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Nucleation interface of Al−Sb alloys on single crystal Al_2O_3 substrate

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Abstract: Lattice structure information of heterogeneous nucleation at nucleation interface was present. The crystal orientation, and interfacial structure characteristic of liquid Al alloys nucleated on the basal surface (0001) Al_2O_3 single crystal substrate were identified by X-ray diffraction (XRD), scanning electron microscopy (SEM) and high resolution transmission electron microscopy (HRTEM) analysis. The preferred crystal orientations of pure Al and Al−1%Sb (mass fraction) alloy adjacent to the nucleation interface were examined as (200) and (220) planes of Al, respectively, and two corresponding orientation relationships were obtained. An improved nucleation efficiency and refined grains were attributed to both the reduced interplanar spacing of preferred orientation and the decrease of lattice misfit from 16.4% to 7.0% in Al−1%Sb/Al_2O_3 nucleation group.
Key words: aluminium alloy; nucleation; interfacial structure; orientation relationship

1 Introduction

Since the first undercooling measurement executed by FAHRENHEIT [1] on the solidification of supercooled water providing the evidence for nucleation barrier, the nucleation of liquid towards a more condensed state attracts extensive scientific interests and technological attentions due to the intimate relationship with initial structure, the size scale of the structure and spatial distribution [2]. It is common practice to introduce nucleating agents during the casting process in order to reduce cast defects, form fine and uniform grains and therefore improve casting quality.

The investigation of ordering phenomena at solid−liquid interface has been carried out by various theoretical and experimental approaches [3−6] from the last century. High resolution transmission electron microscopy (HRTEM) [7,8] enables direct imaging of various interfaces at the atomistic level. For an extensive review on the epitaxy growth of general film in Ref. [9], the experimental evidence of heterostructures was presented across various misfit scales. A good lattice matching at the interface and a small undercooling represent a potent nucleation potency of the nucleating substrate in Al/Al_2O_3 system [10]. While in aspect of grain refining, apart from nucleating agents such as Al−Ti−B in Al alloy and Al−Ti−C in Mg alloy [11−13], trace alloying element, RE, K, Na, Ca, Sr, Ba, Sn, Sb, Bi, P is also added into alloys as microstructure modifier [14−19]. In these elements, Sb is a surface-active element extensively used in Al−Si alloys, Mg−Al−Si alloys and Al-based composites [20−24], to modify eutectic structure forming at relatively high cooling rates for enhanced casting properties. There are a few works on the modification mechanism of Sb to Si, Mg_2Si phases and so on. For example, REN et al [20] presented that the nucleation site of Mg_2Si in Mg−Al−Si alloy, is enriched in Si, Mg and Sb. WANG et al [22] suggested that the coherent precipitation of AlSb can introduce stress into Si crystal forming little defects, which hinders the growth of Si. They further pointed out that Sb and Ba will enhance the modification effect of RE. To date, different modification mechanisms of Sb in Al−Si and Al−Mg−Si alloys have been achieved. However, there is very little information regarding the interface structure formed during both nucleation and solidification processes of common metal casting on the specific

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substrates, such as the usually existed oxide Al_2O_3 in Al alloy. Correspondingly, the nucleation efficiency and refinement effect of Sb element on the heterogeneous nucleation of Al hasn't been investigated thoroughly yet.

This work aims at providing specific experimental results on nucleation behaviour of liquid Al−Sb alloy on a single crystal sapphire substrate and interfacial structure inspired by the nucleation is also investigated. The identity of nucleation interfacial structure is verified by multi-analytical technologies. Following that, the orientation relationships between the nucleation phase and substrate are experimentally determined through HRTEM. The enhanced nucleation efficiency and refining effect of Al−Sb alloy is hereupon evaluated from the crystallographic point of view, using a modified lattice matching model [25].

2 Experimental

A sapphire with basal surface (0001) was used for the nucleation substrate in this work. The surface roughness of the substrate was less than 5 nm. High purity Al (99.999%, mass fraction) was purified using glass fluxing method to remove potential heterogeneous nuclei from liquid Al. To examine alloying element effect on the nucleation behavior of liquid Al and the structure configuration of nucleation interface, Sb (99.999%, mass fraction) was added into liquid Al after purification. The mass fraction of 1% was selected to ensure primary α-Al phase nucleated under an uninterfered environment, to avoid any interfere from the potential compounds, according to Al−Sb phase diagram [26]. Al−1%Sb (mass fraction) alloy was prepared by arc melting under an argon atmosphere, and then directly cast into cylindrical rods with 3 mm in diameter using a suction casting facility.

The Al_2O_3 substrate was firstly cleaned in acetone for 3 min with an ultrasonic cleanser and then placed on a gas cooling platform in a high vacuum chamber (the pressure was 2×10^{-4} Pa). Aluminum and its alloy samples were placed on such an Al_2O_3 substrate and then Al/Al_2O_3, $Al−1\%Sb/Al_2O_3$ couples were heated up to 1300 K (1027 °C) by a laser beam with a heating rate of 20 °C/s. The sample was held at that temperature for 3 min before the laser beam was switched off, and then cooled down at a controlled cooling rate of 20 °C/s under a flowing argon atmosphere. The details can be referred to Ref. [27]. X-ray diffraction (XRD) analysis was employed to detect the crystal orientation of newly formed crystal from the bottom of the sample where nucleation was triggered by the substrate. Scanning electron microscopy (SEM) samples were obtained through conventional metallurgical sample preparation procedure for microstructural analysis. SEM was executed with backscattered electron (BSE) mode by Phenom XL System coupled with energy dispersive X-ray spectroscopy (EDS) analysis. Interfacial structure investigations were carried out by transmission electron microscopy (TEM) and HRTEM using a Tecnai G^2 F20 S-twin TEM instrument. The samples were prepared from cutting slices perpendicular to the interface with a thickness less than 80 μm before ion beam thinned using a Gatan PIPS II precision ion polishing system at 5.0 kV and an incident angle of 4°−6°.

3 Results and discussion

3.1 Crystal orientation of nucleated phase

The crystal orientation of Al alloy adjacent to the nucleation interface was examined on $Al/Al_2O_3(0001)$ and $Al−1\%Sb/Al_2O_3(0001)$ systems using XRD analysis, as shown in Fig. 1. The result shows that the preferred crystal orientation of pure Al was (200). When Sb was added into Al liquid, the preferred crystal orientation of newly formed crystals was changed into (220). Besides the crystals of Al matrix, the AlSb compound was also detected with the (111), (200), (220) and (311) diffraction peaks. It is obvious that the crystal orientation of new crystals nucleated on the (0001) Al_2O_3 substrate was affected by the addition of Sb.

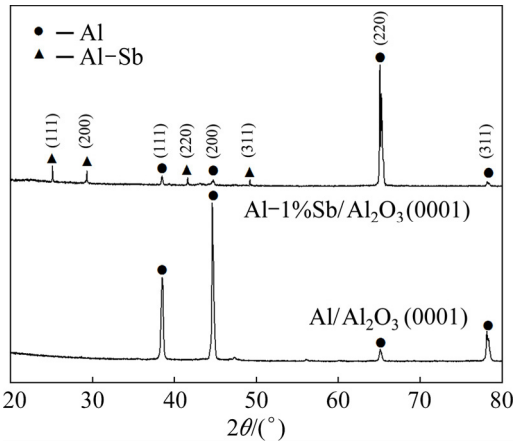

Fig. 1 Crystal orientation of newly formed crystals adjacent to interface in $Al/Al_2O_3(0001)$ and $Al−1\%Sb/Al_2O_3(0001)$ systems

3.2 Interface characteristics

SEM images at crystal−substrate interfaces of $Al/Al_2O_3(0001)$ and $Al−1\%Sb/Al_2O_3(0001)$ systems are presented in Fig. 2. It is seen that both the interfaces between crystal phases and Al_2O_3 substrate were straight and distinct. Two phases appeared in the Al−1%Sb crystals in Fig. 2(b) compared with only one phase in counterpart $Al/Al_2O_3(0001)$ system in Fig. 2(a). The darker and brighter phases marked as "1" and "2" in Fig. 2(b), were confirmed as matrix Al and Al−Sb