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Fractography and transmission electron microscopy analysis of an Al–Li–Cu–Mg–Zr alloy displaying an improved fracture toughness

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Al–2.3 wt % Li–1.8 wt % Mg–0.15 wt % Zr alloys with 0.1 and 0.45 wt % Cu were studied. The materials were cast, homogenized at 530 °C for 24 h, extruded at 400 °C with a reduction ratio of 30 : 1, solution treated at 528 °C for 0.5 h and then artificially aged at 190 °C to an overaged condition. The concerted application of light-, transmission electron- and scanning electron microscopy together with short rod fracture toughness measurements allowed the influence of low level Cu together with 0.15 wt % Zr on the fracture toughness of this alloy system to be studied. It was found that the addition of 0.1 wt % Cu together with 0.15 wt % Zr not only affected the δ' precipitate nucleation and their size and size distribution through Li–Cu–v triple complexes but also affected the β' dispersoid nucleation through Zr–Cu–v triple complexes. This in turn affected the size and size distribution of the δ'/β' duplex precipitates. This causes an increase in the fracture toughness in underaged material artificially aged at 190 °C. This effect was probed in an SEM fractographic investigation and by mechanical testing.

1. Introduction

The addition of Li to various Al alloys has a profound influence on their mechanical properties. For example, Al–Li–X alloys exhibit an increased strength, higher elastic modulus and a lower density when compared to conventional high strength Al alloys. However, the observed low fracture toughness and poor ductility values in this alloy system are problems that remain to be solved. [1]. One way of doing this is by the addition of ternary (or quaternary) alloying elements. Several reports on the effect of Cu, Mg and Zr on the Al–Li binary alloy system exist in the literature [2–5]. Recently Kumar *et al.* [6] and Özbilen [7, 8] studied the effect of low level Cu additions on the mechanical properties of the Al–Li–Mg alloy system during artificial ageing at 190 °C.

In the present study, the effect of low level Cu additions, combined with a 0.15 wt % Zr addition is investigated. The fracture toughness of a Al–2.3 Li–1.8 Mg–0.15Zr alloy with a 0.1 and 0.45 wt % Cu content has been studied in detail using optical- and transmission electron (TEM)- and scanning electron (SEM) microscopy. The TEM was used to probe the microstructure whilst the SEM was used in the fractographic studies. In all the experiments careful control of the chemical composition, processing and artificial ageing cycle of the material was maintained.

2. Experimental procedure

The materials used in the present study were prepared as previously reported [8] with a nominal composition (wt %) Al–2.3 Li–1.8 Mg–0.15 Zr having 0.1 and 0.45 wt % Cu. Materials were homogenized at 530 °C for 24 h which is the optimal treatment suggested by a differential thermal analysis (DTA) study reported in earlier work [8], followed by extrusion at 400 °C with a 30 : 1 reduction ratio and 3.5 mm s^{-1} ram speed. Following the extrusion processing the samples underwent standard solutionizing at 528 °C for 0.5 h and artificial ageing at 190 °C to produce an overaged condition. For optical microscopy of the as-cast and homogenized structures, specimens were prepared by standard metallographic procedure, i.e., grinding with SiC papers, polishing with 3 and 1 μm diamond paste and etching in Keller's reagent. Specimens were examined using a PME Olympos, Tokyo optical microscope. Short rod fracture toughness (FT) measurements and TEM analysis using a JEOL 100 PX Temscan oparating at 100 kV were carried out in the manner previously reported [8]. An Hitachi SEM was used in the fracture surface characterizations of the samples.

3. Experimental results

The microstructure of the as-cast materials showed fine, equiaxed grains approximately 70 μm in size as is shown in Fig. 1a for the alloy containing 0.45 wt % Cu. There was little porosity. Back scattered electron imaging using the SEM on the same alloy as shown in Fig. 1b, showed the presence of segregated phases having a high Cu content in interdendritic regions. They appeared to be bright due to the high atomic weight of Cu. The optimum temperature for the homogenization had been previously determined by DTA analysis [8, 9] and Fig. 1c shows the homogenized structure of the alloy with 0.45 wt % Cu having a fairly uniform microstructure without any segregated phase apart from black etch pits introduced into the structure by the chemical etching. At the end of the heat treatment cycle, samples were subjected to short FT tests in as-extruded (AE), solution treated (ST), underaged (UA), peak aged (PA) and overaged (OA) conditions. The results are shown in Fig. 2. The times to produce the UA, PA and OA states were selected from ageing curves [6, 8, 9]. TEM analysis results for the Al–2.3 Li–1.8 Mg–0.15 Zr alloys with 0.1 and 0.45 wt % Cu are shown in Fig. 3(a–h). In Fig. 3a, very fine (10–15 nm) and numerous δ' coherent precipitates were observed in the 0.1 wt % Cu containing alloy in the UA condition. Fig. 3b, TEM work near $[0\,0\,1]_{\mathrm{Al}}$ shows that the microstructure of the same alloy at the same condition exhibits very fine and dense δ' and $\beta'(\mathrm{Al_3Zr})$ coherent precipitates with typical Ashby–Brown lobe-type strain contrast. While the distribution of the δ' precipitates was uniform, that of the β' particles was non-uniform as can be observed in Fig. 3b. The selected area diffraction (SAD) pattern taken on the area shown in Fig. 3b is shown in Fig. 3c which was taken from the $[0\,0\,1]_{\mathrm{Al}}$ matrix zone axis. The discrete superlattice reflections at $(\frac{1}{2}\;0\,0\,2)$ and $(\frac{1}{2}\;2\,0\,0)$ are due to coherent β'-precipitate particles. The diffuse intensity maxima associated with the superlattice positions that can be seen in Fig. 3c, are due to the very fine, coherent δ'-particles. The OA condition of the same alloy is shown in Fig. 3(d–f). Medium sized ($\sim$25 nm) δ' particles and a few $(\delta' + \beta')$ composite precipitates of $\sim$50 nm size can be observed in the bright field (BF) image (Fig. 3d) and the dark field (DF) image (Fig. 3e) together with the corresponding (Fig. 3f) SAD image in the $[0\,0\,1]_{\mathrm{Al}}$ beam direction exhibiting δ' superlattice reflections with Al-matrix diffraction spots. The OA condition of the alloy with 0.45 wt % Cu is shown in Fig. 3g (BF) and in Fig. 3h (DF). Coarse δ' ($\sim$50 nm) and duplex δ'/β'($\sim$60–100 nm) precipitates, etched $\mathrm{Al_2LiMg}$ grain boundary precipitates and precipitate free zones (PFZ's) at grain boundaries and at around incoherent $\mathrm{Al_2LiMg}$ particles can be observed in Fig. 3g. The corresponding DF image of the same area of the same alloy sample, featured the same microstructural characteristics as those shown in Fig. 3h. Fig. 4(a–f) shows fracture surfaces of the Al–2.3 Li–1.8 Mg–0.15 Zr alloy with 0.1(a–d) and 0.45 wt % Cu (e, f) observed using SEM. In the 0.1 wt % Cu containing alloy, the fracture surface of the ST condition (Fig. 4a) does not show any significant signs of ductile fracture mode whereas ductile fracture is clearly visible in Fig. 4b for the UA condition. A large number of dimples, and areas of plastic deformation during fracture that are characteristic of a ductile fracture mode can be observed. Dimples are also observed in the PA condition of the same alloy (Fig. 4c) but in lower numbers than observed in the UA condition (Fig. 4b). The OA condition of the same alloy had no dimples on the fracture surface (Fig. 4d): which is a characteristic of brittle fracture mode. There were also some minor cracks on the fracture surface. Lower magnification SEM fractographs of the 0.45 wt % Cu containing alloy are shown in Fig. 4e for

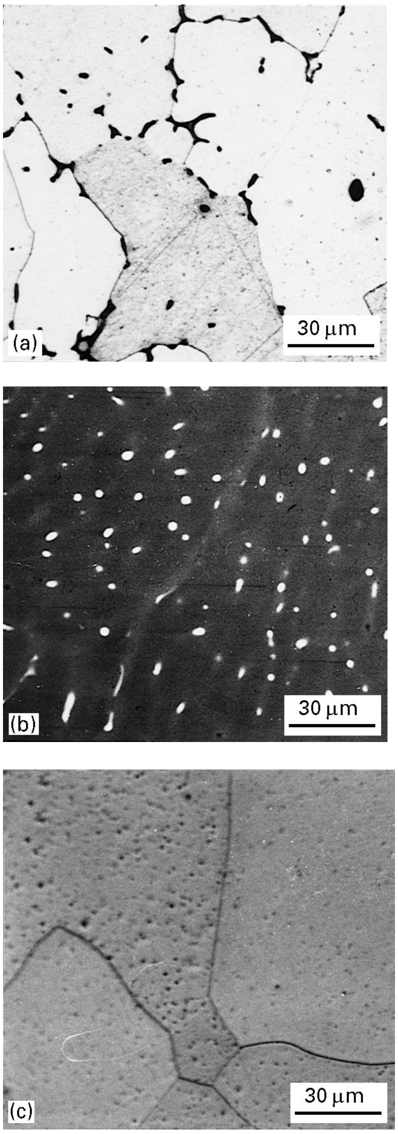


Figure 1 Optical micrographs of the Al–2.3 Li–1.8 Mg–0.15 Zr–0.45 Cu alloy in (a) as-cast and (c) homogenized condition together with (b) a SEM of the as-cast microstructure.

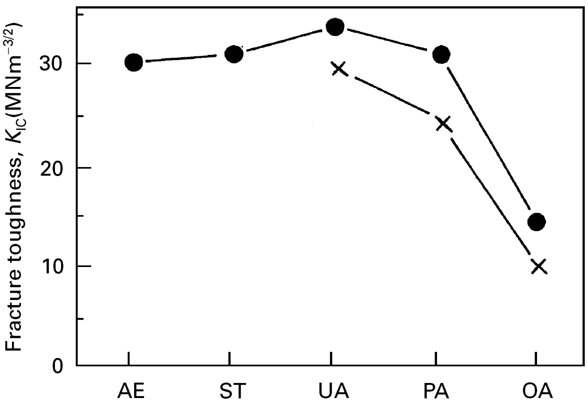
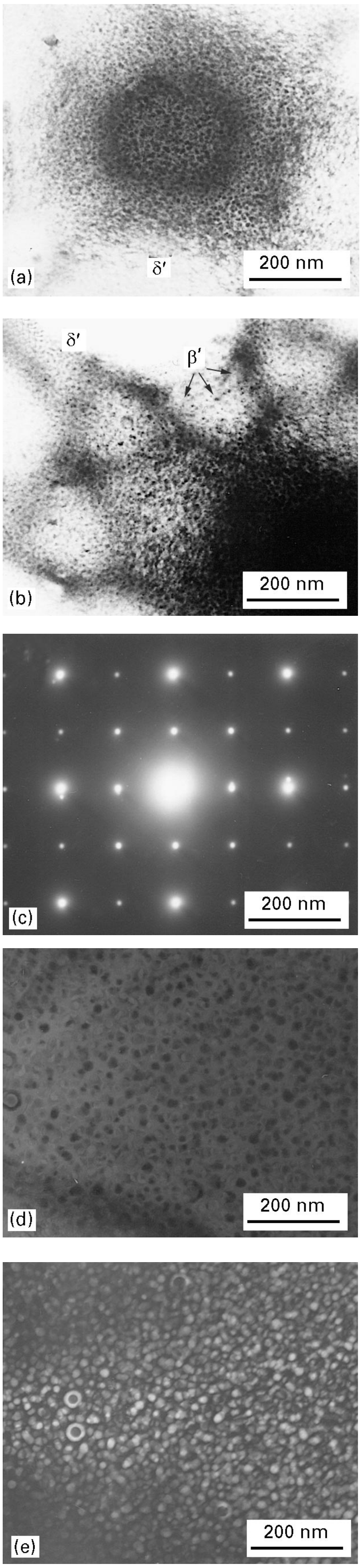


Figure 2 Fracture toughness versus processing conditions in Al–2.3 Li–1.8 Mg–0.15 Zr alloy containing: (●) 0.1 and (×) 0.45 wt % Cu.

the UA-condition and in Fig. 4f for the OA-condition. When the fractographs of the sample in the UA condition (Fig. 4e) showed signs of plastic deformation and a few minor cracks, there were no signs of plastic deformation but wider cracking (Fig. 4f) at the fracture surface of the alloy in the OA condition sample confirms the highly brittle nature of the fracture.



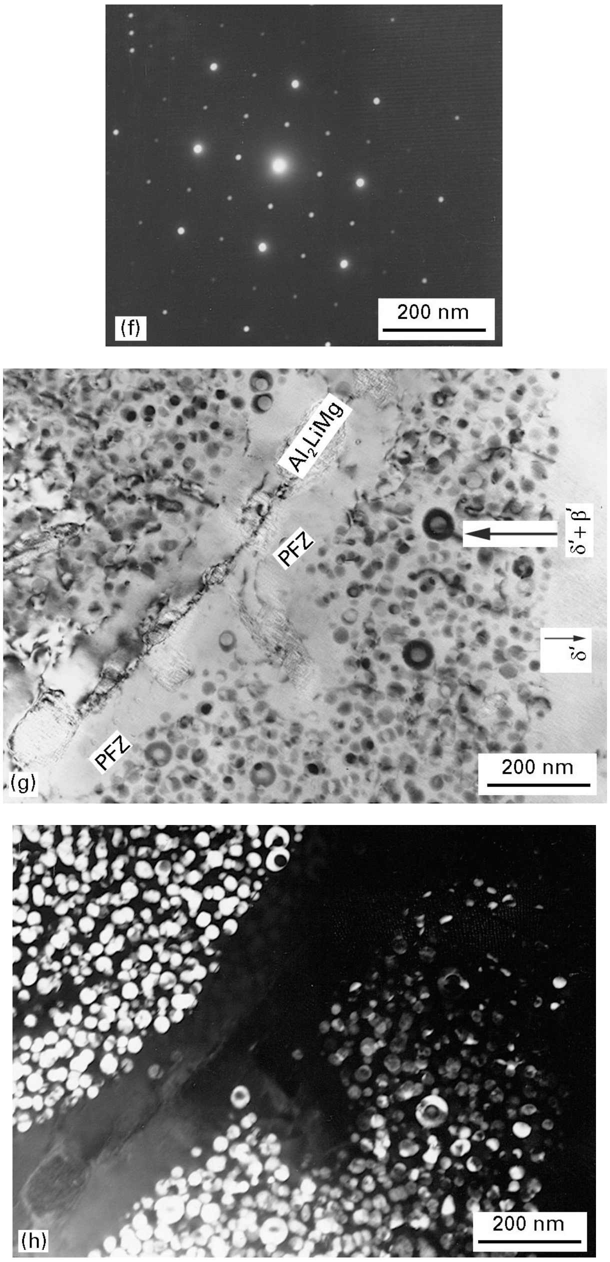


Figure 3 TEM analysis of the Al–2.3 Li–1.8 Mg–0.15 Zr alloy containing 0.1 wt % Cu in (a–c) UA, and (d–f) OA conditions and (g–h) 0.45 wt % Cu in OA condition.

4. Discussion

The as-cast materials contained various amounts of the δ′, δ′/β′, δ and Al_2LiMg phases in agreement with previous reports [8]. The alloy containing 0.45 wt % Cu had a eutectic phase mixture that contained the T_2 (Al_6CuLi_3) phase which was confirmed by the optical and scanning electron microscopy investigation of the as-cast alloy as shown in Fig. 1(a and b). Dark (Fig. 1a) and bright (Fig. 1b) areas at interdendritic regions correspond to this eutectic phase mixture containing the T_2 phase which has a high Cu content with respect to the α-Al matrix and other phases. The T_2 phase also has been found in as-cast Al–Li–Cu–Mg–Zr alloys by chemical analysis [10] and differential scanning calorimetry analysis [8].

Homogenization of the as-cast samples resulted in fairly homogeneous structures. In Fig. 1c the homogenized structure of the 0.45 wt % Cu containing alloy was shown to contain some etch pits but no segregated phases thus confirming the suitability of the application of the homogenization practice to the alloys used in the present study. FT test results (Fig. 2) show that the highest FT value is obtained in the 0.1 wt % Cu containing alloy in the UA condition ($34\ \mathrm{MN\,m^{-3/2}}$). This is consistent with the TEM microstructural investigation and also the SEM fractography results shown in Fig. 3(a–c) and Fig. 4b. Numerous fine δ' and β' particles present in the microstructure and a considerably more dimpled fracture surface that indicate a high level of ductility and plastic deformation during fracture, are signs of improved FT values. The less dimpled nature of the ductile fracture mode in the ST condition of the same alloy confirms the lower FT values than that in UA condition. In the same alloy, the FT values decrease in the PA and OA conditions as shown in Fig. 2. The fracture surface mode changes from ductile to brittle as the sample changes from the UA condition to the OA condition as shown in Fig. 4(b–d). TEM work on the same alloy in the OA condition (Fig. 3(d–f)) showed uniformly distributed coarse δ' and non-uniformly distributed very coarse ($\delta' + \beta'$) composite precipitates that cause lower FT values. Increasing the Cu content to 0.45 wt % results in an overall decrease in the FT values with respect to the 0.1 wt % Cu containing alloy regardless of the artificial ageing steps. This is due to the very coarse δ', δ'/β' duplex precipitates, the Al_2LiMg grain boundary equilibrium particles and PFZ's at grain boundaries and also around Al_2LiMg particles as is shown in Fig. 3(g and h). The FT value ($16\ \mathrm{MN\,m^{-3/2}}$) in the 0.1 wt % Cu containing alloy in the OA condition is higher than that ($8\ \mathrm{MN\,m^{-3/2}}$) in the 0.45 wt % Cu containing alloy at the same heat treatment condition (Fig. 2). The TEM and SEM results are consistent with this result. The observation of Al_2LiMg precipitates and no PFZ's, finer δ' and $\delta' + \beta'$ particles (Fig. 3(d and f)) in the 0.1 wt % Cu containing alloy in the OA condition results in the observed higher FT values. A comparison of Figs 4d and 1f shows that as the content of Cu increases the amount of minor cracks also increases. The brittle nature of the fracture mode is also more clear in Fig. 4f of the 0.45 wt % Cu containing alloy. This suggests that the increase in Cu, together with Zr, causes more intense co-planar slip resulting in lower FT values. The intense co-planar slip may result from the increase in size of the δ' and δ'/β' duplex precipitates, as observed in the present study, with the increase in Cu content. Microstructural observation using TEM and fractographic studies using SEM indicate that Cu together with 0.15 wt % Zr improves the FT by refining the size, size distribution and density of the δ' precipitates as previously reported [6, 8]. Zr also affects the size of the δ' precipitates by supplying the sites for nucleation of δ' precipitates since it is well known that in Zr containing alloys duplex δ'/β' precipitates form with δ' encapsulating the β' nuclei [11] as well as the δ' particles observed in the matrix as shown in Fig. 3. Thus, Zr can affect the size and size distribution of the δ' precipitates which results in

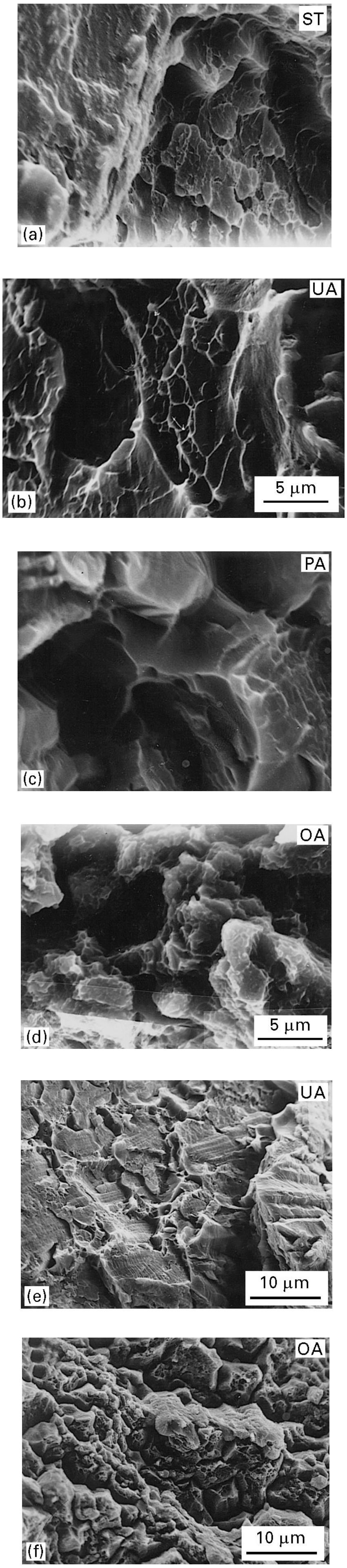


Figure 4 SEM fractographs of the Al–2.3 Li–1.8 Mg–0.15 Zr alloy with (a–d) 0.1 wt % Cu and (e, f) 0.45 wt % Cu.

modified FT values (Figs 2 and 4). The influence of the Cu and Zr content on the δ' precipitation has been discussed in detail in previous publications [6–8, 9, 12]. For this, Li–Cu–v and Zr–Cu–v triple complexes were proposed to explain the observed effects. For example, Zr–v and Zr–Cu–v complexes may help in the nucleation of β' dispersoids (Fig. 3(b and c)) which in turn also act as nucleation sites for δ'/β' duplex precipitates (Fig. 3(d and e, g and h)). Since the mobility of the Zr–Cu–v triple complexes is less than that of Zr–v binary complexes, the Zr–Cu–v complexes act as nucleation sites and therefore cause the formation of numerous fine δ' and β' precipitates (Fig. 3(a and b)) on which δ'/β' duplex precipitates form. Uniformly distributed small, spherical δ' particles, non-uniformly distributed δ'-coated β' particles (i.e., $\delta' + \beta'$ composite precipitates) have an effect on the FT value. This is because this property is highly influenced by the size distribution of precipitate particles of the relevant phase.

5. Conclusion

As-cast structure of the Al–2.3 Li–1.8 Mg–0.15 Zr alloy with 0.1 and 0.45 wt % Cu contain fine, equiaxed grains of an ~70 μm size. An increase in the Cu content results in a ternary eutectic containing the T_2 phase at the interdendritic regions. The correct homogenization treatment produces a fairly uniform structure without any segregated phases. Small additions of Cu at the level of 0.1 wt % together with 0.15 wt % Zr affect (i) the δ' precipitate nucleation, (ii) their size and size distribution through Li–Cu–v triple complexes and (iii) in the β' dispersoid nucleation through Zr–Cu–v triple complexes which in turn also affects the size and size distribution of the δ'/β' duplex precipitates. The size and size distribution of both δ' and δ'/β' precipitates have an effect on the fracture toughness of the Al–2.3 Li–1.8 Mg–0.15 Zr alloy. Low additions of Cu, combined with 0.15 wt % Zr, increased the fracture toughness values of the alloys studied in this work that were underaged artificially at 190 °C after proper casting, homogenization, extrusion and solutionizing treatments. The fracture mode of the alloys can be changed from ductile to brittle, regardless of the Cu content, by moving from artificially underaged material to artificially overaged material.

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