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Intermetallic Al_2Cu orientation and deviation angle measured by the rotating directional test during directional solidification

K. Gao*, S.M. Li, H.Z. Fu

State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi' an 710072, P. R. China

*Corresponding author. E-mail: gaoka9222005@163.com, lsm@nwpu.edu.cn, fhzh@pub.xaonline.com

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ABSTRACT

The orientation and deviation angle of intermetallic Al_2Cu phase in directionally solidified Al-40wt. %Cu hypereutectic alloy were investigated using a rotating orientation X-ray diffraction (RO-XRD) method. Experimental results show that preferred planes (110) and (310) of the Al_2Cu phase occur at 10 μm/s and the growth directions of the two planes are not well aligned with the heat flux direction. The growth direction of the preferred plane (110) has a 7.24 º~11.43 º angle with the heat flux direction. For the direction of the plane (310), its deviation angle attains 2.68 º~20.82 º. Besides, the measured data agree well with the previously reported results, indicating that the RO-XRD method is an effective method for measuring the orientation and deviation angle of the phase in polycrystalline materials. Copyright © 2011 VBRI press.

Keywords: Directional solidification; intermetallic compound; RO-XRD; orientation and deviation angle.

Gao Ka, a PhD student who is presently studying and working at State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, P.R. China. He is also undertaking at the National Natural Science Foundation of China through Grant No. 50971101 and 51074127 projects and the fund of the State Key Laboratory of Solidification Processing in NWPU (34-TP-2009). His research interest is the study of intermetallic phase orientation during directional solidification.

Li Shuangming, a Professor and PhD of materials science, who had graduated and obtained doctor of engineering degree in 1999 from Northwestern Polytechnical University, in China. He is working on the solidification processing fundamentals, single crystal growth and microstructure characterization at State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, P.R. China. He had published more than sixty research papers in English and Chinese. He is the member of Chinese Society of Aeronautics and Astronautics.

Fu Hengzhi, a Professor, Academician of Chinese Academy of Engineering and PhD of materials science, who had graduated and obtained doctor of engineering degree in 1962 from PhD, Leningrad Polytechnical University, in Russia. He is working on the solidification theory and technology on non-equilibrium solidification, near rapid directional solidification and ultra-refinement of microstructure, directional solidification of superalloys, rare-earth magnetic materials and their microstructures and properties, electro-magnetic confinement of Ni-base and Ti-base alloys as well as the growth orientation and structure control of intermetallic materials at State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, P.R. China. He had published more than 350 research papers in English and Chinese.

Introduction

Dendrite is the most common and important growth manner during alloy solidification. In constrained growth, for example, the dendrite often grows along with a certain angle deviating from the heat flux direction. This dendrite growth orientation and behavior in directional solidification have attracted much attention, since the dendrites with various directions have different growth characteristics which remarkably affect the properties of the solidified alloy [**1-5**]. Indeed, some methods have been proposed to measure the dendrite growth orientation, including the X-ray diffraction (XRD) [**6, 7**], the electron backscatter diffraction (EBSD) technique [**8**] and the transmission electron microscope (TEM) [**9**]. The EBSD method can be used to determine the crystal orientation relationship at micro-scale, and TEM can give the exact crystal orientation relationship in a selected area. However, to characterize the crystal orientation relationship at macro-scale, the XRD method has some advantages for low testing cost, high efficiency as well as the simply sample processing [**10, 11**]. In single crystal growth, the XRD is often applied to determine the preferred growth direction. For an instance, a single crystal copper prepared by continuous casting shows that the direction of crystallization is [100], and the degree of this direction deviation from the crystal axis is less than 5 º [**12**]. For polycrystalline materials, few investigations have been reported for the crystal orientation and deviation angle measured by the XRD [**13, 14**].

In this study, different intermetallic Al_2Cu orientations and deviation angles are revealed to illustrate the growth behavior of the intermetallic Al_2Cu phase. Hence the objective of this paper is to investigate the intermetallic Al_2Cu orientation and deviation angle in directional solidification of Al-40wt. %Cu hypereutectic alloy by the rotating orientation X-ray diffraction (RO-XRD) method.

Experimental

Principle of RO-XRD

To determine the crystal orientation at macro-scale, the Bragg diffraction angel θ and the main diffraction planes need to be determined by the routine XRD method. **Fig. 1** shows the schematic principle of the rotating orientation X-ray diffraction (RO-XRD) method. In **Fig. 1(a)**, if the diffraction plane has a deviation angle Φ to the sample surface, the detector fixed at the θ_1 with the sample surface can receive the crystal plane diffraction lines. Further, the sample rotates along its normal line and the detector again receives the crystal plane diffraction peaks at the angle θ_2, as shown in **Fig. 1** (**b**). Combining with the **Fig. 1** (**a**) and **1** (**b**), the deviation angle $\Phi = (\theta_2 - \theta_1)/2$ can be obtained as indicated in **Fig. 1** (**c**).

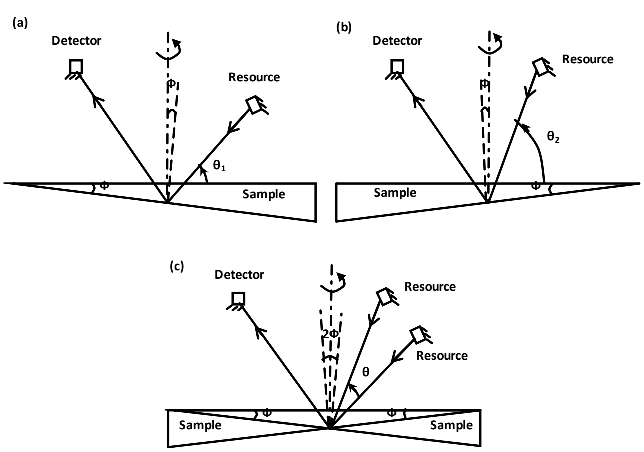
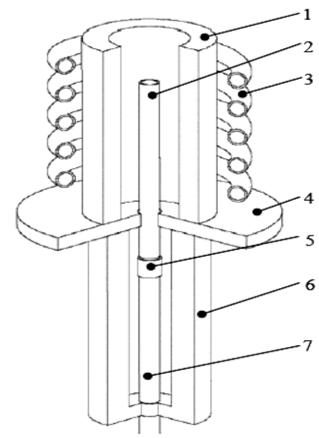


Fig. 1. Principle diagram of the Rotating directional test.

Methods and apparatus

The Al-40wt. %Cu alloy was prepared using the 99.99% purity Aluminum and Copper, and the 4 mm diameter rods with the length of 100 mm were cut from the ingot by the electrical discharge machining. The machined sample composition was determined by the chemical analysis method. Directional solidification experiments were carried out on a vertical Bridgman furnace at a thermal gradient of 250 K/cm described elsewhere [**15**]. The schematic diagram of the directionally solidified apparatus is shown in **Fig. 2**.

In directional solidification, the samples were firstly heated up to 850 ºC and kept this temperature for 20 min, and then pulled down at constant pulling rates. After the pulling distance attained 50 mm, the sample was rapidly quenched into a liquid Ga-In-Sn pool in order to obtain the solid/liquid interface shape.



1. Graphite heater, 2. Sample, 3. Inductor coil, 4. Heat insulating piece, 5. Connector, 6. Coolant container, 7. Pulling rod.

Fig. 2. Schematic diagram of directionally solidified experiment.

Directionally solidified samples were polished and etched using solvent Kroll with the agent of 1 ml HF, 3 ml HNO_3, and 46 ml H_2O. A Lecia DM4000M optic microscope was employed for metallographic examination and a Rigaku's D/max2400 X-ray diffractometer was used to measure the orientation of the directionally sample by RO-XRD.

Results and discussion

Microstructure

The part of Al-Cu binary phase diagram is shown in **Fig. 3**, and the experimental alloy composition of Al-40 wt. %Cu is marked by the black dotted line.

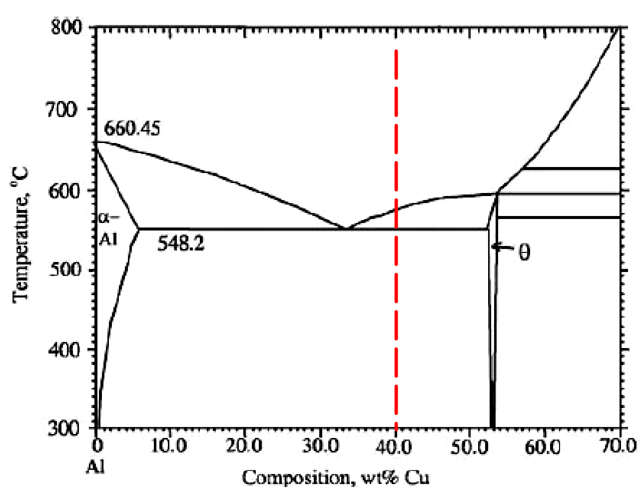


Fig. 3. Phase diagram of Al-Cu alloy [**16**].

The solidified microstructures of the Al-40% Cu consist of α-Al and θ-Al_2Cu phases based on the Al-Cu phase diagram. Due to the Al-40% Cu alloy being a hypereutectic alloy, the θ-Al_2Cu phase will be precipitated as the primary phase from the melt, and then the remaining liquid develops in the form of the coupled eutectic (Al/Al_2Cu). **Fig. 4** shows the directionally solidified microstructures of the Al-40% Cu at the growth rate of 10 μm/s. It implied that the grey parts were the primary Al_2Cu phase, since the size of primary Al_2Cu phase was larger than that of the coupled eutectic. Thus, the primary Al_2Cu phase and the eutectic (Al/Al_2Cu) can be clearly distinguished in the solidified structure.

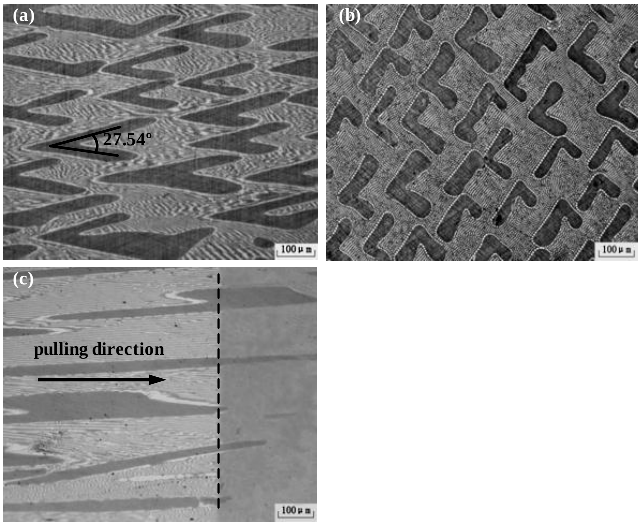


Fig. 4. Directionally solidified microstructures of Al-40wt. %Cu hypereutectic alloy at the solidification rate of 10 μm/s: (a) longitudinal section; (b) transverse section; (c) appearance of solid/liquid interface.

In addition, the growth pattern of the primary Al_2Cu phase was observed as the regular "V" shape in longitudinal section shown in **Fig. 4** (**a**) and (**b**), its morphology appeared as the "L" shapes in transverse section [**17**]. From **Fig. 4** (**c**) where the black dashed line indicated the solid/liquid interface, the grey primary Al_2Cu phase grew along with the pulling direction, and led the growth of the eutectic, because the interface temperature of the primary Al_2Cu phase was slightly larger than that of the coupled eutectic. To get the preferred growth direction or oriented crystal plane of the Al_2Cu phase in **Fig. 4**, the XRD diffraction measurement was employed. The experimental results are shown in **Fig. 5**.

Oriented crystal planes in the Al_2Cu phase

As indicated in **Fig. 5**, various crystal planes of the Al_2Cu phase, such as (200), (211), (112), (202), (222) as well as (420) can occur at normal solidification conditions. However, in directional solidification, only two crystal planes (110) and (310) appeared, and the crystal plane (220) can be attributed to the crystal plane family (110). Hence, the various crystal planes (200), (211), (112), (202) and (420) were overgrowth and only the crystal planes (110) and (310) were preferred planes during directional solidification. Considering in directional solidification, the heat flux direction can be well controlled, the growth of some no-preferred crystal planes may be inhibited and preferred crystal plane growth is promoted. Because only two crystal planes (110) and (310) appeared in the experiment, they were selected to do further analysis.

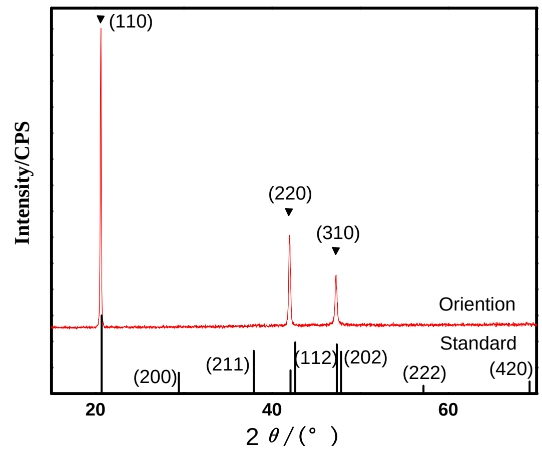


Fig. 5. X-ray diffraction patterns for Al-40wt. %Cu alloy.

The diffraction intensity of the crystal plane (110) in **Fig. 5** is the strongest, indicating that the primary Al_2Cu phase grows along this crystal plane as shown in **Fig. 4**, since it occupies the maximum volume fraction of the solidified microstructure.

Fig. 6 shows the butterfly diagram of symmetrical Gaussian envelope lines for the crystal plane (110), in which the RO-XRD method was employed by fixing the diffraction angle 2θ. It can be seen that two pairs of symmetrical Gaussian peaks take place, which shows the crystal plane (110) not well-aligned with one direction. Consequently, from **Fig. 6**, the deviation angle Φ between the crystal plane and the sample macro-surface varying from 7.24 º to 11.43 º can be measured by using the equation $\Phi = (\theta_2 - \theta_1)/2$.

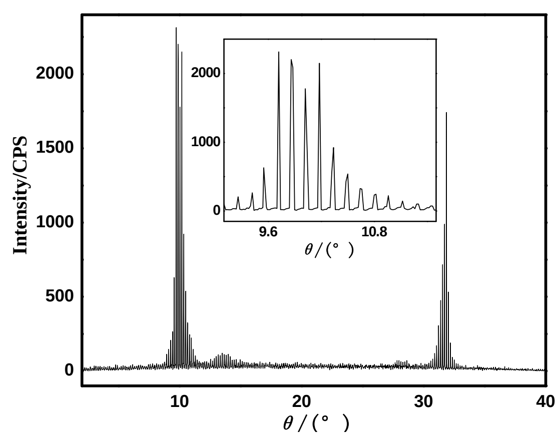


Fig. 6. RO-XRD patterns of (110) plane of Al-40wt. %Cu alloy.

In addition, the Gaussian peaks near the angle 10 º are magnified as shown in the upper part of the **Fig. 6**. Many small diffraction peaks within the angle from 9 º to 11.5 º were developed, meaning the growth direction fluctuation due to the changes of local alloy concentration and temperature field.

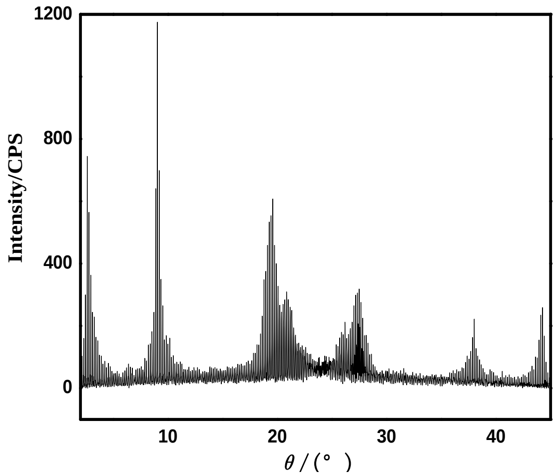


Fig. 7. RO-XRD patterns of (310) plane of Al-40wt. %Cu alloy.

For the crystal plane (310) in the Al_2Cu phase, when the detector fixed at 2θ= 47.26 º, the corresponding butterfly diagram of **Fig. 7** can be obtained. For the same reason, the growth direction of the Al_2Cu phase along the crystal plane (310) had a large dispersion and the deviation angle Φ changed from 2.68 º to 20.82 º.

Since Al_2Cu phase belongs to the tetragonal crystal system, the **Eq. 1** [**18**] is used to calculate the angle θ between crystal planes, in which $(h_1k_1l_1)$ and $(h_2k_2l_2)$ are the indices of crystallographic planes, and a, c are the lattice parameters. For the Al_2Cu phase, a= 0.6066 nm, c= 0.4874 nm. Introducing these data into the formula, the angle between the crystal planes (110) and (310) can be calculated to be 26.46 º. And in the experiment, the inclined angle of "V" shaped Al_2Cu phase was also measured in **Fig. 4** and found this value of 27.54 º close to 26.46 º, in agreement with the analysis of the XRD results. It also implies that the growth directions of these two planes are not well aligned with the heat flux direction.

$$\theta = \cos^{-1}\left[\frac{\frac{h_1h_2+k_1k_2}{a^2}+\frac{l_1l_2}{c^2}}{\sqrt{\frac{h_1^2+k_1^2}{a^2}+\frac{l_1^2}{c^2}}\sqrt{\frac{h_2^2+k_2^2}{a^2}+\frac{l_2^2}{c^2}}}\right] \quad (1)$$

Besides, the previously reported results indicated that the diffraction peaks of the intermetallic Al_2Cu phase directionally solidified were the crystal plane (110) and (310) [**9,19,20**]. The crystal plane (110) was the rapid growth crystallographic direction of tetragonal crystal system [**21**]. But under the influence of processing parameters, such as the growth rate and temperature gradient in directional solidification, the non-oriented crystal planes (310), (211), (112), (202) [**21**] may be developed. Nevertheless, the previously reported results do not include the deviation angle and dispersion degree, which are important to deeply understand the microstructure formation and morphology evolution in directional solidification of Al-40%Cu hypereutectic alloy.

Conclusion

1. During directional solidification of Al-40%Cu alloy, the preferred planes (110) and (310) of the Al_2Cu phase appear at 10 μm/s and the growth directions of these two planes are not well aligned with the heat flux direction.
2. The growth direction of the preferred plane (110) has an angle of 7.24 º~11.43 º with the heat flux direction and the deviation angle attains 2.68 º~20.82 º for the direction of the plane (310).
3. The RO-XRD method is an effective method for measuring the orientation and deviation angle of the intermetallic Al_2Cu phase in directional solidification of Al-40%Cu hypereutectic alloy.

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