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Review

Characterization of microstructures and growth orientation deviating of Al_2Cu phase dendrite at different directional solidification rates

Ka Gao [a,*], Shaojie Song [b], Shuangming Li [a], Hengzhi Fu [a]

[a] Zhengzhou University of Aeronautics, Zhengzhou 450015, China
[b] State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an 710072, China

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At different directional solidification rates (10, 20 and 100 μm/s), microstructures and growth orientation variations of Al_2Cu dendrite in Al-40%Cu alloy were characterized. When solidification rates were ranged from 10 to 100 μm/s, three-dimensional microstructure of Al_2Cu dendrite changed from faceted L-shaped patterns to non-faceted complex dendrite morphology in transverse section. By the macro and micro orientation analysis characterize methods, [001] growth direction of Al_2Cu dendrites with different morphologies was determined. The deviation angle between [001] direction and the heat flow direction was increased with solidified rate increasing. The experimental results showed that the regular solidified microstructure and growth orientation along the heat flow direction could be well controlled under lower directional solidification rate.

Contents

1. Introduction

In the process of dendrite growth, the primary dendrite can form various morphologies and microstructure distributions under directional solidification [1–5], which could result that the toughening methods mainly including the fine grain toughening and the second phase toughening occurs. This seriously leads to the crack propagation path deflecting, which improves the fracture toughness and affects mechanical property of the alloys. Thus, the study on dendrite growth is a significant practical importance [6,7]. So far more works indicated that dendrite microstructure evolution was strongly related on the directional solidification rate and the alloy composition, especially the former effect factor. The effect of solidification rate on dendrite growth morphology should been concerned. Li et al. [8] observed various Si dendrite microstructures and the columnar to equiaxed dendrite transition in Al-Si alloy

* Corresponding author.
E-mail address: gaoka9222005@163.com (K. Gao).

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under different directional solidification rates. Wang et al. [9] studied the microstructure evolution of Al-Cu alloy through changing the directional solidification rate. They found that the planar-to-cellular transformation of Al dendrite morphology occurred with the solidification rates increasing. Besides, Liu et al. [10] investigated the interface morphologies of single crystal super alloy CMSX-2 and found that the evolution of interface morphologies of dendrite bad been changed with directional solidification rate increasing. The above results indicated that the solidification rate played a vital role in dendrite growth.

On the other hand, when dendrite morphologies evolution occurred at different directional solidification rates, its growth orientation variations were also accompanied [6]. Then, dendrite would grow along its growth orientation in growth process. So it is very important to investigate the growth orientation variations of dendrite. Until now, more previous studies ascribed that there had been three main specific directions for dendrite growth under directional solidification: the dendrite growth orientation, the heat flow direction and the preferred crystallographic orientation [11]. And then the relationship among them was transformed with the solidification rate changing. Ardakani et al. [12] showed that the growth orientation of nickel-base single crystal super alloys was [001] direction, which was between the heat flow direction and the preferred orientation during directional solidification. Further, Esaka and Shionzuka et al. [13] found that the dendrite growth orientation gradually turned toward its preferred orientation with the directional solidification rate increasing. However, until recently, the deviation degree between them were not given and discussed, which would lead to the dendrite growth direction not controlled well. Therefore, it is necessary to provide effective analysis methods to characterize dendrite growth orientation deviating during directional solidification.

Thus, in this work, the two-dimensional (2D), three-dimensional (3D) microstructure evolution and growth orientation of Al_2Cu dendrite were investigated during directional solidification. Moreover, the deviation relationship between the dendrite growth orientation and the heat flow direction was characterized and analysed by the macro orientation (the rotating orientation X-ray diffraction) and the micro orientation (the electron back-scattered diffraction) analysis methods.

2. Experimental procedures

2.1. Materials

Al-40 wt.% Cu alloy used in this study was prepared in a vacuum induction melting furnace with purity aluminium (99.9 wt.%) and copper (99.99 wt.%). The composition of the ingot measured by chemical titration is Al-39.2 wt.% Cu. The cast sample were enveloped in high purity Al_2O_3 tube with a inner diameter of 4 mm and length 100 mm. Directional solidification experiments were carried out using a Bridgman vertical vacuum furnace described elsewhere [14]. In this study the thermal gradient was measured using a Φ0.5 mm NiCr–NiSi thermocouple and its value was about 250 K/cm at 10 μm/s. In directional solidification process, the sample was heated by a graphite heater at 750 °C and then held isothermal for 20 min. Subsequently, the sample was moved downwards at 10, 20, and 100 μm/s, respectively. In order to keep the S/L interface, when the directional solidification distance reached 50 mm, the sample was quenched into a liquid Ga–In–Sn pool.

2.2. Characterization

The directionally solidified samples obtained in the experiments were then sectioned horizontally and vertically, respectively. And the specimens polished and etched using solvent Kroll of H_2O (46 mL) + HNO_3 (3 mL) + HF (1 mL) for about 15 s. The scanning electron microscopy (SEM, JSM-6390A) were employed to photograph the specimens microstructures. The materialise's interactive medical image control system (Mimics) software was applied to reconstruct the three-dimensional (3D) microstructures images of the primary Al_2Cu phase. The growth orientations were investigated by means of the X-ray powder diffraction (XRD, D/max-3) and the electron back-scattered diffraction (EBSD) in scanning electron microscopy (SEM, Zeiss Supra 55) equipped with the Channel 5 EBSD system (HKL Technology-Oxford instrument). In addition, the deviation angle in directional solidified alloy was measured by the rotating orientation X-ray diffraction (RO-XRD) method in X-ray diffractometer (XRD, Rigaku's D/max2400).

3. Results and discussions

3.1. Microstructure evolution (2D and 3D)

It is well known that the solidified microstructures of Al-40%Cu alloy consist of primary θ-Al_2Cu phase and eutectic (Al/Al_2Cu) phase based on the Al-Cu phase diagram shown in Fig. 1. In directional solidification process, when the melt temperature was cooling down, the θ-Al_2Cu phase would be precipitated firstly as the primary phase from the liquid metals and grew along its growth directions to form dendrite morphology. With a large number of Al_2Cu dendrite precipitating and growing, the composition of the remaining liquid melt was gradually reduced to the eutectic composition. Then the remaining liquid occurred the eutectic reaction and developed in the form of the coupled eutectic (Al/Al_2Cu). When at directional solidification rate of 10 μm/s, the primary Al_2Cu dendrite could be clearly distinguished from the eutectic in Fig. 2(a). The dendrite displayed regular faceted L-shaped patterns [15] in transverse section and the eutectic exhibited regular lamellar structure morphology. When solidification rate increased to 20 μm/s, Al_2Cu dendrite displayed irregular non–faceted L-shaped patterns and I-shaped patterns shown in Fig. 2(b). The surface of the dendrite began roughening, which resulted in its edges and corners gradually disappeared. Moreover, the dendrite size was smaller than alloy at 10 μm/s. While the primary Al_2Cu dendrites surface roughened further with the solidification rate increasing. Then at 100 μm/s the dendrite displayed irregular complex three lobately morphology with non-faceted characteristics in Fig. 2(c). The dendrite edges and corners were completely smooth and its size was smallest, which

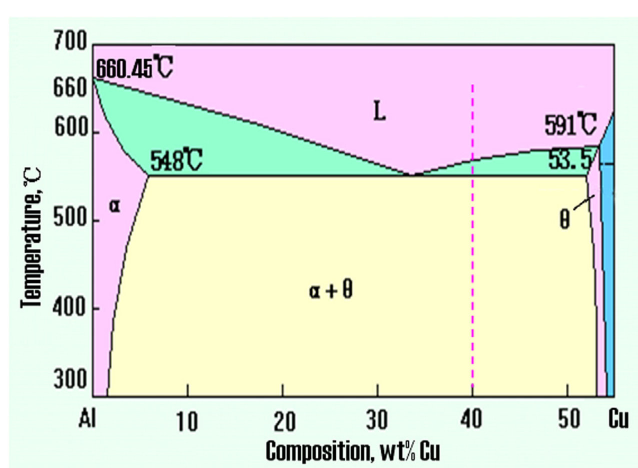

Fig. 1. Phase diagram of Al-Cu alloy.

differed from the microstructures shown in Fig. 2(a). That implies that the microstructure of the Al_2Cu dendrite can be transformed by changing its growth rate [16,17].

Similar to the internal structure of the human body analyzed by X-ray computed tomography systems (CT) in medicine, in this work milling off the surface of the sample with the stayed step-size layer by layer by the serial sectioning technique [18,19], the microstructures images of the polished samples were obtained. Then, the obtained images were transferred into Materialise's interactive medical image control system, fixed position, edited, and reconstructed into three-dimensional (3D) microstructure of Al_2Cu dendrite during different directional solidification rates shown in Fig. 2, which provides a novel way to deeply understand the crystal growth of the solidified phases. At 10 μm/s, Al_2Cu dendrite was observed having faceted edges and surface, and its growth orientation deviated significantly from the heat flow direction in Fig. 3(a). Preliminary research results [20] showed that the growth orientation of Al_2Cu phase was its [001] direction during directional solidification. By cutting its three-dimensional microstructure along the radial direction, the two-dimensional (2D) morphologies in transverse section displayed the L-shaped pattern, which was consisting with the above result (in Fig. 2(a)). While at 20 μm/s, solidification rate increasing caused the faceted interface instable [21]. In Fig. 3(b), the 3D microstructure of Al_2Cu dendrite had non-faceted and coarse surface. Then the dendrite was observed as similarity L-shaped pattern and grew not along the heat flow direction. When the solidification rate increased to 100 μm/s, Al_2Cu dendrite displayed long rod-shaped dendrite with non-faceted surface, and there had been a large deviation angle between its growth orientation and the heat flow direction in Fig. 3(c). The above analysis showed that the solidification rate changing affected greatly the Al_2Cu dendrite morphology.

3.2. Growth orientation analysis

Seen from the three-dimensional microstructures in Fig. 3, the deviation angle between the growth orientation of Al_2Cu dendrite and the heat flow direction became larger with the growth rate increasing. Firstly, based on reported by Huang et al. [22], in this study the Al_2Cu dendrite growth orientations were measured by using XRD. Fig. 4 is the XRD spectrums for Al-40%Cu alloy in transverse sections at different rates. Only two diffraction peaks (110) and (004) of Al_2Cu dendrite appeared in the sample at 10 μm/s. It indicated that the growth orientations of Al_2Cu dendrite dendrite were the normal direction of (110) and (004) planes ([110] direction and [001] direction). At 20 μm/s, the diffraction peak (110) disappeared. Then (330) and (004) planes appeared. Here Al_2Cu dendrite growth orientations were also its [110] direction and [001] direction. When at solidification rate of 100 μm/s, the crystal plane (330) disappeared and only the diffraction peak (004) increased. Apparently, Al_2Cu dendrite had a strong preferred growth orientation at its [001] direction.

Further, the growth orientations of Al_2Cu dendrite under directional solidification were investigated and characterized by the EBSD analysis. Fig. 5 shows the EBSD maps in the transverse section, the corresponding (100)-pole figures and inverse pole figure of Al_2Cu dendrite at different directional solidification rates, respectively. The L-shaped patterns could be observed at solidification rate of 10 μm/sin Fig. 5(a). From the pole figure, the center position was the heat flow direction. Three poles of Al_2Cu phase were sited near the center and circle of the pole figure (Fig. 5(b)). It is easy to deduced that Al_2Cu dendrite had oriented with its [001]

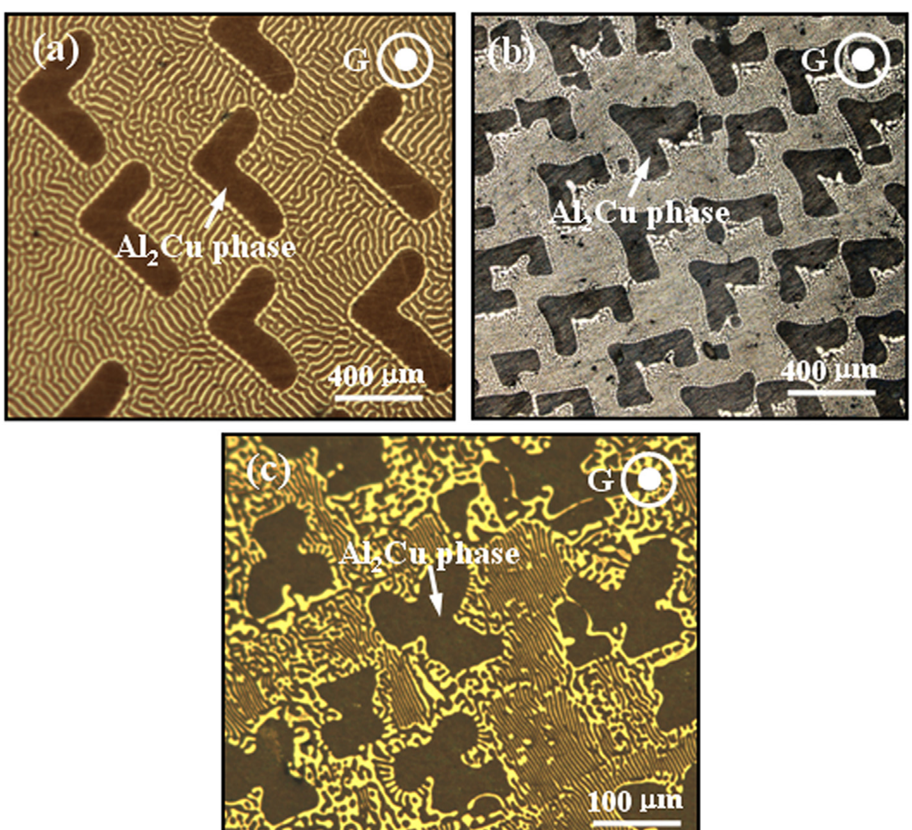

Fig. 2. Directionally solidified microstructures of Al-40%Cu hypereutectic alloy at different solidification rates in transverse section: (a) at 10 μm/s, (b) at 20 μm/s, and (c) at 100 μm/s.

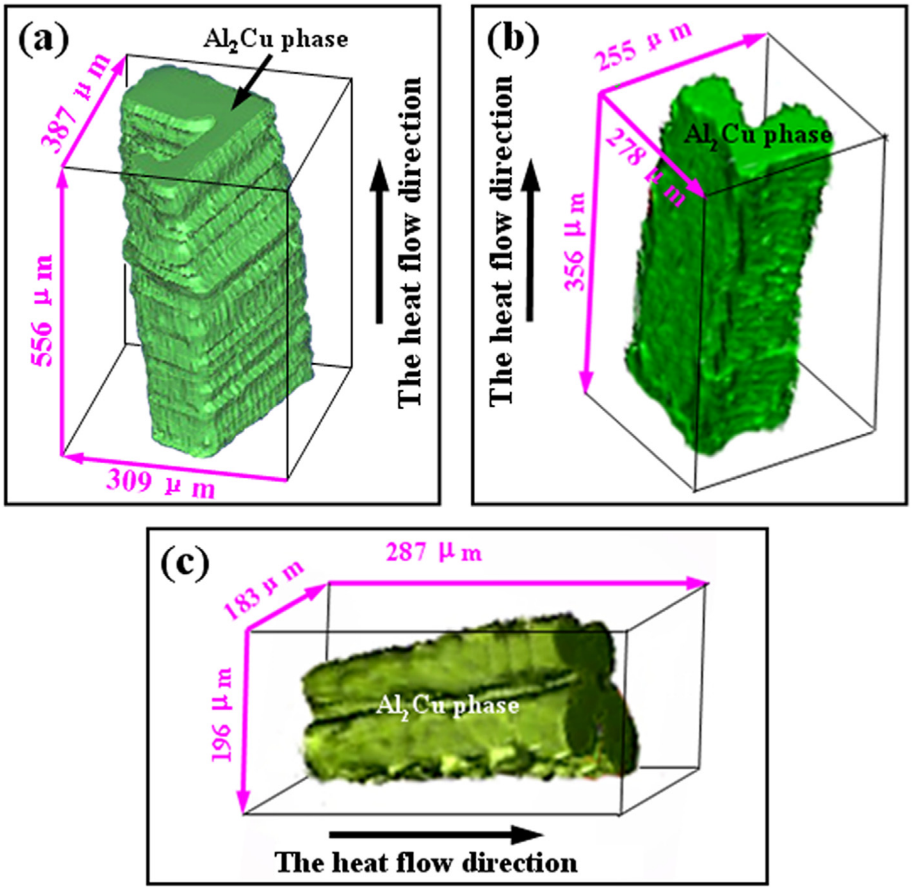

Fig. 3. The three-dimensional (3D) microstructure of the directionally solidified Al-40%Cu alloy at different solidification rates: (a) at 10 μm/s, (b) at 20 μm/s, and (c) at 100 μm/s.

crystal direction by the application of directional solidification. And the [001] direction was near by the heat flow direction. Then the deviation angle between [001] direction of Al_2Cu dendrite and the heat flow direction was about 7.69 °. Meanwhile, in the inverse pole figure of Fig. 5(c), the preferred growth orientation of Al_2Cu dendrite was only its [001] direction. With the solidification rate increasing, the mainly growth orientation of Al_2Cu phase were also its [001] direction both in the samples at 20 and 100 μm/s; seen from the Fig. 5(d) to (i). The deviation angle between its [001] direction and the heat flow direction were about 8.02 ° and 8.74 ° shown in Fig. 5(e) and (h), respectively, which were given in Table 1. The above results indicated that with solidification rate decreasing, the growth orientation of Al_2Cu dendrite was further deviated from the heat flow direction. That was agreeing well with the 3D microstructure results in Fig. 3.

3.3. The characteristics of the deviation angle

The following experiments were designed to characterize the deviation degrees changing between growth orientation of the Al_2Cu dendrite and the heat flow direction at different directional solidification rates. According to the above research results, we know that the (110) plane diffraction intensity of Al_2Cu dendrite is the strongest in the XRD diffraction spectrum standard. In addition, the normal direction of (110) plane is perpendicular to the [001] direction (Fig. 6). Then the deviation degree between Al_2Cu dendrite growth orientation and the axial direction (the heat flow direction) could be characterized by the deviation angle between the (110) plane and the longitudinal-section surface of the directional solidified sample. Therefore, in this work, we selected the (110) plane to do further analysis. Fig. 7 shows the butterfly diagram of symmetrical Gaussian envelope lines for the crystal plane (110) at different solidification rates, in which the RO-XRD method [23,24] was employed.

When at solidification rate of 10 μm/s, two pairs of symmetrical Gaussian peaks occurred in Fig. 7(a), indicated that there were not

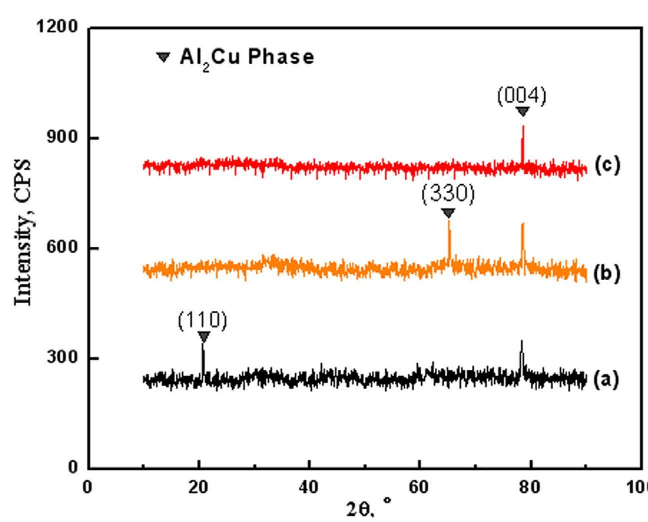

Fig. 4. XRD patterns of Al-40% Cu alloy at different directional solidification rate: (a) at 10 μm/s, (b) at 20 μm/s, and (c) at 100 μm/s.

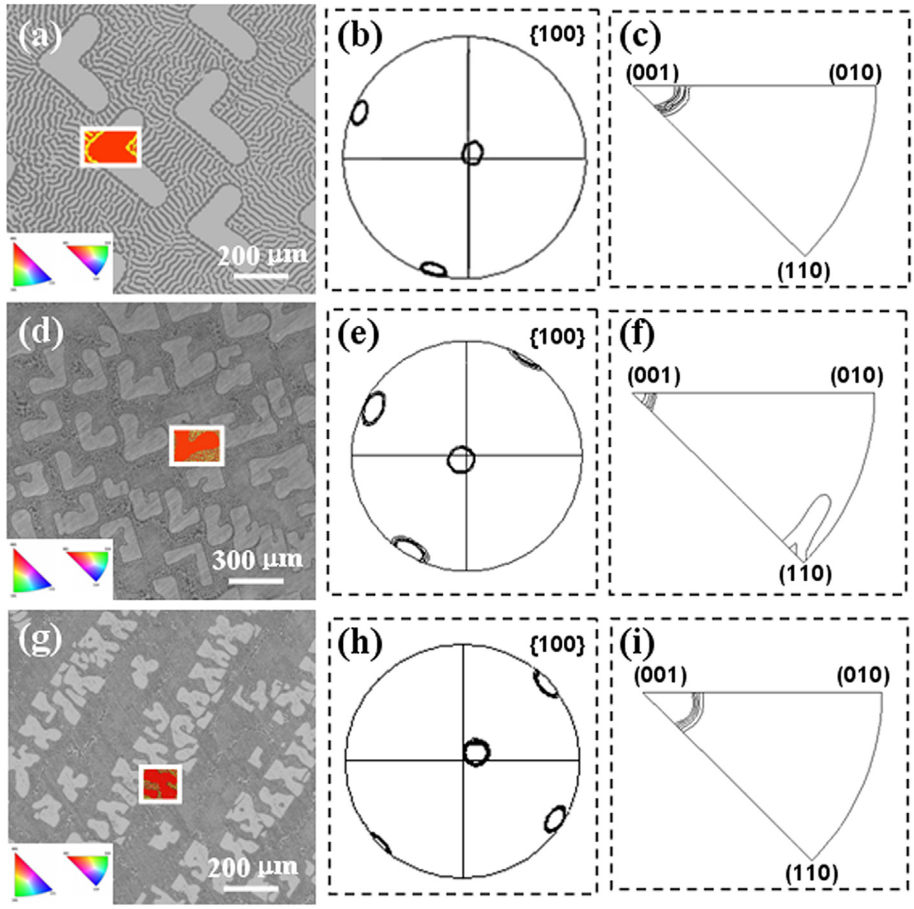

Fig. 5. (a–c) the transverse section microstructure, the corresponding (100)-pole figure of Al_2Cu phase and inverse pole figure of Al-40%Cu hypereutectic alloy at 10 μm/s, respectively; (d–f) the transverse section microstructure, the corresponding (100)-pole figure of Al_2Cu phase and inverse pole figure of Al-40%Cu hypereutectic alloy at 20 μm/s, respectively; and (g–i) the transverse section microstructure, the corresponding (100)-pole figure of Al_2Cu phase and inverse pole figure of Al-40%Cu hypereutectic alloy at 100 μm/s, respectively.

less than two grains appeared in the sample macro-surface. On the basis of the work with Guo et al. [25], the deviation angle was calculated. The (110) plane deviated from the heat flow direction about 7.24 ° (shown in Table 2), which was almost equal to the deviation angle between Al_2Cu dendrite [001] direction and the heat flow direction shown in Table 1. In the same way, the deviation angles between (110) plane and the heat flow direction were calculated about 7.73 ° and 8.91 ° at 20 μm/s and 100 μm/s shown in Fig. 7(b) and (c), respectively. The deviation degree became lager with the solidification rates increasing, which was consistent with the EBSD results.

3.4. The mechanism of the growth orientation deviating

Al_2Cu dendrite morphology had significant differences at different rates. Especially the degrees of deviation between growth orientation and the heat flow direction were changing. The mainly reason was that the relationship changing between the preferred orientation and the heat flow direction resulted in the dendrite growth orientation deviating with the directional solidification rate changing [11,13].

Table 1
The deviation angle between [001] direction of Al_2Cu phase and the heat flow direction.

Sample	The deviation angle (°)
The sample at 10 μm/s	7.69 °
The sample at 20 μm/s	8.02 °
The sample at 100 μm/s	8.74 °

Table 2
The deviation angle between (110) plane of Al_2Cu phase and the heat flow direction.

Sample	The deviation angle (°)
The sample at 10 μm/s	7.24 °
The sample at 20 μm/s	7.73 °
The sample at 100 μm/s	8.91 °

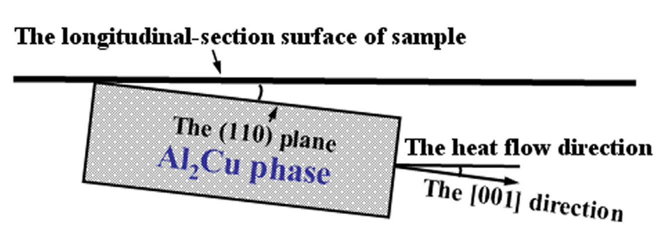

Fig. 6. Schematic diagram of two type of deviation angle.

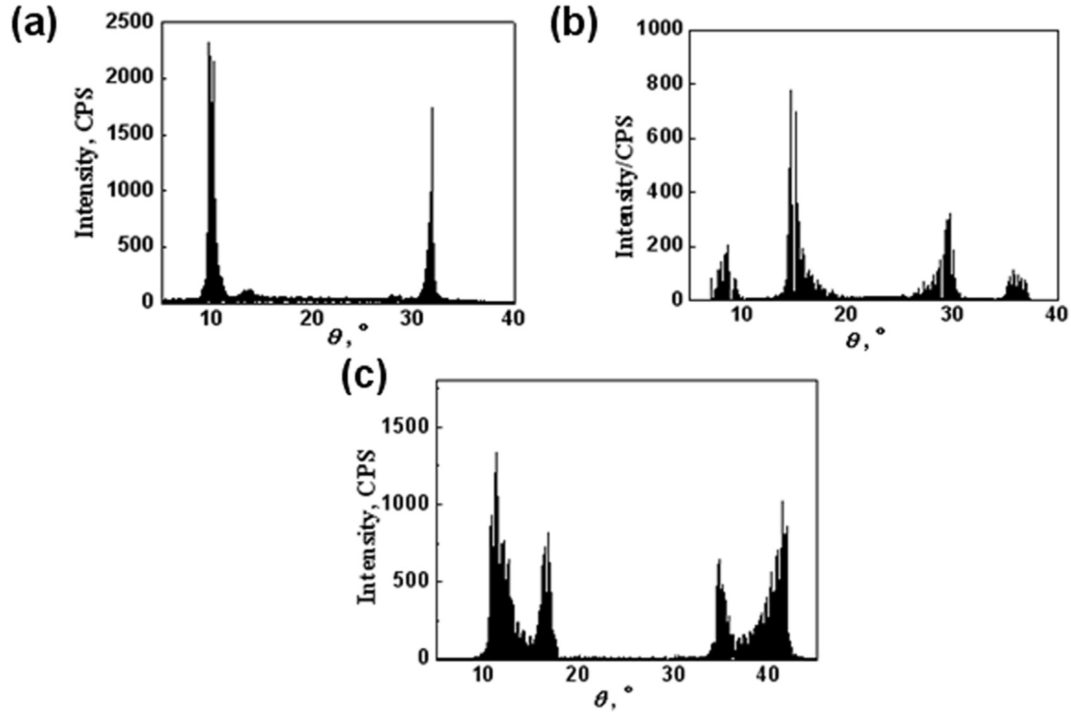

Fig. 7. RO-XRD patterns of (110) plane of Al_2Cu dendrite at different solidification rates: (a) at 10 μm/s, (b) at 20 μm/s, and (c) at 100 μm/s.

In directional solidification process, the growth orientation was between the preferred orientation and the heat flow direction in Fig. 8(a). When the solidification rate was lower, the heat flow factor would play a mainly role on dendrite growth. Then the dendrite would grow near along the heat flow direction, and deviated from its preferred orientation, as shown in Fig. 8(b). With the solidification rate increasing, the solute gradient destabilizes the interface. The interface of Al_2Cu dendrite was instability, leading to solute enrichment. Then the rate of atoms deposition on different crystal planes would be changed, which is related on the interfacial surface energy. Atoms aggregated easily on the crystal plane with higher interfacial surface energy, because of fewer satisfied bonds, which resulted in the anisotropy of the interfacial surface energy. While the anisotropy of interfacial surface energy gradually determined the dendrite growth. Its growth orientation began to deflect to the preferred orientation (Fig. 8(c)), which resulted in the deviation degrees between growth orientation and the heat flow direction gradually increasing. Based on the above theories, in this work the preferred orientation of Al_2Cu dendrite was its [110] direction, which is perpendicular to the heat flow direction. If the solidification rate was increasing, the growth orientation of Al_2Cu dendrite would approach to its [110] direction and deviate from the heat flow direction. Then the dendrite grew along its growth orientation, which had a large deviation angle with the heat flow direction.

During directional solidification, the microstructure and growth orientation of the Al_2Cu dendrite were influenced by solidification rate. The results showed that the regular solidified microstructure and growth orientation along the heat flow direction could be well controlled under lower directional solidification rate. Moreover, this work provided effective micro and macro orientation analysis methods to characterize dendrite growth orientation variations during directional solidification.

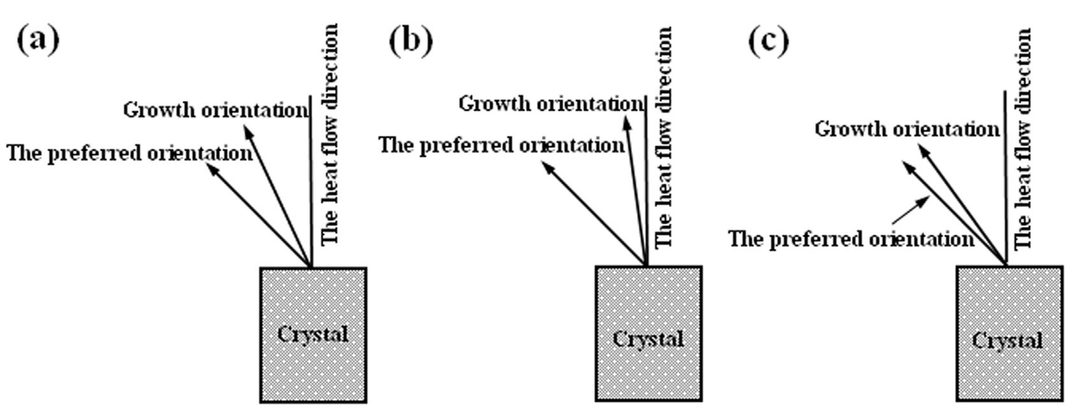

Fig. 8. Relationship of preferred orientation, growth orientation and the heat flow direction: (a) at normal solidification rate, (b) at lower solidification rate, and (c) at higher solidification rate.

4. Conclusions

The microstructures and growth orientation of Al_2Cu dendrite in directionally solidified Al-40%Cu hypereutectic alloy under different directional solidification rates were investigated. With the solidification rate ranging from 10 to 100 μm/s, the primary Al_2Cu dendrites in transverse section changed from faceted L-shaped patterns to non–faceted complex morphology. By using the serial sectioning technique, the three-dimensional (3D) microstructures of Al_2Cu dendrite at different rates were observed. The Al_2Cu dendrites all grew along its [001] direction at solidification rate of 10, 20 and 100 μm/s. With the solidification rate increasing, the deviation angle between growth orientation of Al_2Cu dendrite and the heat flow direction increased by micro and macro orientation analysis methods. The deviation degree was influenced by the solidification rate. A brief description for mechanism of the growth orientation deviating was given.

Acknowledgments

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