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2012 IOP Conf. Ser.: Mater. Sci. Eng. 27 012032

(http://iopscience.iop.org/1757-899X/27/1/012032)

IOP Conf. Series: Materials Science and Engineering **27** (2011) 012032 doi:10.1088/1757-899X/27/1/012032

Crystal orientation and morphology in Al-Ag-Cu ternary eutectic

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Abstract. Ternary eutectics provide a unique opportunity for studying the effects of complex microstructure formation, as three distinct phases must be formed simultaneously from the melt. In order to produce fully coupled three-phase growth, Al-Ag-Cu at the ternary eutectic composition was directionally solidified in a constant temperature gradient of 3 K/mm at velocities between 0.2 and 5.0 μm/sec. Under these conditions, the two intermetallic phases appear to grow as closely coupled rods in an α(Al) matrix, with the solidification velocity affecting the specific morphologies chosen by the rods and the general degree of alignment of the structure. Crystal orientations were examined by EBSD to determine if variations in morphology within a single sample are due to specific orientation relationships. Although no conclusive connection to morphology has yet been found, two different sets of orientation relationships between the three phases have thus far been identified.

1. Introduction

As the performance and safety demands on engineering materials continue to rise, the use of multi-component, multi-phase alloys processed under carefully controlled conditions becomes ever more common. This is one of the reasons for the recent scientific interest, summarized by Hecht et. al. in [1], in understanding the behavior of these types of complex systems. One particular type of complex microstructure is that which occurs in ternary eutectic systems. While binary eutectics are generally well understood, ternary eutectics, by adding an extra degree of freedom, open up a far wider array of possible microstructures, beyond simple lamellar or rod formations, and make for an excellent platform for the study of complex microstructure formation.

Although the presence of ternary eutectic structures are reported fairly often in literature, systematic studies of them have been extremely limited. One of the few systems which has been the subject of limited studies is aluminum-silver-copper. The phase diagram in Figure 1 shows the three phases that form the single ternary eutectic point at 774 K: α(Al), θ (Al_2Cu), and ζ (Ag_2Al). This alloy serves as a nice model system because, unlike many systems, the eutectic structure has approximately equal volumes of each phase, allowing for true three-phase coupled growth. It also has three unfaceted phases [2] and physical properties that make it convenient for experimental work. The classic work on the Al-Ag-Cu system was carried out by Cooksey and Hellawell [3] and by McCartney et al. [4]. More recently, De Wilde et. al. [5, 6] has described several different morphologies resulting from both directional solidification and unconstrained growth. While the exact form of the microstructure varies depending on solidification conditions, in all cases an association between the Al_2Cu and the Ag_2Al phases is reported. De Wilde attributes this both to mass transport reasons and to the predicted presence of a low-energy boundary between the two intermetallic phases, although the crystallographic orientations of the three phases in this system have never been investigated. In this work, we attempt to characterize the morphologies favored by different solidification velocities and to identify the primary crystallographic orientations of the three phases and any common planes between them.

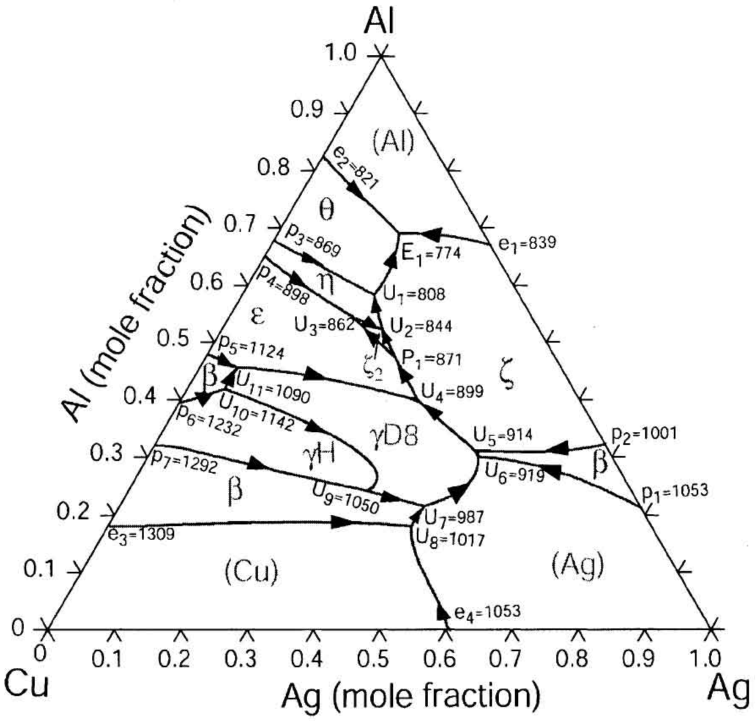

Figure 1. Liquidus surface of the ternary phase diagram showing the three-phase eutectic at E_1 [7].

2. Experimental procedure

The ternary eutectic composition for this system has the composition of Al-12.8Cu-18.1Ag (at%), as given by the phase diagram shown in Figure 1. High purity (99.999%) aluminum, silver and copper were used to prepare a melt with the ternary eutectic composition. From the initial cast ingot, cylindrical rods of 8 mm diameter and 105 mm length were cut and directionally solidified using the ARTEMIS furnace facility [8], which holds the molten sample inside a silica aerogel crucible. Aerogels exhibit extremely low levels of heat conduction, which allows for negligible radial heat loss and nearly planar isotherms during solidification. In addition, their transparency enables optical monitoring of the solidification front and precise determination of the solidification velocity. All samples were solidified with a thermal gradient of 3 K/mm and velocities between 0.2 and 4.0 $\mu m/s$.

After processing, the samples were cut and polished for examination in both the transverse and longitudinal planes. Energy dispersive x-ray spectroscopy (EDS) was used to determine overall composition and composition of individual phase regions. Electron backscatter diffraction (EBSD) was used to determine the crystallographic orientations of the phases on the longitudinal sections.

3. Results and Discussion

Under the range of solidification conditions tested, the samples displayed both primary dendritic phases and regions of two-phase eutectic. The type and volume fraction of dendrites was dependent on the precise composition of the alloy and the location within the sample. Figure 2 shows an example of a region with both large primary α(Al) dendrites (#1) and smaller two-phase α(Al)-Al_2Cu dendrites (#2). In all cases, however, large regions of fine three-phase eutectic away from the influence of the primary phases were available for examination.

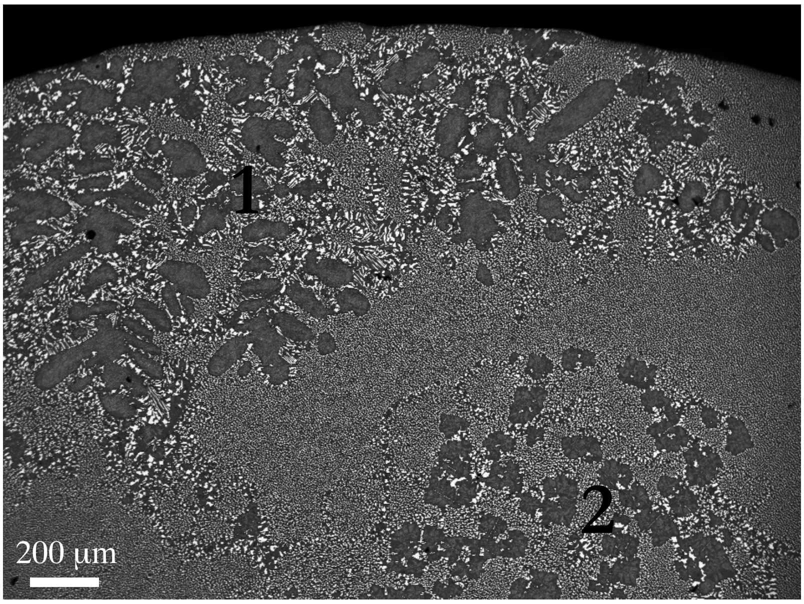

Figure 2. Overview of a sample showing primary α(A) dendrites (#1) and two-phase α(Al)-Al_2Cu dendrites (#2) surrounded by fine three-phase eutectic.

Figure 3 shows a detailed view of two of the different observed eutectic morphologies. These are transverse cross-sections, perpendicular to the growth direction. The image on the left shows the two intermetallic phases growing as rods with roughly rectangular cross-sections arranged in an alternating pattern to form 'ribbons' running through the structure, with each ribbon separated by regions of α(Al). This type of structure has been previously described as 'brick-like' [9] and 'chains with alternating intermetallic links' [4]. The alignment of the ribbons or 'chains' was observed to vary considerably, and generally increased with decreasing solidification velocity. The image on the right in Figure 3 shows a different morphology where many of the Ag_2Al regions are elongated in the direction of the ribbons, while the Al_2Cu regions remain as rods with very irregular cross-sections. A variety of methods for quantifying these structures and the effects of changing solidification velocity and thermal gradient are described in another recently submitted work.

While solidification velocity and temperature gradient have a measurable effect on the morphology of the eutectic structure, considerable variations in morphology within the same cross-section of a single sample were also observed, and it was hypothesized that these differences are the result of different crystal orientations. Investigation of the crystal orientations with EBSD has thus far revealed two distinct sets of orientation relationships, although a clear relationship between orientation and morphology has not yet been found.

For both crystal orientations, the growth directions of the three phases are always the same. The α(Al) (face centered cubic) and the Al_2Cu (tetragonal) both grow face-on with the [001] direction aligned to the growth direction. The hexagonal Ag_2Al grows edge-on with the $[11\bar{2}0]$ aligned with the growth direction, making the basal planes also parallel to the growth direction. Only a small degree of misalignment between these directions and the growth direction was

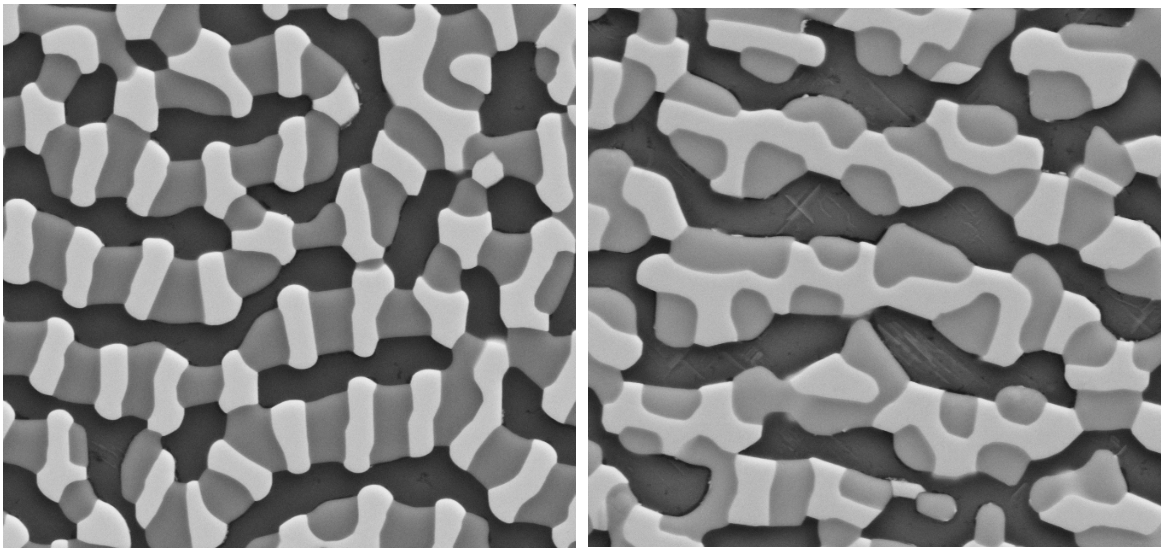

Figure 3. Eutectics morphologies found in two different grains of the same transverse cross-section solidified at 4.0μm/s. The white phase is Ag_2Al, the light gray phase is Al_2Cu, and the dark gray phase is α(Al). The crystal orientations of these specific grains are unknown.

observed. Additionally, different phase regions in a single grain showed a very high degree of alignment in all directions, with variations of less than 2°.

In the first orientation relationship, one of the {130} planes in the α(Al) is aligned with a {100} plane in the Al_2Cu phase. Additionally, a {110} plane of the Al_2Cu is aligned with a $\{10\bar{1}0\}$ (prismatic) face of the Ag_2Al. An example of the pole figures illustrating this relationship is shown in Figure 4, and a schematic showing the alignment of the unit cells is given in Figure 6. The growth direction (z) is at the origin of these pole figures, so the location of the spots for the common planes along the outer edge of the figures indicates that the common planes are aligned parallel or nearly parallel to the growth direction, a necessity for continuous fibers such as those observed in longitudinal cross-sections of these samples. The alignment of the $\{110\}^{Al_2Cu}$ and $\{10\bar{1}0\}^{Ag_2Al}$ planes with the microstructure is reasonably good. The average difference between the direction indicated by the pole figures and the angle perpendicular to the dominant interface between the two intermetallics is about 11°, which is within the range possible for an irrational interface.

In the second orientation relationship, the {130} plane in the α(Al) is again aligned with a {100} plane in the Al_2Cu phase. However, in this case, a {140} plane of the Al_2Cu is now found to be aligned with the {0001} (basal) plane of the Ag_2Al (see Figures 5 and 6). Although this second set of planes is again aligned correctly (that is, vertically), the angle between the pole figures and the dominant interfacial alignment in the microstructure appears to be greater than 30°, indicating that this may not be a true common plane. However, it can still be used to identify this particular orientation relationship until the true common plane, if one exists, is found. In neither this orientation relationship or the first has a common plane between the α(Al) and the Ag_2Al been identified.

In previous work with this system done on alloy compositions along the α(Al)-Al_2Cu univariant grove, two orientation relationships between $\alpha(Al)$ and Al_2Cu were observed: $\{130\}^{\alpha(Al)}//\{100\}^{Al_2Cu}$ and $\{111\}^{\alpha(Al)}//\{2\bar{1}1\}^{Al_2Cu}$ [10]. In the binary eutectic, the latter is more common, but in the ternary eutectic only the former has thus far ever been observed. It appears that the presence of the Ag_2Al phase stabilizes that particular orientation relationship.

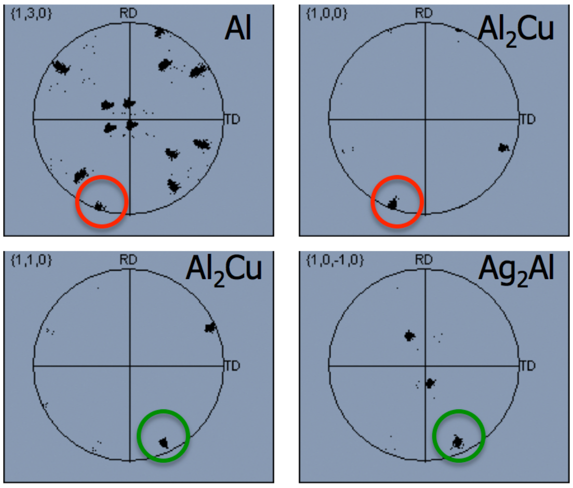

Figure 4. Pole figures showing the common planes for the first orientation relationship.

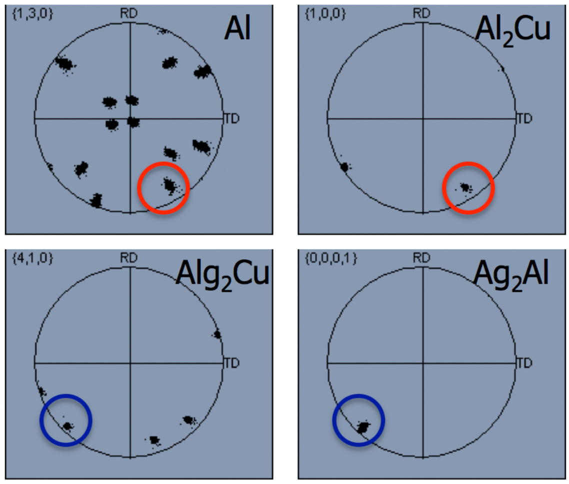

Figure 5. Pole figures showing the common planes for the second orientation relationship.

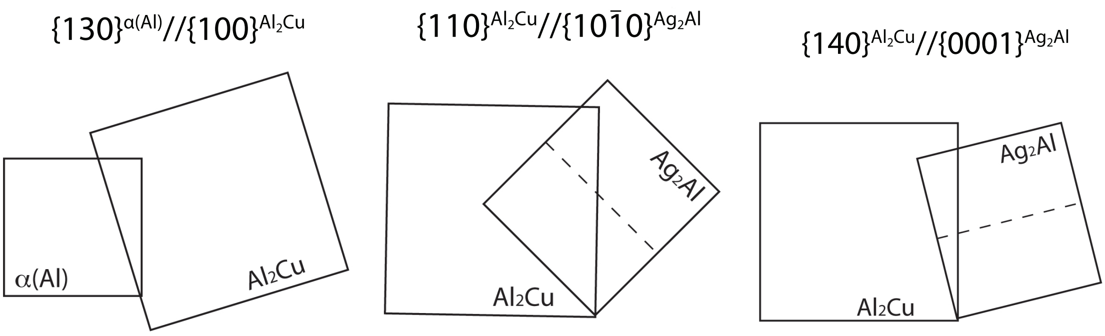

Figure 6. Relative positions of the unit cells for each of the three observed orientation relationships, when looking along the growth axis. Dotted line indicates an edge in the hexagonal cell, perpendicular to the basal plane.

More work is necessary to understand the mechanism behind it.

The work carried out to date provides only a preliminary understanding of the role of crystal orientation in Al-Ag-Cu ternary eutectics. More data from a larger number of eutectic grains is necessary to determine if other orientation relationships appear, and what correlation, if any, exists been orientation relationship and morphology. The Ag_2Al phase sometimes shows preferred interface planes when sectioned in the transverse direction, and identification of these planes by EBSD is an interesting problem. Polishing of the sample so that all three phases produce a diffraction pattern is a difficult task, especially while maintaining all the phases at the approximately the same height. This is a challenge that must be overcome before the necessary data can be reliably collected. Ion milling has shown promise as a method and, if optimized, may prove useful for three-dimensional imaging of these samples as well.

4. Conclusions

The formation of the ternary eutectic structure in the Al-Ag-Cu system is a complex process that depends on both solidification conditions and crystal orientation. Using EBSD, two different orientation relationships have been identified. In both cases, the growth direction of all three phases is fixed, along with the orientation relationship between α(Al) and Al_2Cu, which has $\{130\}^{\alpha(Al)}$ and$\{100\}^{Al_2Cu}$ as the common plane. Unlike in the α(Al)-Al_2Cu binary system, this is the only orientation relationship observed between these two phases. Two different orientation relationships have been observed between Al_2Cu and Ag_2Al, and in one case a common plane has been identified with matches well with the interfaces observed in the microstructure. No common planes have been identified between α(Al) and Ag_2Al. Work is ongoing to obtain a better understanding of these complex microstructures and the driving forces behind them.

Acknowledgements
The authors wish to acknowledge the invaluable help of Ulrike Hecht and Matthias Kolbe in obtaining and analyzing the crystallography data.

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