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Microstructural characteristics of splat-quenched aluminium-copper alloys

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The microstructural characteristics of a range of splat-quenched aluminium–copper alloys are described. The variation of the microstructure as a function of thickness and composition has been determined, by use of conventional 100 kV and high-voltage electron microscopy, and also by ion-beam thinning techniques. With increasing thickness alloys containing < 13 at. % Cu undergo the coventional aluminium–copper precipitation sequence, but alloys of > 13 at. % Cu exhibit a range of degenerate, radial and parallel eutectic structures. Restriction of examination to standard areas, undergoing similar cooling paths, has enabled valid comparison to be made between specimens of different composition.

1. Introduction

The techniques of splat-quenching metals and alloys directly from the liquid state to room temperature, or below, are well established [1]. The rapid cooling rates ($> 10^{6}\,^{\circ}$C sec^{-1}), frequently result in non-equilibrium metastable phase formation, and a comprehensive review [2] of the wide range of materials investigated has recently been published. The aluminium–copper alloy system, in particular the eutectic (17.3 at.% Cu) composition, has been the subject of several investigations, (e.g. [3–6]), using both X-ray and electron microscope techniques. Nevertheless, there is still considerable uncertainty concerning the microstructural characteristics of these alloys, particularly the distribution of phases within the specimen, and their relationship to the local cooling rate. Such uncertainty is common in many splat-quenched alloys, due to the wide point-to-point variation in cooling conditions within the splat. This paper reports a transmission electron microscope study of the microstructural characteristics of splat-quenched aluminium and six alloys containing up to 17.3 at.% copper. The work has emphasized the influence of composition at approximately constant cooling rate [7]. In addition, the effect of thickness at constant composition has been characterized, using both high-voltage electron microscopy and ion-beam thinning of splatted material.

2. Experimental

Pure (99.999%) aluminium and copper were used to prepare six alloys containing 2, 5, 7, 13, 15 and 17.3 at.% Cu. Casting and homogenization were carried out under high-purity argon. Specimens (100 mg) were splat-quenched from 850° C onto a water-cooled substrate using a modified Duwez gun system, which has been described in detail elsewhere [8]. Prior to operation, the system was evacuated to 2×10^{-5} Torr and flushed with high-purity argon. Quenches were performed under reduced pressure (400 Torr) of high purity argon using a 5 MN m^{-2} blast of the same gas. Specimens obtained were extremely porous, adhering closely to the substrate. Small fragments were removed using a fresh scalpel blade, and observed in a Philips E.M. 301 or A.E.I. EM 7, electron micro-

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scope, either directly or after subsequent ion-beam thinning.

3. Results and discussion

Specimens studied were invariably crystalline, and the effects of composition and specimen thickness on the microstructure will be treated separately. (Amorphous regions were observed, but as reported previously [9], these are either artefacts introduced during splatting or a consequence of specimen oxidation.)

3.1. Variation of microstructure with composition

The as-splatted specimens had many crystalline areas transparent to 100 kV electrons. Such a region, with an overall morphology typical of every composition, is shown in Fig. 1. The specimen has a columnar grain structure as has been observed occasionally before [6]. A characteristic narrow band of increased thickness at the edge gives rise to an increase in the projected width of the grain boundaries at A. The regions immediately adjacent to the thick edge were taken as having approximately constant cooling rate and were therefore used as standard regions for comparison of microstructure from specimen to specimen [7]. The occurrence of the columnar grain structure in Fig. 1 is consistent with heat transfer in splat-cooling occurring parallel to the specimen/substrate interface [10] in contrast to the perpendicular direction often assumed in theoretical treatments [11, 12] of splat-cooling kinetics.

Pure aluminium specimens contained a mixture of Frank and prismatic dislocation loops (see Fig. 2). In contrast, there were no dislocation loops in the alloy specimens. The presence of Frank loops

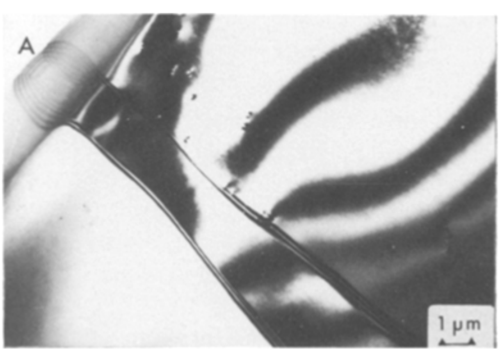

Figure 1 As-splatted specimen of Al–5 at.% Cu showing the overall morphology typical of electron transparent regions.

Figure 2 As-splatted specimen of pure aluminium containing a high density of Frank and prismatic dislocation loops.

implies that there are no significant quenching stresses in these areas, since the resultant dislocations would produce prismatic loops alone.

Using the thin standard regions for comparison of microstructures, it has been found that all alloys, up to and including the eutectic composition, were apparently single phase. These single phase regions were devoid of fine scale structure in the image and ⟨1 0 0⟩ streaks in diffraction patterns. Occasionally in alloys containing ~ 5 to 12 at.% Cu, non-standard cooling conditions were reflected in the presence of grain-boundary precipitates (see Fig. 3a). Similarly above ~ 13 at.% Cu a degenerate eutectic microstructure was occasionally observed in such areas (see Fig. 3b). It is assumed that such regions have undergone a slower cooling rate than standard regions. This may arise either from "lift-off" of the edge of the specimen from the substrate, or from re-heating of the thin regions as a result of subsequent droplets solidifying in their vicinity.

3.2. Variation of thickness

The effect of thickness was studied in three ways, namely, progressing into thicker regions from the

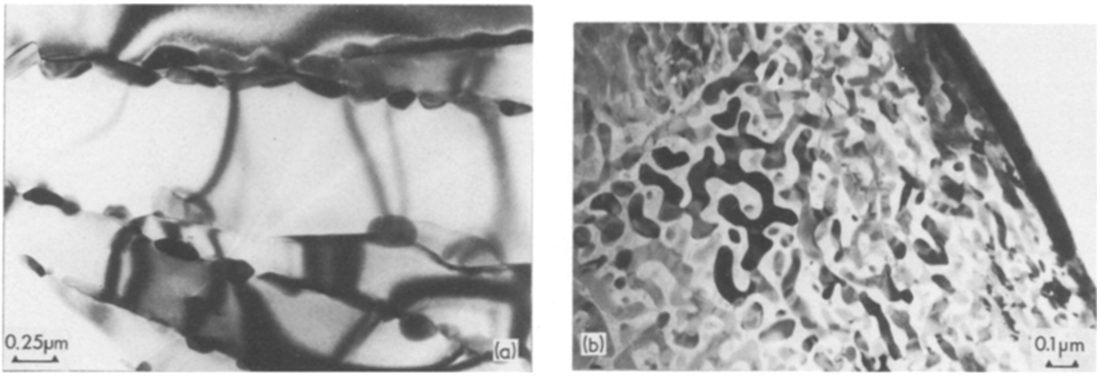

Figure 3(a) Grain-boundary precipitates in splat-quenched Al–7 at.% Cu. (b) Degenerate eutectic structure in splat-quenched Al–17.3 at.% Cu.

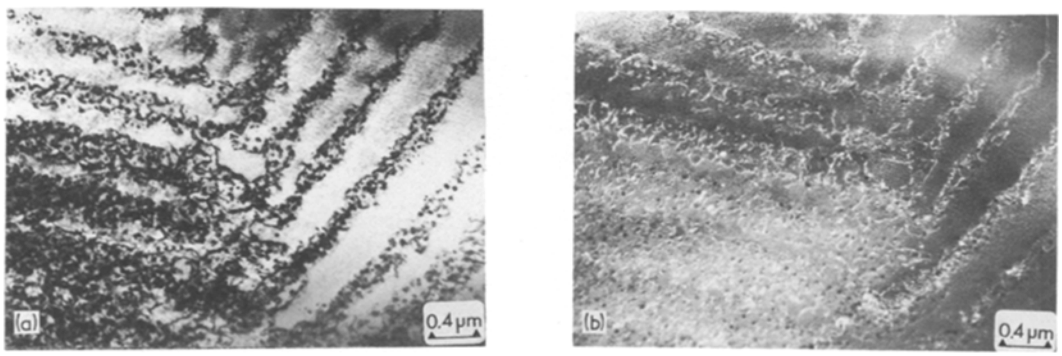

Figure 4(a) Banded arrays of θ particles on $\{1\,1\,1\}$ planes in Al–7 at.% Cu. (b) Weak beam image showing the dislocation substructure associated with (a).

standard thin region, using (a) 100 kV then (b) 1 MeV electron microscopy and (c) ion-beam thinning to reveal the microstructure of the bulk splat. Increasing thickness generally reflected the expected effect of slower cooling rate. Thus, for specimens containing < 13 at.% Cu, as thickness increased, first GP zones were detected by $\langle 1\,0\,0 \rangle$ streaking in diffraction patterns, then precipitate was observed in the sequence $\theta'' \rightarrow \theta' \rightarrow \theta$ similar to that in annealed splatted aluminium–copper eutectic [6]. However, when θ occurred it was either on grain boundaries or in a banded array (see Fig. 4a) associated with slip on $\{1\,1\,1\}$, as shown by the weak beam image in Fig. 4b. These dislocations must have been generated above room temperature by quenching stresses, to enable heterogeneous nucleation of θ to occur during subsequent cooling. In alloys containing >13 at.% Cu increasing thickness produced a less degenerate eutectic (see Fig. 5a) which eventually becomes lamellar, and is part of a radial rosette-type structure as revealed by high-voltage electron microscopy (see Fig. 5b). Similar structures have been observed previously [3, 13] but only by use of replica techniques from bulk-splatted specimens.

In ion-beam thinned specimens exhibiting the bulk microstructure, the predominant precipitate was θ which, above ~ 13 at.% Cu, was primarily part of a lamellar microstructure of the type shown in Fig. 6. These are not part of the radial lamellar microstructure shown in Fig. 5b, and are parallel for distances of several microns.

In order to compare the present results with those of other workers, it is necessary to know the approximate cooling rates. These have been estimated in the conventional manner [3] using interlamellar spacing. Well defined lamellae were only observed in specimens ion-beam thinned from the bulk and from the finest spacing observed (~ 20 nm) a cooling rate of $5 \times 10^{8}\,^{\circ}\mathrm{C\,sec^{-1}}$ was calculated. Knowing the maximum rate of ion-beam thinning, an upper limit to the as-splatted thickness was determined. The lower limit was taken to be observed thickness, and this thickness

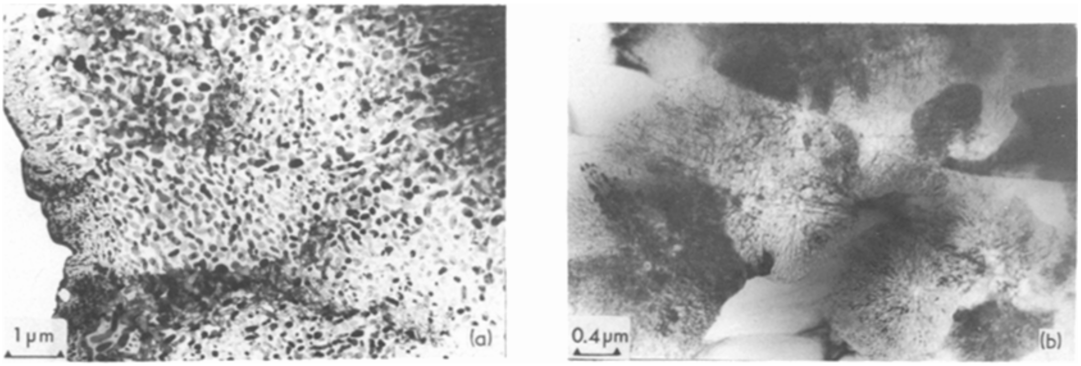

Figure 5(a) Variation of the degenerate eutectic structure with thickness in Al–17.3 at.% Cu. (b) Radial lamellar structure revealed by high-voltage electron microscopy, in Al–17.3 at.% Cu.

Figure 6 Parallel alternate lamellae of θ and aluminium–copper solid solution, from ion-beam thinned specimen of Al–17.3 at.% Cu.

variation introduced an error of ~ 10% into the cooling rate calculation. However, the cooling rate is the same order of magnitude as that reported by other workers using the same equipment [6, 8], and also similar to the values quoted in other investigations of aluminium–copper alloys. Thus it is considered reasonable that the thinner regions exhibiting degenerate eutectic, θ'', GP zone and single-phase structures have cooled at correspondingly faster rates.

It has been generally assumed [3, 5, 14] that degeneracy occurs when the cooling rate is too rapid to permit simple eutectic lamellar growth. However, Scott [15] reported contrary results when using a glass substrate to improve thermal contact at the expense of thermal conductivity. More rapidly cooled specimens were reported to have a lower proportion of degenerate eutectic, and a correspondingly higher proportion of fine lamellae. In this investigation, as shown clearly in Fig. 5a, the eutectic degeneracy is more marked at the thinnest, most rapidly cooled region of the specimen. In the thick regions of the specimen the structure is parallel lamellae (see Fig. 6). This would appear to confirm that the lamellar structures form at lower cooling rates, whilst degeneracy occurs above a certain critical value.

Studies of quenched aluminium–copper eutectic made in the same equipment [6] have reported bulk specimens consisting entirely of single-phase material, for similar quenching rates. This condition was only observed in the present investigation for specimens containing < 5 at.% Cu. It must therefore be concluded that for some reason, anomalously high cooling rates must have been achieved throughout the bulk foil. On the basis of our evidence it is considered that, in the more concentrated alloys, bulk material will generally exhibit some form of precipitation for conventional splat cooling rates.

4. Conclusions

(1) With increasing thickness (decreasing cooling rate) splat-quenched aluminium–copper alloys containing < 13 at.% Cu exhibit the conventional

breakdown of single-phase solid solution to GP zones $\rightarrow \theta'' \rightarrow \theta' \rightarrow \theta$.

(2) The matrix θ precipitation in alloys containing < 13 at.% Cu occurs heterogeneously on dislocations arising from quenching strains.

(3) With increasing thickness in alloys containing from 13 to 17.3 at.% Cu, the single-phase solid solution breaks down to a fine degenerate structure, which further develops into lamellae, which may be of a radial nature at intermediate cooling rates, or parallel at slow cooling rates.

(4) Extensive precipitation invariably occurs throughout the bulk of all specimens containing > 5 at.% Cu.

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