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ScienceDirect
Acta Materialia 55 (2007) 5934–5947
Acta MATERIALIA
www.elsevier.com/locate/actamat

First-principles calculations of β″-Mg_5Si_6/α-Al interfaces

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Received 26 January 2007; received in revised form 11 June 2007; accepted 29 June 2007
Available online 29 August 2007

Abstract

The metastable β″-Mg_5Si_6 phase is often the most effective hardening precipitate in Al-rich Al–Mg–Si alloys. Two important factors that control the precipitate morphology are the strain energy and the interfacial energy between the precipitate and the matrix. By means of a first-principles supercell approach and density functional theory calculations, we have studied the interfacial properties between β″-Mg_5Si_6 and α-Al. We carefully construct a large number of interfacial cells in order to elucidate preferred interfacial terminations and orientations, as well as atom alignment and intermixing across the interface. Each of the low-energy interfaces we found possesses two key attributes: a high number of Al–Si bonds across the interface, and a face-centered cubic topological alignment of atoms across those interfaces. Our first-principles results yield quantitative values for the interfacial energies, lattice mismatches and strain energies that can be used in future predictions of precipitate morphologies as a function of size.

Keywords: β″-Mg_5Si_6/α-Al interface; Interfacial energy; Lattice mismatch; First-principles

1. Introduction

Precipitation hardening is utilized to strengthen a wide variety of alloy systems. An example is the class of commercially important Al–Mg–Si based alloys which are strengthened by a number of metastable precipitate phases [1–12], where the needle-shaped β″-Mg_5Si_6 precipitates are often the main contributor to hardening [11]. Beginning with the supersaturated solid solution (SSS), the generic precipitation sequence in Al–Mg–Si alloys is generally believed to be [12]:

$$\text{SSS} \rightarrow \text{Mg/Si clusters} \rightarrow \text{Guinier-Preston zones} \rightarrow \beta'' \rightarrow \beta' \rightarrow \beta$$

In practice, the sequence can be even more complex [1,3,10–14], and a number of other metastable phases, such as U2, U1 and B′, may also form along with β′, depending on alloy composition and the heat treatment time and temperature.

One of the key factors that control the mechanical properties of precipitate-hardened alloys is the precipitate morphology, i.e. the size and shape of precipitates. In order to predict [15,16] the precipitate microstructural evolution and thus mechanical properties, it is critical that the thermodynamic driving forces and kinetic mechanisms that lead to various precipitate shapes be understood.

The morphology of a precipitate is primarily determined by two competing energetic contributions, i.e. the interfacial energy between the precipitate and the matrix and the coherency elastic strain energy generated due to the lattice mismatch between the precipitate and the matrix. Obtaining these quantities directly from experiments can be difficult due to the metastable nature of many precipitates. For example, in many cases, including β″-Mg_5Si_6, only the constrained lattice parameters are available experimentally (e.g. from high-resolution transmission electron microscopy (TEM) or diffraction

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1359-6454/$30.00
doi:10.1016/j.actamat.2007.06.045

approaches [11,17]). But experimental data for stress-free lattice parameters (and hence the lattice mismatch), as well as elastic constants and interfacial energies, are not typically available. First-principles total energy and crystal structure calculations provide a computational tool capable of giving quantitative predictions for these hard-to-measure quantities.

The main objective of this paper is to search for low energy interfaces between the β″-Mg_5Si_6 precipitate and the α-Al matrix from a first-principles approach. Extensive calculations were performed to examine the effects of interfacial termination, atomic alignment and intermixing [11], and interfacial orientations [18,19]. Though interfacial energies have been previously calculated using first-principles calculations in other systems (see e.g. [20–23]), almost all of these previous calculations have been focused on systems where both phases are high-symmetry cubic phases, often with simple small-unit-cell crystal structures. In contrast, the precipitate/matrix interfaces studied in this work involve the relatively low-symmetry monoclinic β″-Mg_5Si_6 precipitate with a complex stoichiometry and crystal structure, and a high symmetry cubic α-Al matrix. As a result of the complexity of this system, there are a large number of degrees of freedom to consider in constructing the interfaces, including where to "cut" the crystals of the matrix and precipitate and how to "join" them – interfacial orientation, interfacial termination, atom alignment and atomic arrangement near the interface. In the following sections, the crystal structure of β″-Mg_5Si_6, its relationship with α-Al (face-centered cubic, fcc) and the supercell models for the β″-Mg_5Si_6/α-Al interface are described. We then give first-principles results for the interfacial energies and strain energies, as well as the stress-free mismatch in β″-Mg_5Si_6/α-Al. In addition, we investigate a recently proposed model for one of the β″-Mg_5Si_6/α-Al interfaces in which an intermixing tendency across this interface has been deduced from high-resolution electron microscopy (HREM) and electron diffraction (ED) measurements [11].

2. Crystal structures and interface models

2.1. Structural relationship between β″-Mg_5Si_6 and α-Al

Fig. 1 illustrates the structural similarity [11] between β″-Mg_5Si_6 and fcc α-Al by showing a 22-atom supercell of the α-Al fcc lattice in the form of a conventional monoclinic unit cell (CMUC) of β″-Mg_5Si_6. In this representation, the lattice vector $\mathbf{b}_{\beta''}$ ([010]) of the β″-Mg_5Si_6 is parallel to the [001] axis of α-Al ($\mathbf{b}_{\beta''} = \mathbf{c}_{Al}$) and the other two lattice vectors of β″-Mg_5Si_6 are defined by $\mathbf{a}_{\beta''} = 2\mathbf{a}_{Al} + 3\mathbf{b}_{Al}$ (i.e. the [230] direction in α-Al) and $\mathbf{c}_{\beta''} = -1.5\mathbf{a}_{Al} + 0.5\mathbf{b}_{Al}$ (i.e. the $[\bar{3}10]$ direction of α-Al). As noted in Ref. [11], even a precise one-to-one atom mapping between β″-Mg_5Si_6 and α-Al can be obtained if one shifts the corner Mg and $\mathbf{a}_{\beta''}/2$ Mg atoms by $\mathbf{b}_{\beta''}/2$.

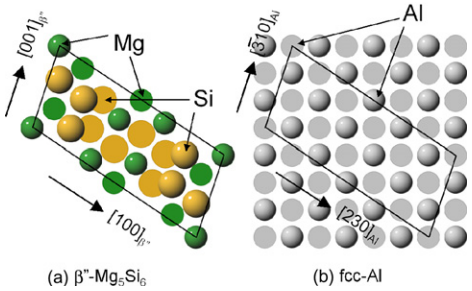

Fig. 1. The crystal structure of β″-Mg_5Si_6 (a) and its relation with α-Al (b). Mg: green (small balls); Si: orange (large balls); Al: gray. The solid balls show atoms at the paper surface and flat plates show atoms at $\mathbf{b}_{\beta''}/2$ (~0.2025 nm) into the paper surface. It can be seen that the one-to-one atom correspondence can be assigned between the two parallelograms, with only the Mg atom at the corner of the β″-Mg_5Si_6 parallelogram being shifted $\mathbf{b}_{\beta''}/2$ if being compared the α-Al parallelogram. Note that the Mg atom located at the $[100]_{\beta''}$ side of the β″-Mg_5Si_6 parallelogram is equivalent to corner Mg atom by translational symmetry. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2.2. Interfacial orientations

A reconstructed exit wave of a typical β″-Mg_5Si_6 needle in α-Al by Andersen et al. [11] showed the existence of two types of interfaces parallel to the needle axis $\mathbf{b}_{\beta''}$ (needle-∥), i.e. one parallel to the (100) crystal plane of β″-Mg_5Si_6 and another parallel to the (001) crystal plane of β″-Mg_5Si_6. Along with these two observed interfacial orientations, for completeness, we assume a third interface be the plane paralleling to the (010) crystal plane of β″-Mg_5Si_6 (needle-⊥). The interfacial orientation relations for these three types of interfaces are summarized as follows:

A: $(130)_{Al}\|(100)_{\beta''}$(needle-∥)

B: $(001)_{Al}\|(010)_{\beta''}$(needle-⊥)

C: $(\bar{3}20)_{Al}\|(001)_{\beta''}$(needle-∥)

where the labels A, B and C are used simply to reflect, respectively, the interfacial orientation direction of β″-Mg_5Si_6 for that interface.

3. Supercells

In our first-principles calculations, we adopt coherent models along all interfacial orientations considered. Table 1 summarizes the interfacial orientations, alignments, terminations and interfacial intermixing of all interfacial supercells considered in this paper.

The conventional monoclinic unit cell (CMUC) shown in Fig. 1 is used as the "building block": the interfacial supercells are built using multiple CMUC units, with the 22 atoms of each CMUC having stoichiometries Al_{22} or $Mg_{10}Si_{12}$ on either side of the interface. It should be noted that if one shifts the corner Mg atom of β″-Mg_5Si_6 (equivalent to the $\mathbf{a}_{\beta''}/2$ Mg atom) by $\mathbf{b}_{\beta''}/2$, the