occupy easily at any position, leading to the mental Al grow not in a certain direction. On the other hand, the interface between the solid and liquid for nonmetal Si is atomically flat, called facet, leading to the nonmetal Si grow in the certain direction. For crystal Si, the certain direction is along <2ll> or <110> crystal orientation of {111}crystal plane, forming the acicular structure.

Due to the directional solidification of nonmetal Si, the crystal Si would grow approaching to or departing from each other. So the thickness of the Si branches would not uniform as shown in Fig.2. There would be lack of Si element near the solid-liquid interface if two neighboring crystal Si approach to each other. When the space between them gets to a minimum λ_m, the growth would be stopped due to surface energy effect. On the other hand, when the space between them gets to a maximum λ_M, the nonmetal Si would change the direction to the Si enrichment region through twin crystal or sub grain boundary[10], and form the Si crystal branches to connect to each other. The space between them would be reduced, and vary around an average value. So the branched eutectic Si would be solidified to an intercross structure, and the eutectic cell is difficult to be observed.

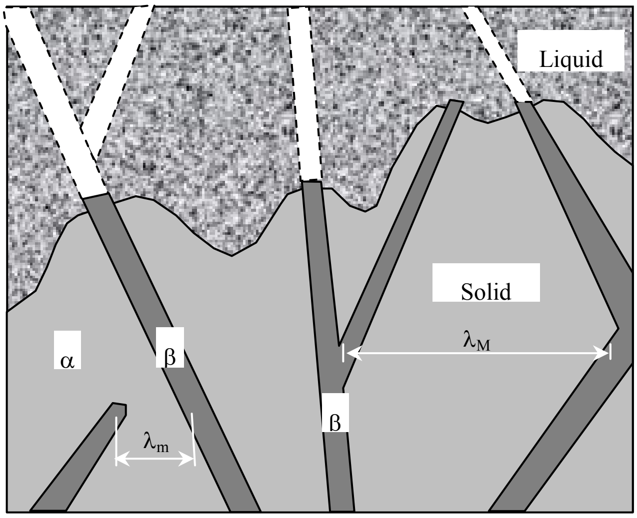

Fig.2 Schematic diagram of the growth of an irregular eutectic showing the disappearance of a lamellae at $\lambda=\lambda_m$ and the branching of a lamellae at $\lambda=\lambda_M$

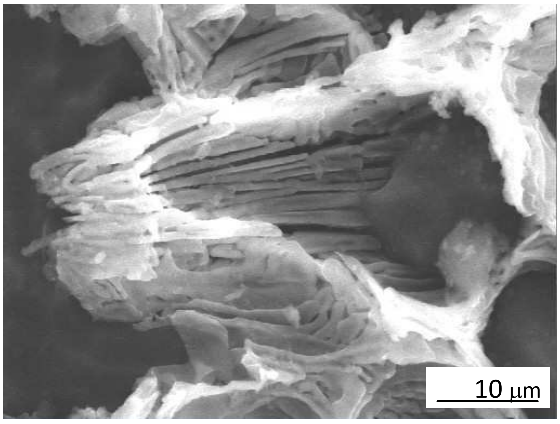

Fig.3 Scanning electron micrograph of Al-13at. %Si alloy solidified at 5 GPa

The SEM micrograph of the eutectic structure of Al-13at.%Si alloy solidified under high pressure of 5GPa is shown as Fig.3. The primary α phase and eutectic α phase were removed due to the heavy etching, leading to form concave pit, in which the morphology of the eutectic lamellar could be found. From Fig.1 and Fig.3, it could be found that the eutectic structure of Al-Si alloy solidified under pressure of 5GPa is very different from that of under normal pressure, which is curved and parallel lamellar, rather than the complicated acicular ones under normal pressure. The space between two neighboring Si layers is constant and the rosette eutectic cell is shown clearly.

The reason why the microstructure solidified under high pressure is different from that under normal pressure may be that the growth of Si crystal have changed from the facet fashion of nonmetal to the non-facet fashion of mental. The growth direction is no longer along a certain orientation, but changing direction continuously. And there is no approaching to or departing from growth tendency between two neighboring layers, resulting of the constant space between two neighboring layers. This indicates that the growth mode have changed from the facet fashion under normal pressure to the non-facet fashion under high pressure as shown in Fig.4. The principle is as following:

When the eutectic crystallization is under high pressure, the leading phase act as the nucleation center and eutectic cell grow around this center. The growth velocity of eutectic Si crystal in front of the solid-liquid interface is no longer faster than the one under normal pressure, but being depressed by the high pressure. According the thermodynamics relation:

$$\left[\frac{\partial(\Delta G_v)}{\partial P}\right]_T = V^L - V^S = \Delta V \tag{1}$$

where ΔGv is free energy, P is pressure, V^L and V^S are the molar volume of liquid and solid, respectively. If the volume of nucleation phase is less than that of liquid phase, i.e., $\Delta V > 0$, then the free energy ΔGv increase as the pressure increase (for example Al, Cu). On the other hand, if the volume of nucleation phase is great than that of the liquid phase, i.e., $\Delta V<0$, than the free energy ΔGv will decrease as the pressure increase (Si, Ge). So during the eutectic growth of Al-Si alloy under high pressure, the pressure effect is in favor of Al phase. According to the nucleation theory, the growth velocity of crystal can be expressed as:

$$U = f\frac{D}{a_0}\left[1 - \exp(-\frac{\Delta G_v}{RT})\right] \tag{2}$$

where f is geometrical factor of interface, D is diffusion coefficient, a_0 is average space of atoms, and R is gas constant. When the cooling velocity is small, i.e., $\Delta Gv/RT<<1$, equation (2) can be approximately expressed as:

$$U \approx \frac{fD\Delta G_v}{a_0 RT} \tag{3}$$

Partial derivative of equation (3) to P can lead to the relation as following:

$$\left(\frac{\partial U}{\partial P}\right)_T = \frac{fD}{a_0 RT}\left[\Delta G_v\left(\frac{\partial \ln D}{\partial P}\right)_T + \left(\frac{\partial \Delta G_v}{\partial P}\right)_T\right] \tag{4}$$

Due to $\left(\frac{\partial \ln D}{\partial P}\right)_T = -\frac{\Delta V^*}{RT}$, and $\left(\frac{\partial \Delta G_v}{\partial P}\right)_T = \Delta V$,

Substituting above relation into equation (4) can derive relation as following:

$$\left(\frac{\partial U}{\partial P}\right)_T = \frac{fD}{a_0 RT}\left[-\Delta G_v\frac{\Delta V^*}{RT} + \Delta V\right] \tag{5}$$

where ΔV^* is diffuse activation volume. When the cooling velocity is small, i.e., $\Delta Gv/RT<<1$, equation (5) can be approximately expressed as:

$$\left(\frac{\partial U}{\partial P}\right)_T = \frac{f}{RT}\cdot\frac{\Delta V}{a_0}\cdot D \tag{6}$$

Because the atomic diffusivity in alloy fondant decrease as the pressure increase, so the pressure effect decrease the diffusivity for both Al atoms and Si atoms, and the influence to diffusivity D in equation (6) is same to both Si crystal and Al crystal growth. Therefore, the item $\Delta V/a_0$ in equation (6) will play the key role in determining which phase (Al phase or Si phase) will precipitate first. According to equation (1), $\Delta V > 0$ for Al element, and $\Delta V < 0$ for Si element. So the increase of pressure will favor the growth of Al phase, and restrain the growth of Si phase.

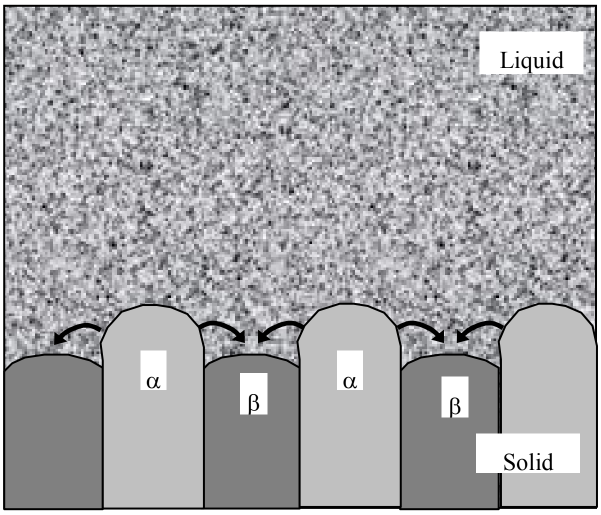

Fig.4 Schematic diagram of the growth of an irregular eutectic at high pressure

Since the Si phase was depressed and Al phase was promoted during the solidification of Al-Si alloy under high pressure, so there is the convex eutectic Al phase at the both side of the Si phase, leading to the solid-liquid interface of facet Si phase is concave face as shown in Fig.4. Therefore, the facet Si phase will lose the condition of facet growth mode, and the Si phase will grow according to non-facet (atomically rough) growth mode. The reason is that the facet growth mode can only take place when the solidification is carried through on a convex solid-liquid interface, and the solid-liquid interface is tangency with the plane of facet shown as the right panel of Fig.5. On the contrary, if the interface between solid and liquid is concave face, the new atomic layer will provide the growth step around the solid phase at any place in the concave face. So the facet growth, which was a lateral growth mode, will be difficult to carry on. The interface between the two phases Si is concave plane, which make the facet could not grow according to the facet mode. So the eutectic crystallization could only carry on according to the nonfacet-nonfacet mode.

Summary

The growth of Si phase was no longer the facet one, but changed to the continuous growth of rough face. However, the growth of Al phase was still the continuous growth when the Al-Si eutectic alloy solidified under high pressure. Therefore, the growth of both Si phase and Al phase became the nonfacet-nonfacet eutectic mode. The two interactional phases grow to form the rosette eutectic cell.

Acknowledgments

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