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Crystallographic study of grain refinement in aluminum alloys using the edge-to-edge matching model

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Abstract

The edge-to-edge matching model for describing the interfacial crystallographic characteristics between two phases that are related by reproducible orientation relationships has been applied to the typical grain refiners in aluminum alloys. Excellent atomic matching between Al_3Ti nucleating substrates, known to be effective nucleation sites for primary Al, and the Al matrix in both close packed directions and close packed planes containing these directions have been identified. The crystallographic features of the grain refiner and the Al matrix are very consistent with the edge-to-edge matching model. For three other typical grain refiners for Al alloys, TiC (when $a = 0.4328$ nm), TiB_2 and AlB_2, the matching only occurs between the close packed directions in both phases and between the second close packed plane of the Al matrix and the second close packed plane of the refiners. According to the model, it is predicted that Al_3Ti is a more powerful nucleating substrate for Al alloy than TiC, TiB_2 and AlB_2. This agrees with the previous experimental results. The present work shows that the edge-to-edge matching model has the potential to be a powerful tool in discovering new and more powerful grain refiners for Al alloys.

Keywords: Aluminum alloys; Grain refinement; Crystallography; Edge-to-edge matching

1. Introduction

Grain refinement in aluminum alloys has been studied for half a century and is aimed at improving the soundness of cast alloys. Although many new techniques, such as the electromagnetic vibrational method [1] and rapid solidification [2–4], have been developed, adding grain refiners as nucleant (or inoculant) is still the most common method for refining the grain structure of aluminum alloys during the solidification processes. The most commonly used commercial master alloys are based on the Al–Ti–B system, as originally proposed by Cibula [5]. For wrought alloys, the most commonly used master alloy is Al–5wt%Ti–1wt%B, which contains TiB_2 and Al_3Ti particles. The Al_3Ti particles dissolve rapidly upon dilution into wrought alloys, providing Ti solute, which is very effective at providing for constitutional undercooling that decreases the growth rate of grains and facilitates further nucleation. For a considerable amount of time it was assumed that Al_3Ti particles must be the heterogeneous nucleation sites as they are a pro-peritectic phase, which it was assumed could nucleate the solid through a pro-peritectic reaction and furthermore had very good lattice matching with Al. It is now generally considered that TiB_2 particles are the nucleation sites as they are more stable in aluminum melts [6–8], although there is still some debate about whether these particles require a coating of Al_3Ti to be active [9,10]. More recently, grain refiners with lower ratios of Ti:B have been developed for use in Al–Si foundry alloys. These master alloys

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contain AlB_2 and (Ti, Al)B_2 particles. It is understood that these master alloys are superior to the Al–5Ti–1B type grain refiners because the AlB_2 and (Ti, Al)B_2 particles are 'more potent' nucleant particles than TiB_2 and Al–Si foundry alloys and do not require the excess Ti for constitutional undercooling due to the high level of Si. Another group of commercial grain refiners are based on the Al–Ti–C system [11], a system also originally proposed by Cibula [12]. In these grain refiners TiC particles are thought to be the nucleating phase and these grain refiners can be used in situations where elements that poison the grain refinement by Ti:B grain refiners are present, e.g. Zr, Cr or Li [13,14].

One of the important issues to consider in understanding grain refinement is the potency of the substrate. The potency of a substrate is related to the free energy difference required to drive the phase transformation. Classical heterogeneous nucleation relates this to the wetting angle between the substrate and the nucleating solid [15]. However, more recently it has been realized that the wetting angle is so low during typical grain refinement processes that this model is not a useful description of heterogeneous nucleation during grain refinement. Alternatives such as the absorption model of nucleation [16] have been proposed as improved descriptions of nucleation during heterogeneous nucleation. Regardless of which model accurately describes the process of heterogeneous nucleation, it is important to note that each of these models consider that the interfacial energy between the substrate and the solid is critical to the potency of a substrate. The energy on the newly formed interface must be lower than the surface energy of the same area of the interface if it formed directly in liquid metal. For a low interfacial energy, the two solids have to be coherent or partially coherent, which leads to a certain orientation relationship (OR) between these two phases. In a coherent or partially coherent interface, strain energy is present if there is not exact matching between the substrate and the solid. In order to minimize this strain energy, it requires maximum atom matching on the interface.

There has been some research that has investigated the lattice matching of the grain refiner particles with aluminum, particularly for the Al_3Ti phase, which has a tetragonal structure. The ORs published by Davies [17] and by Arnberg [18] are quite different. Davies only reported some plane parallelisms between the two crystals. This can not really be called an OR. Arnberg [18] reported the following ORs between Al_3Ti and Al. They are as follows.
The A–B–K OR1

$$(0\ 1\ 1)_{Al_3Ti} \parallel (0\ 1\ 2)_{Al}, \quad [0\ 1\ 0]_{Al_3Ti} \parallel [0\ 1\ 0]_{Al}$$

The A–B–K OR2

$$(0\ 0\ 1)_{Al_3Ti} \parallel (0\ 1\ 0)_{Al}, \quad [0\ 1\ 0]_{Al_3Ti} \parallel [0\ 1\ 0]_{Al}$$

In addition, Kobayashi et al. [19] observed another OR in Al–Ti alloy using electron diffraction analysis.
The K–H–S OR

$$(1\ 1\ \bar{2})_{Al_3Ti} \parallel (1\ \bar{1}\ \bar{1})_{Al}, \quad [\bar{1}\ 1\ 0]_{Al_3Ti} \parallel [0\ 1\ \bar{1}]_{Al}$$

This was considered to have low lattice disregistry between the two lattices. The lattice matching between the borides and aluminum has also been considered previously. TiB_2, AlB_2 or (Ti, Al)B_2 are isomorphous compounds with a hexagonal structure. As boride particles were found at the center of the Al grains [20,21], they have been considered as active heterogeneous nuclei. In the 1970s, Naess [20] reported a relationship between TiB_2 and Al matrix as $(3\ 1\ 1)_{Al} \parallel (20\ \bar{2}\ 1)_{TiB_2}$, and Marcantonio [22] found an OR between AlB_2 and Al.
The Marcantonio OR

$$(1\ 1\ 1)_{Al} \parallel (0\ 0\ 0\ 1)_{AlB_2}, \quad [\bar{1}\ 1\ 0]_{Al} \parallel [1\ 1\ \bar{2}\ 0]_{AlB_2}$$

Another nucleation substrate in Al alloys is the compound TiC with a face-centered cubic (FCC) structure. The only work on the crystallography of TiC and Al was undertaken by Cissé et al. [23]. They reported an OR as follows.
The C–B–K OR

$$(0\ 0\ 1)_{Al} \parallel (0\ 0\ 1)_{TiC}, \quad [0\ 1\ 0]_{Al} \parallel [0\ 1\ 0]_{TiC}$$

Throughout the previous research, people have realized the importance of the crystallography of the nucleation substrate and Al matrix in grain refinement. Normally it is considered that the lattice disregistry for TiB_2 and Al has been quoted as 5.9% at room temperature and this decreases as the temperature increases [24]. The lattice disregistry between AlB_2 and Al was determined to be 3.5% according to Tøndel [25], which is why AlB_2 particles are thought to be of greater potency than the TiB_2 particles. The (Al, Ti)B_2 particles are in between. It seems the lattice disregistry correlates well with observations. However, the concepts of either the lattice disregistry or lattice mismatch are very confused. Do they imply lattice structure similarity, or interplanar spacing mismatch between planes, or interatomic spacing mismatch along directions? In addition, because the lattice disregistry is calculated based on lattice parameters only, it actually represents the correlations between lattice points rather than atoms. For more complicated structures, the actual atoms that form the crystal and the interface between crystals have been ignored. To solve the problem, the present paper applies the recently developed edge-to-edge matching model [26–30] to examine the crystallographic features between the Al solid and the commonly used grain refiners, including Al_3Ti, TiB_2, AlB_2 and TiC, in order to understand, from a crystallographic point view, the mechanism of the grain refinement resulting from these compounds. The focus of this examination is to use the model to predict whether any ORs between the nucleating substrates and the Al matrix exist in a