Grain Size Measurements in Mg-Al High Pressure Die Castings Using Electron Back-Scattered Diffraction (EBSD)

Amanda Bowles^{1,2,*1}, Kazuhiro Nogita², Matthew Dargusch^{1,2},
Cameron Davidson^{1,3} and John Griffiths^{1,3,*2}

¹Cooperative Research Centre for Cast Metals Manufacturing (CAST), UDP 055, University of Queensland, QLD 4072, Australia

²Division of Materials, School of Engineering, University of Queensland, Brisbane, QLD 4072, Australia

³CSIRO Manufacturing and Infrastructure Technology, PO Box 883, Kenmore, QLD 4069, Australia

Optical metallographic techniques for grain-size measurement give unreliable results for high pressure diecast Mg-Al alloys and electron back-scattered diffraction mapping (EBSD) provides a good tool for improving the quality of these measurements. An application of EBSD mapping to this question is described, and data for some castings are presented. Ion-beam milling was needed to prepare suitable samples, and this technique is detailed. As is well-known for high pressure die castings, the grain size distribution comprises at least two populations. The mean grain size of the fine-grained population was similar in both AZ91 and AM60 and in two casting thicknesses (2 mm and 5 mm) and, contrary to previously published reports, it did not vary with depth below the surface.

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1. Introduction

Interest in the grain size of magnesium high pressure die castings is two-fold. *First*, magnesium benefits from potent grain size strengthening, deriving greater strength increases from a reduction in grain size than is the case for aluminium alloys. The Hall-Petch parameter, k_y , depends on alloy content and texture and varies from 0.2 to 1.25 MPa m^{-1/2} (see Ref. 1 for a review), in magnesium alloys compared to ~0.05 MPa m^{-1/2} in aluminium alloys.²⁾ However, unlike many castings and wrought alloys the grain size distribution in thin-walled high pressure die castings does not consist of one population. Large dendritic grains form in the shot sleeve and they are then carried through the gate (where some are fragmented) into the die cavity while a second population of small equiaxed grains is formed in the die cavity and these comprise the majority of the structure.³⁻⁷⁾ The effect of this mixed distribution on the Hall-Petch relationship is crucial to an understanding of the yield strength and it is, therefore, important to measure the grain size distribution. *Second*, many authors have noted that magnesium high pressure die castings have a “skin”, and some have claimed that it is characterised by an especially small grain size.⁸⁾ The skin is believed to increase the strength and ductility of castings;⁸⁻¹⁰⁾ measurements of the grain size variation with depth below the surface are required to test this claim and to assess the importance of such a fine-grained region on tensile properties.

Unfortunately, there are a number of difficulties associated with the measurement of the grain size in high pressure die cast Mg-Al alloys. Principally, no etchant is available that reliably reveals the grain boundaries in as-cast Mg-Al alloys (this, for reasons that are not clear, is not true for solution-treated alloys). An alternative to etching is available in high aluminium alloys such as AZ91. In these alloys the as-cast

microstructure consists of primary α grains and an ($\alpha + \beta$) eutectic which forms an almost-continuous network around the grain boundaries and which is readily observed. However, boundaries that lack the β (Mg₁₇Al₁₂) phase will be missed and the grain size will then be over-estimated. It can also be difficult to define the large dendritic grains formed outside the die cavity since the eutectic forms between the dendrite arms and the size of these grains can then be under-estimated. This method is, of course, not easily applied to alloys such as AM60 because the lower volume fraction of the eutectic phases means that the cell boundaries are not completely decorated. Another method, applicable to both AM60 and AZ91, is to solution heat treat the castings, dissolving the β phase and enabling etching to reveal the grain boundaries, but careful selection of a heat treatment time is required to avoid grain growth.

Electron back-scattered diffraction (EBSD) mapping can overcome the limitations of the conventional methods described above. EBSD patterns are Kikuchi patterns and these are determined by the orientations of the grains being examined. The orientation at a particular point where the electron beam is positioned can be determined by commercially available software and, by scanning the beam in a grid pattern, a map of the grain orientations is produced. When a discontinuous change in the orientation occurs, this signifies that a grain boundary has been crossed. In this way, the entire grain structure of a selected region can be mapped.¹¹⁻¹³⁾ The technique is being increasingly used for grain size determination and, while it unlikely to supplant established optical techniques due to the time-consuming nature of specimen preparation and the limited size of the area analysed, it can be invaluable in materials where standard optical techniques prove difficult to apply or where details of misorientation are required. It is difficult to apply optical analysis to Mg(Al) alloys in the as-cast state and previous attempts to use EBSD for these alloys have not been successful (but see Ref. 7) The reasons for this are unclear but appear to revolve around methods for surface preparation. Specifically, preparing the surface by mechanical polishing (e.g., with diamond polish-

*1Present address: Oak Ridge National Laboratory, Metals and Ceramics Division, TN 37931, USA

*2Corresponding author, E-mail: john.griffiths@csiro.au

Table 1 The grain size (equivalent circle diameter) of the fine-grained population as measured by EBSD at a number of depths below the casting surface in AZ91 (2 mm thick), AZ91 (5 mm thick) and AM60 (5 mm thick). Maximum grain sizes are from a separate population.

Alloy	0.2% proof stress and fracture strain	Depth below surface, μm	Mean grain size, μm	Maximum size, μm	No. of grains measured
AZ91 (2 mm)	161 MPa 2.2%	40	5.7	20	127
		70	5.1	23	157
		500	7.7	29	131
		1000	5.7	43	144
AZ91 (5 mm)	142 MPa 5.9%	150	6.1	33	153
		180	6.0	34	165
		500	6.0	32	177
		1000	6.7	34	181
		2500	6.5	52	430
AM60 (5 mm)	121 MPa 11%	100	5.1	27	747
		550	5.0	43	688
		2000	7.8	68	311

ing agents) or by electropolishing does not produce a surface finish good enough to allow EBSD mapping.

In this paper we describe the successful production of EBSD maps using ion-beam milling to prepare the surface. Data for three different sets of high pressure die castings are presented.

2. Experimental Procedure

Two magnesium alloys were used: AZ91D (9.0Al/0.8Zn/0.2Mn) and AM60B (5.9Al/0.01Zn/0.3Mn). High pressure die castings in the form of tensile specimens were produced on a 250 tonne Toshiba cold chamber die casting machine (see Ref. 14 for details of the casting conditions). The specimens had a rectangular cross-section with a width of 10 mm, and the thickness, t , was either 5 mm or 2 mm for the AZ91D specimens and was 5 mm for the AM60B specimens. The gauge length was 50 mm.

The tensile properties were published in Refs. 5 and 14, and are summarised in Table 1.

Sample preparation for EBSD involved specimens being mechanically polished to a 1 μm finish and then cold-stage argon-ion-beam milled. A gun tilt of 13° was used at 3.5 kV and 0.8 mA and the milling time was $\sim$ 1 hour. After milling, the specimens were stored under vacuum before being transferred to the SEM. In contrast to experience with, for example, aluminium alloys it was difficult to guarantee satisfactory specimens in that the EBSD patterns were sometimes unacceptably noisy. In such cases the specimens had to be re-milled, and small adjustments were usually made to the ion-beam current. A Philips XL30 scanning electron microscope equipped with an Oxford Instruments EBSD analyser was used. The system was operated at 20 kV with the sample tilted to 70°. The smallest detectable grain was $\sim$ 1 μm , so that all grain size calculations/distributions were made with a minimum equivalent circle diameter of 1 μm . Orientation maps were produced to reveal the structure of the material in which misorientations of greater than 15° were recorded as being distinct grains. The choice of 15° is far greater than the instrument resolution which is $\sim$ 3° but we

make two points. *First*, using a figure of 3° would demand very high quality surface preparation and, as noted above, this was difficult to achieve. *Second*, we estimate that the fraction of grains missed at 15° is negligible ($\sim$ 1%) and, indeed, preliminary runs using 5°, 10° and 15° produced similar numbers. A qualitative examination was made of the influence of misorientation angle on the delineation of grain boundaries in two samples. These were compared to grain boundaries determined from the same fields by a manual estimation from orientation images. This showed that using 5° discrimination and, to a lesser extent, 10°, gave a false identification of tiny grains. On the other hand, 5° discrimination would occasionally split large grains at low-angle grain boundaries, while 10° and 15° would sometimes result in counting of apparently different adjacent grains as a single larger one. These would bias the distributions somewhat, although the numbers involved were few enough to result in only minor differences in the mean grain size. It was concluded that 15° was the optimum discrimination, giving the most self-consistent measure of grain size.

A final source of error, that is not accounted for in this study, is that a few large grains in the centre of castings exceeded the size of an individual field of view for EBSD. Such heterogeneity would require far more fields in order to characterise it fully, as well as a modified acquisition or analysis technique. The fields of view for this work were deliberately chosen to avoid the large grains.

3. Results and Discussion

The microstructures of high pressure die castings of AZ91 and AM60 have been described by numerous authors.^{8,15,16} Figure 1(a) is a typical micrograph and shows the presence of the large α grains that formed outside the die cavity and that are segregated towards the centre of the casting. Figure 1(b) shows the primary α (Mg rich) grains with a divorced eutectic of β (Mg₁₇Al₁₂) and α (Al rich). The dark-etched regions around the β phase seen in Fig. 1(b) are commonly referred to as eutectic α , (in this case labelled α') and are enriched in aluminium.

Fig. 1 Optical micrographs of AZ91. (a) the large white grains are the primary α phase and the surface of the casting is at the top of the micrograph. Image (b) shows grain boundary β phase precipitates, the primary α phase and the aluminium rich α' . Etching conditions: 1 s in Nital (2% concentrated nitric acid in ethanol).

Fig. 2 Images of AZ91 (2 mm thick). Top pair show a 15° EBSD grain orientation map and the associated secondary electron (SE) image obtained during acquisition of EBSD data. Bottom pair show the optical and SEM secondary electron images of the same region after removal of the carbon contamination followed by etching. All images are to the same scale shown by the 30 μm bar in the centre.

Figure 2 is a series of images from the EBSD analysis together with an optical micrograph of the same area. The area used for the EBSD mapping was visible under the optical microscope because of carbon contamination on the surface, and microhardness indents were made at the corners

of this area to act as markers for subsequent optical metallography. The electron image shows damage produced from the ion beam milling (long rows of dimples running diagonally across the field of view). The optical and secondary electron images show approximately the same

Fig. 3 EBSD grain maps of AZ91 (2 mm thick) at depths of 40 μm , 70 μm , 500 μm and 1000 μm below the casting surface in a transverse section. All maps are to the same scale, shown by the 20 μm bar in the centre.

region as that analysed (the EBSD map is produced when the sample is tilted 70° so it is difficult to match the images exactly even though a tilt correction was applied). The large grain labelled A in the EBSD map is visible in the top left corner of all of the images.

The optical and scanning electron images illustrate the difficulties in using the eutectic grain boundary decoration to measure grain size. EBSD grain maps were obtained at a number of depths below the casting surface. An example of a series from AZ91 (2 mm thick) is shown in Fig. 3. At depths less than 100 μm the microstructure consists primarily of equiaxed fine α grains while at $\sim 1000 \mu\text{m}$, several large α grains can be seen (see also Fig. 1).

The grain size data are presented as histograms in Fig. 4. The effect of a few large grains on the distributions is seen in the area plots, it being less obvious in the number plots.

There have been many studies of the grain size distribution in annealed alloys^{17–19} although not, it appears, of chill-cast structures. Most authors take, as a starting point, the lognormal distribution in which the data form a straight line on a logarithmic probability plot. Figure 5 shows the results of tests to investigate whether this is true of the data in Fig. 4.

It can be seen that the $\log(\text{grain diameter})$ data fit a normal distribution only moderately well, with the 40 μm , 70 μm and 500 μm data showing a positive deviation from the straight line at larger grain sizes (13 to 20 μm). All data show a slight

negative deviation at grain sizes below 3 to 4 μm , with the effect being slightly more pronounced in the 1000 μm data. Similar plots were made for the other two sets of castings and these showed the same general features. There are several possible explanations for such behaviour. First, and fundamentally, the size distribution in chill castings cannot be exactly lognormal since the impingement of growing grains during solidification sets an upper limit to their size, even though the distribution may approximate lognormal at intermediate sizes. Such an upper size limit will appear as a positive deviation at that end of the distribution. However, we do not at present have a large enough dataset to examine whether this explanation is valid. A second possibility is related to the previous one. Schückher¹⁸) shows that such deviations are characteristic of log-probability plots for linear measurements of grain size even when the volume distribution is lognormal. However, we believe this explanation may be invalid in the present case because the deviations in Fig. 5 appear to be too severe. A third explanation is that the population is mixed (it is readily shown that adding data points at one end, for example, of a lognormal distribution has this effect) and the optical micrographs certainly suggest this is the case. A useful rule-of-thumb²⁰) is that if the area of the largest grain differs from the mean by more than a factor of 8 then the distribution is likely to be mixed, and this criterion is easily met for the present data, see Table 1.

Fig. 4 Histograms showing the number of grains (counts) and the grain area as a function of equivalent circle diameter (ECD) from the EBSD maps in Fig. 3, at 40 μm , 70 μm , 500 μm and 1000 μm below the casting surface.

Gifkins²¹⁾ has earlier proposed a similar criterion but with a factor of 4. Finally, we have already commented that the size resolution for the technique is $\sim 1 \mu\text{m}$ so that the data at that end of the graph are less reliable than elsewhere.

Unfortunately, the sample size here is too small to allow quantitative estimates of the means of possible constituent populations and much further work would be needed to improve the statistics. The populations cover a diameter range in excess of two orders of magnitude, and analysing sufficient fields of view would require an unjustifiably long amount of machine time. As a first approximation to determining the relative amounts of the two populations, we note that the fraction solidified prior to injection through the gate is reported to be in the range of 10–20%^{4,6)} and our own earlier analysis of these present die castings found that the volume fraction of large α grains (defined as any with an area greater than $110 \mu\text{m}^2$) is 4%, 4% and 10% in the AZ91 (5 mm thick), AZ91 (2 mm thick) and AM60 (5 mm thick) castings, respectively.⁵⁾

Despite these uncertainties, the means for the four datasets, assuming straight lines in Fig. 5 in the interval $16\% < p < 84\%$ (i.e. $\pm$ one standard deviation), are given in Table 1 together with data for the other two types of castings. The maximum grain sizes in the field of view are also given

although they are not part of the recorded means; these grains are possibly part of the separate populations. The logarithms of the standard deviations are given by the slope of the lines in Fig. 5 and are, typically, ~ 0.4 . This is only slightly larger than the value of ~ 0.3 found for several annealed metals¹⁸⁾ and cannot of itself be used to assert that the population is mixed although as pointed out in the previous paragraph, the maximum grain sizes measured are a strong indication of that.

Table 1 shows that the mean grain size of the fine-grained population measured by EBSD ranges from 5 to 8 μm and does not show a systematic variation with the casting thickness, alloy composition or, especially, depth below the surface. This last finding conflicts with three earlier reports of a significant variation in the grain size of the fine-grained population with depth below the surface. Sequeira *et al.*⁸⁾ report that this grain size increased from approximately 0.5–4 μm at the surface to 10 μm in the centre of AZ91 castings (1, 2 and 6 mm thick). Similarly, Carlson²²⁾ found an increase from $\sim 13 \mu\text{m}$ at the edge to 26 μm at the centre of an AM60 casting. Finally, Rodrigo *et al.*²³⁾ found that the grain size of samples from 2 mm thick AM60 castings was 7 μm at the edge and 17 μm in the centre. In all these cases standard optical metallography was used to measure the grain size and, as pointed out in the Introduction, there are several difficulties with this technique. It is possible, therefore, that the optical methods have unique observational errors that do not apply to EBSD. Interestingly, Weiss and Davies²⁴⁾ compared EBSD with a conventional metallographic method for measuring the grain size of an aluminium alloy. They found that the optical technique overestimated the grain size by more than 40% and considered the EBSD data to be more reliable. Clearly, further work is needed to make direct comparisons of optical and EBSD methods for Mg-Al alloys in the same way as done by Weiss and Davies. A final comment regarding our calculations of grain size (and those of the other authors cited here) is that the numbers refer to the apparent grain size as measured on a polished cross-section. That is, no corrections have been made to transform the data to the true three-dimensional grain size.

4. Conclusions

The following conclusions can be drawn from this work:

- (1) If appropriate surface preparation methods are used, EBSD can be used to measure the true as-cast grain size and grain size distribution in high pressure die cast Mg-Al alloys.
- (2) The grain size distribution in the castings examined is mixed although the sample size is too small to make good estimates of the means of the constituent populations.
- (3) The grain size of the fine-grained population in the three groups of castings examined in this work does not depend on alloy content, casting section size nor, contrary to previously published data, with distance below the cast surface.

Fig. 5 A logarithmic probability plot to test for lognormality of the small grain-size data from Fig. 4 (AZ91: 2 mm thick casting).

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