Letters

Some comments on the microstructure of rapidly quenched Al–Cu alloys

Recently, Williams and Edington have published in this journal several articles on the microstructure of splat-quenched Al–Cu alloys [1–3]. As contributors to this field we would like the opportunity to present a number of comments on these results and some observations of our own.

It is common practice to use the Al–Cu eutectic alloy as a monitor of cooling rate in splat-quenching [4, 5]. Usually the spacing of the lamellar structure is measured and a growth rate determined by extrapolation of data obtained from directional solidification experiments. From a knowledge of the foil thickness and by making assumptions about the nature of the cooling, a cooling rate may be determined. It has always been assumed that for the maximum cooling rate a high conductivity substrate is required although one of us (MGS) showed some years ago that the growth rates obtained when using a glass substrate were as high as those on copper [6]. Kijek and Matyja [7] have recently verified this result by showing that under otherwise similar conditions the cooling rate, as determined by the lamellar spacing method, is virtually independent of substrate material (copper, stainless steel, Al_2O_3 and glass were used). Likewise they have shown that the average supersaturation of Cu in the α(Al) phase (also a measure of cooling rate) does not change when the copper substrate is substituted by glass. These results are explained when one notices that although a glass substrate has a considerably lower thermal conductivity than a copper one the thermal contact between the melt and the substrate is much better in the former case. We have two pieces of evidence for this: first, the measured values of the splat–substrate heat transfer coefficient are higher for glass than for copper; second, scanning electron microscopy of the bottom surface of the splat-quenched foils shows that the area of contact with glass is greater than with copper. The improved contact is presumably a consequence of the flatness of the glass and its tendency to soften in contact with the melt; both features give enhanced wetting of the substrate by the molten alloy.

A problem in using lamellar spacing estimates of cooling rates is that the lamellar morphology is not obtained over the entire specimen and it is difficult to relate other microstructures to cooling rate. At the highest rates complete retention of the copper in aluminium solid solution is possible, even up to the eutectic composition [8]. With less severe quenching the excess copper is precipitated in various morphologies. Williams and Edington [3] claim that in alloys containing up to 13 at.% Cu the sequence is GP zones, θ'', θ' and θ with decreasing cooling rates, whereas alloys with > 13 at.% Cu have a lamellar eutectic structure which breaks down to a degenerate morphology at high cooling rates. This correlation of microstructure and cooling rate, however, is based on the assumption that cooling rate is a function of specimen thickness, a situation which is true only if the heat flow is everywhere perpendicular to the substrate. There is considerable evidence that this is not always the case and areas which have lifted off the substrate may have cooled considerably slower than thicker regions which have remained in contact [9]. The similarity of the microstructures observed by Williams and Edington in the thicker parts of their specimens to those obtained by annealing the supersaturated solid solution [8] leads us to propose two mechanisms for their formation. First, thick areas may be formed by the impingement of molten droplets on previously solidified regions causing an initially supersaturated solid solution to decompose; second, in the author's experience the heat liberated during ion-beam thinning is often sufficient to cause decomposition of a metastable structure. Both of these processes would lead to the microstructures observed and we suggest they may be more likely than the formation of the metastable phases θ' etc. directly during quenching since this would require cooling rates considerably slower than those usually associated with splat-quenching.

The degenerate eutectic morphology has been observed in many splat-quenching studies of this alloy and generally has been assumed to result from a high cooling rate, i.e. it is implied that at some cooling rate a transition from lamellar to degenerate occurs. If this is the case then the "critical" growth rate must vary from specimen to specimen since in some cases a degenerate structure has co-existed with a smallest lamellar spacing of 800 Å [5], whereas in others lamellae as fine as 100 Å have been obtained [10]. Although there is some evidence from other solidification work that a degenerate structure can result on isothermal solidification at high undercoolings [11, 12], the morphology is usually obtained at either very slow growth rates [13] or under conditions of non-uniform heat flow [14]. The latter case may well be the situation in splat-quenching where, as mentioned earlier, heat flow and therefore solidification do not occur unidirectionally from the substrate, particularly if the thermal contact between the melt and substrate is poor. We suggest therefore that a degenerate structure need not represent a particularly high cooling rate; it may be just a reflection of the non-uniformity of the heat flow conditions. In support of this we notice that in several cases an increase in the thermal contact parameter (either by use of a glass substrate [6] or by roughening a copper one [4] has been accompanied by an increase in the proportion of lamellar eutectic. It is also worth noting that the degenerate structure usually predominates near the surface of the specimen in contact with the substrate where again one might most expect non-uniform heat flow conditions. By analogy it is interesting to note that the first few millimetres of growth of directionally solidified rods often have a degenerate structure [14].

Finally, a feature commonly observed in splat-quenched Al–Cu alloys but rarely discussed is a radiating lamellar arrangement. Such non parallel lamellae may be part of a colony structure which occurs at a high ratio of growth rate to temperature gradient [13] but usually they have been observed to nucleate at primary particles even though the alloy is of eutectic composition. Their observation both in the plane of the foil [3, 7] and in transverse section [4, 5] suggests they may in fact be spherical arrangements which have grown as a consequence of nucleation within the liquid layer rather than at the advancing liquid–solid interface. EDAX analysis shows that the particles at the centre are a supersaturated solid solution of Cu in Al and are surrounded by a halo of $CuAl_2$ [15] consistent with observations elsewhere that the lamellar arrangement cannot grow directly from the primary Al-rich phase [16]. Similar eutectic "nodules" have been observed in more conventionally cooled eutectic alloys [13, 16].

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Reply to "Comments on the microstructure of rapidly quenched Al–Cu alloys"

In their article on the microstructure of rapidly quenched Al–Cu alloys, Scott *et al.* [1] not only make a number of general statements, but also refer to our earlier experimental results [2–4] and make specific criticisms of [4] alone. These are based upon:

(1) Our proposed correlation between decreased cooling rate and increased thickness of splat in specific, characteristic areas produced by single drop spreading.

(2) The influence on the microstructure of subsequent heating associated with either ion beam thinning or overlapping sequential droplet solidification.

Before discussing these points it is necessary to summarize the approach we have used to select areas for electron microscopy in the as-splatted material. We have pointed out earlier [2, 3] that this must be done carefully because different electron transparent regions in the splat do not necessarily experience the same cooling rate, particularly if the droplet–droplet or droplet–substrate reactions discussed by Vitek [5] are involved. Furthermore, we have expressly recognized the complicating factors of heat flow parallel to the surface and indeed presented evidence for it ([4], Fig. 1) in the form of columnar grain structures. In order to compare regions with reasonably equivalent cooling rates, such effects must be minimized. We suggested [3] that this could be done by comparing regions produced by single drop spreading that are transparent to 100 kV electrons.

The abundance of single drop spreading regions can be maximized by control of the splatting conditions. Furthermore, they may be recognized readily in our splats of Al alloys containing up to 33% Cu, since there was a band of single phase material near the edge of the thinnest regions and a narrow rim of thicker material at the edge. An example is shown in Fig. 1 which also demonstrates that nearby thicker regions were usually two phase. Clearly the single phase regions would be expected to have the fastest cooling rate, while the nearby thick regions cooled more slowly. Single drop spreading electron transparent regions of this type were selected for study in the work described in [4] in the as-splatted condition.

We now consider the two points made by Scott *et al.* [1].

(1) In areas selected using the above criteria, non-equilibrium cooling with significant heat flow parallel to the substrate will be reflected by changes in the microstructure that would lead to rejection of the area for possible study. So would sequential solidification of overlapping droplets. For example columnar grains may be produced, or phase transformations may occur, in the thinnest (normally single phase) regions. Therefore we do not accept the possibility of significant differences in heat flow and cooling rate in our selected areas.

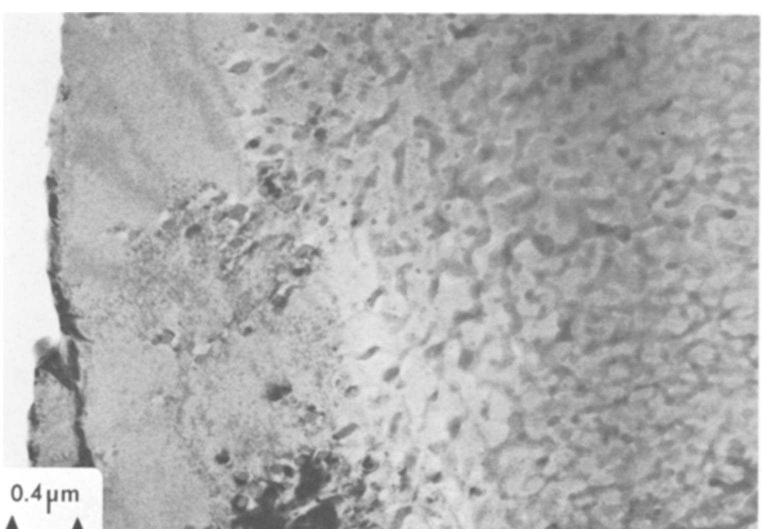

Figure 1 The edge of a typical thin area in an Al–33% Cu alloy produced by single droplet spreading. The edge of the foil is single phase, whereas the thicker regions are degenerate eutectic.

(2) Subsequent heating due to sequential solidification of droplets does not apply to our studies of the as-splatted condition because we did not observe surface precipitation, which is characteristic of decomposition on annealing splat quenched specimens (Scott and Leake [6]) and supersaturated thin foils in general [7]. A similar argument may be used to reject heating during ion beam thinning which in our experience normally arises from incorrect selection, and/or inadequate control of thinning conditions and/or poor thermal contact with support materials. Control specimens of quenched supersaturated Al–4% Cu alloys did not decompose as a result of ion beam thinning under identical conditions.

In conclusion it is obvious that interpretation of splat quenched microstructures is not straightforward in view of the many possible variables affecting the formation of the microstructure, and the lack of independent measurement of the variables that may control the solidification and precipitation processes at different points in the specimen. Our original use of a microstructural standard was an attempt to bring some degree of rationality to the issue. Obviously if the assumptions inherent in our standardization are not operating, then different factors will affect the observed microstructure. This observation was assumed to be trivial. We feel strongly that our method of determining the principal factors affecting the microstructure is still the only rational way available at present to permit comparison of microstructures from different splatted specimens, and different regions of the same splat.

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Cyclic fatigue of polycrystalline alumina in direct push–pull

Whilst considerable effort has been made in the study of fracture strength and creep properties of ceramic materials, comparatively little is know about their behaviour under cyclic fatigue conditions . Dynamic fatigue testing of brittle materials seems to be a difficult problem and in those few cases where it has been attempted it has yielded a wide scatter in the data, so that statistical methods of analysis have had to be employed [1–2].

More recently a limited amount of data has become available [3] on mechanical fatigue of Al_2O_3 which seems to show that this material is susceptible to dynamic fatigue. These results were obtained in repeated *uni-directional bending*, and as far as can be ascertained the influence of cyclic tension compression on fatigue behaviour of ceramics has never before been investigated. A method of dynamic fatigue testing of brittle materials in direct push–pull is described in this note. The method has been successfully applied to fatigue testing of "Lucalox" alumina.

The fatigue tests were carried out in a "Mayes" servohydraulic testing machine equipped with a low capacity pump for smooth running up to frequencies of 10 Hz. The use of a hydraulic machine is important to ensure axial movement of the loading ram. The gripping fixture consists simply of two adaptors (one mounted on the moving ram, the other on the load cell) each one containing a standard "Marlco" friction spring collet. The specimens, with cylindrical shoulders, are thus