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Influence of barium addition on the microstructure and the rheological behaviour of partially solidified Al–Cu alloys

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Abstract

The effect of a barium addition to Al–Cu alloys on the morphology of liquid films in the mushy zone has been studied. It is shown that barium improves wetting of the solid phase by the liquid and thus, delays coalescence of the grains. In addition, tensile and shear tests have been carried out during solidification to determine the influence of Ba on the mechanical behaviour of the mush. It is observed that the fracture stress in tension is affected by the presence of Ba at the very end of solidification whereas an effect of Ba is not detectable in shear. It is proposed that this is due to delayed coalescence for Ba-treated alloys for which liquid films embrittle the alloy in tension up to very high solid fractions.

Keywords: Solidification; Aluminium alloys; Mechanical properties testing; Wetting

1. Introduction

Hot tearing is a severe defect widely encountered in industrial casting processes of aluminium alloys. It occurs during solidification when the alloy is in a mushy state where both solid and liquid phases coexist. Towards the end of solidification solid dendrites start to coalesce and solid permeability drops dramatically. Thus, the liquid phase that remains in the mush cannot accommodate the thermal strain, the solidification shrinkage and the possible casting strains experienced by the ingot. This can lead to the formation of pores that may degenerate into hot tears. Many studies on hot tearing phenomena have been performed [1–4] and several hot tearing criteria have been suggested [5–8]. The rheological behaviour of the mushy zone plays a great role in the hot tearing susceptibility of the alloy. Constitutive equations have been proposed to model the mechanical behaviour of semi-solid alloys accounting for the effect of the stress state [9–13]. They are generally based on the viscoplastic behaviour of the solid alloy extended into the semi-solid state by taking into account the presence of liquid films. This is done by introducing either an internal cohesion variable by Ludwig et al. [9] and Martin et al. [10] or the fraction of grain boundary area covered with liquid by Van Haaften et al. [11]. These models agree that, at high solid fractions, the presence of thin liquid films that wet the dendrites has a great influence on the rheology. The presence of such liquid films towards the end of solidification depends on the wetting properties of the solid phase and more specifically on the solid–liquid interface energy γ_{sl}. A decrease of γ_{sl} enables more efficient wetting of the solid phase by the liquid and thus, leads to the embrittlement of the mush. Gullo et al. [14] have studied the influence of barium as an additional element on thixoforming of Al–Mg–Si alloys. They concluded that barium decreases γ_{sl}.

The aim of this study is firstly to verify the surface active role of barium for Al–Cu alloys in the semi-solid state and secondly to determine the consequences of the addition of barium for the mechanical behaviour of the mushy zone through tensile and shear tests carried out during solidification at small strains (<0.3) and slow strain rates ($<10^{-3}\ s^{-1}$).

2. Materials

An alloy for the study was cast in Alcan CRV solidification laboratory. The base metal was 99.99% Al in order to eliminate possible influence of other elements on the results. It was alloyed with 4.26 wt.% Cu and 350 ppm Ti, grain refined

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with 1 kg $tonne^{-1}$ Al–5%Ti–1%B commercial grain refiner rod, stirred for 5–10 min and then cast at 700 °C into sampling moulds. Ti was introduced through a Al–10 wt.%Ti master alloy (322 ppm) and through the grain refiner (28 ppm). Depending on the heats, 307 ppm Ba (±12 ppm) was also added to the melt as a pure metal prior to casting. The chosen steel mould (28 mm inner diameter, 60 mm height and 1 mm wall thickness) provided a maximum cooling rate of 8 K s^{-1}. This relatively low cooling rate allows one to obtain a sufficiently coarse microstructure to assure good resolution upon microstructural observation. For this observation, the specimens were prepared by standard polishing techniques (grinding on various SiC paper and final polishing using Alumina suspension). Examination was carried out with a Philips XL30 SEM.

3. Results and discussion

3.1. Effect of barium on wetting properties

The average grain size of the grain refined alloy is $d = 140 \pm 10\,\mu m$. The addition of barium shows no refining effect; on the contrary it seems to slightly coarsen the microstructure (Table 1). In addition, there is no evidence of a phase containing Ba. Indeed, no information is available concerning the possible alteration of the binary Al–Cu phase diagram by a small addition of Ba. At a level of 0.03%, the modification of the phase diagram, if any, should be very small and the possible formation of additional phase containing Ba should not change the microstructural parameters in a significant manner.

High contrast between primary aluminium and the Al_2Cu phase allows these two structural constituents to be easily distinguished by SEM observations. As shown in Fig. 1, eutectic (Al + Al_2Cu) is observed in wider areas between primary Al dendrites while in thinner zones only films of Al_2Cu are present, resulting from divorced eutectic. This observation is consistent with the results of Pompe and Rettenmayr [15] who have studied the microstructural changes which occur during quenching of Al–6.8 wt.% Cu alloys. Indeed, they have shown that for such a low cooling rate (8 K s^{-1}) the microstructure present at a given temperature in the semi-solid state is modified during final solidification. The (Al) phase of the eutectic agglomerates with primary (Al) dendrites leaving the Al_2Cu phase alone into narrow interdendritic spaces. Hence the observed microstructure does not accurately reflect the solid/liquid microstructure that has been present above the solidus.

Table 1
Average grain size d, S_v and $S_v d$ determined by image analysis for samples with and without barium

	d (μm)	S_v (μm^{-1})	$S_v d$
Without barium	140 ± 10	0.024 ± 0.03	3.3 ± 0.6
With barium	150 ± 10	0.033 ± 0.03	5 ± 0.8

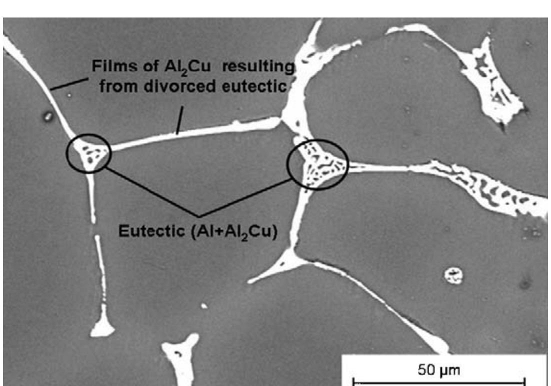

Fig. 1. SEM micrograph of the Al–4 wt.% Cu alloy showing eutectic areas and films of Al_2Cu.

The detection and measurement of some geometric features can be achieved by image analysis. The interface density S_v (interface area per total volume) has been calculated:

$$S_v = \frac{4L_s}{\pi A_T}, \tag{1}$$

where L_s is the length of the interface between the eutectic (or Al_2Cu) and Al dendrites and A_T is the total area of the micrograph. The word "eutectic" will be used in the following whether it is really the eutectic or just Al_2Cu.

Note that the S_v value calculated after solidification by Eq. (1) is artificially higher than the actual value in the semi-solid state [16] owing to the microstructural changes occurring during cooling of the samples. However, it has been also shown that this parameter is less sensitive to changes during quenching, as compared to the solid fraction [15]. Nevertheless, it allows the effect of barium on the solid–liquid interface to be characterised since alloys with and without barium have been solidified under the same conditions. Table 1 indicates that the S_v parameter is 40% higher in the case of the samples treated with barium and the difference between the alloys is even higher when the dimensionless parameter $S_v d$, which represents the interface area per grain, is considered. This observation shows that the addition of barium leads to a significant increase of the amount of solid–liquid interfaces, thus, improving the wetting of the solid phase by the liquid at the end of solidification.

In order to determine the position of the eutectic films, an Electron Back Scattering Diffraction (EBSD) analysis (Fig. 2a) has been performed to measure the crystalline disorientation between grains. This technique has been combined with microstructural observations (Fig. 2b) to plot the thickness of the eutectic films present between two grains as a function of the disorientation of the grains. It is to be noted that the eutectic films are mainly located at the interfaces between grains. All the measurements have been carried out on films located between two different grains. Fig. 3 shows the results obtained for samples with and without barium. It is restricted to the grains having disorientations lower than 20° and to eutectic films with a thickness larger than 1 μm (Fig. 2b). For these chosen eutectic films, a high magnification was used so that the thickness of the films can be measured with an accuracy of ±0.1 μm.

Fig. 3 indicates that samples without barium do not have observable films for grain disorientation lower than 10° while