Edited by J.F. Nie, A.J. Morton and B.C. Muddle

Influence of Magnesium and Copper Additions on the Low Temperature Thermal Stability of Al-Li Alloys

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Keywords: Al-Li-Mg, Al-Li-Cu, thermal stability, strength, toughness, DSC, electrical resistivity.

Abstract

Separate magnesium and copper additions have been made to a base Al-1.7 wt% Li alloy to investigate their effect on the low temperature stability (70°C) of the microstructure when in the lightly-aged condition. Increasing concentrations of magnesium and copper produce increasing instability by encouraging larger volume fractions of δ´ phase to form during exposure. This enhanced δ´ formation is accompanied by an increase in proof stress and a decrease in fracture energy, the effect being larger in Al1.7LiXCu alloys than in Al1.7LiXMg alloys. Changes in the δ´ volume fraction are the result of a shift in the α/δ´ solvus boundary in Al1.7LiXMg alloys, and heterogeneous nucleation of δ´ on GP_{cu} zones in the case of Al1.7LiXCu alloys.

1. Introduction

Aluminium alloys based on Al-Li respond to age hardening and develop their strength principally from the precipitation of small spherical particles of δ´ (Al_3Li). In order to achieve a satisfactory combination of strength and ductility this type of alloy is often underaged to below peak strength (e.g. 24h at 150°C). In this condition, exposure of the alloy to service temperatures of 50-100°C produce a slow increase in proof stress and a gradual decrease in fracture energy [1]. This instability of the microstructure needs to be accounted for by appropriate design procedures in the aerospace industry.

Various explanations for the low temperature instability of 8090 (Al-Li-Cu-Mg) alloys have been proposed. These include, the segregation of lithium atoms to grain boundaries [2-3], precipitation of fine δ´ dispersions in the matrix [4-5], and formation of GP zones [6]. To isolate the factors that cause instability, work has been undertaken on binary Al-Li alloys. It was shown that the principal microstructural instability at low temperatures (70-100°C) was the continued precipitation of fine δ´ [7]. The present work stems from this study; separate additions of magnesium and copper have been made to Al-Li alloys in order to investigate whether these additions influence the low temperature stability of the microstructure. The concentrations of magnesium and copper studied include those close to 1420 (Al-Li-Mg) and 2090 (Al-Li-Cu-Cu) alloys. Some work is reported in the literature on the low temperature stability of Al-Li-Mg alloys [8] and Al-Li-Cu alloys [9-10], but no systematic approach looking at a range of magnesium and copper concentrations exists. This has been the focus of the present work.

2. Experimental

All the alloys studied contained 1.7±0.05wt%Li, 0.06-0.07wt%Zr, together with separate additions of 1.2wt%Mg, 3.0wt%Mg, 1.2wt%Cu and 3.0wt%Cu. The alloys were prepared by induction melting under argon, cast, homogenised, extruded to bar, hot rolled and finally cold rolled to 1.6mm strip. After solution treatment all alloys had a fine grained equiaxed and recrystallised microstructure. Ageing was carried out in silicone oil baths for 24h at 150°C. This was followed by ageing at 70°C for up to 1000h to simulate service conditions.

Differential scanning calorimeter (DSC) determinations were performed in a Perkin Elmer DSC7 instrument using a heating rate of 20 deg/min. Transmission electron microscopy (TEM) was carried out in a Jeol 2000 FXII, using dark field imaging with 001 superlattice reflections. Electrical resistivity measurements were made in liquid nitrogen using a standard four-probe potentiometric technique. The Kahn tear test was used to measure fracture energy.

3. Mechanical Properties

A standard age of 24h at 150°C resulted in a uniform dispersion of spherical δ' precipitates in all the alloys. Additionally, in the 1.7Li3.0Cu alloy coarse T_1 (Al_2CuLi) plates and a fine dispersion of GP_{cu} zones were also present. Exposing the aged alloys for times up to 1000h at 70°C caused the formation of further fine δ' precipitation. The development of this δ' resulted in an increase in proof stress and a decrease in fracture energy, as shown in Figure1 and Figure 2. The effect of copper on the mechanical property changes during exposure are larger than those produced by magnesium additions. The decrease in fracture energy observed in both alloys is accompanied by an increasingly brittle fracture surface as revealed by SEM.

4. Microstructural Characteristics

TEM showed a bimodal distribution of δ' in both 1.7LiXMg and 1.7LiXCu alloys after the exposure treatment. Coarse δ' of radius 6-7 nm was present as a result of the standard age of 24h at 150°C. Fine δ' was also present as a result of precipitation during exposure. In all alloys, the size of the exposure δ' was 1-2 nm and therefore difficult to measure its volume fraction by TEM. Further characterisation of the exposure δ' was therefore undertaken by DSC, particularly with regard to the measurement of volume fraction. A typical DSC plot before and after exposure of the 1.7Li3.0Mg alloy is shown in Figure 3. A single endothermic peak is observed and this represents the dissolution of both the coarse δ' (from the standard age at 150°C) and the fine δ' (from 1000h exposure at 70°C). Subtracting the enthalpy value (ΔH) obtained after ageing at 150°C from the ΔH value obtained after exposure gives the ΔH value arising from exposure precipitates only. Calibration of ΔH values from known volume fractions of δ' phase enables the volume fraction of δ' precipitated during exposure to be found. These calculated values for 1.7LiXMg alloys are given in Figure 4. Increasing the magnesium concentration from 0 to 3% has increased the volume fraction of exposure δ' from 0 to 0.03.

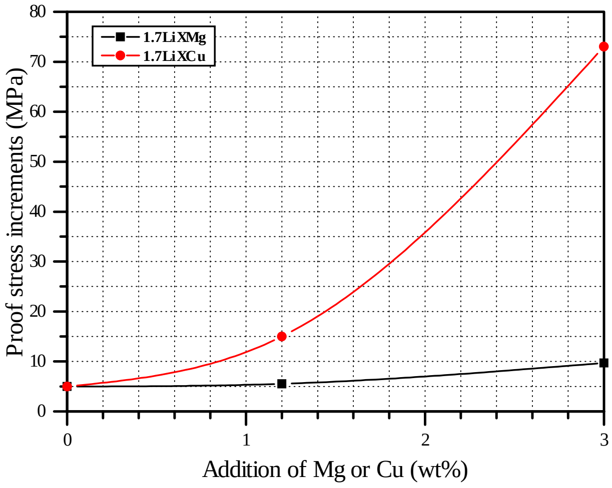

Figure1: Effect of exposure at 70°C on the proof stress of 1.7LiXMg and 1.7LiXCu alloys.

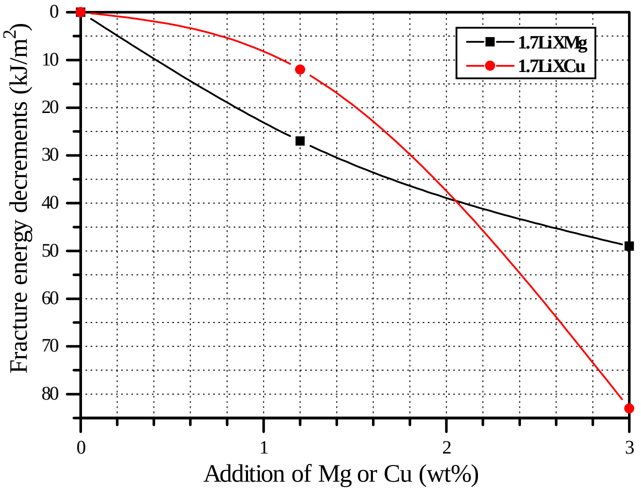

Figure2: Effect of exposure at 70°C on the fracture energy of 1.7LiXMg and 1.7LiXCu alloys.

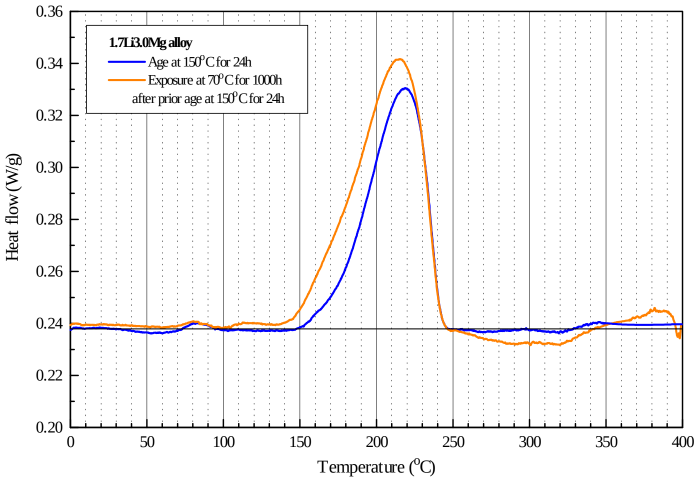

Figure 3: Effect of 1000h exposure at 70°C on the DSC characteristics of the 1.7Li3.0Mg alloy.

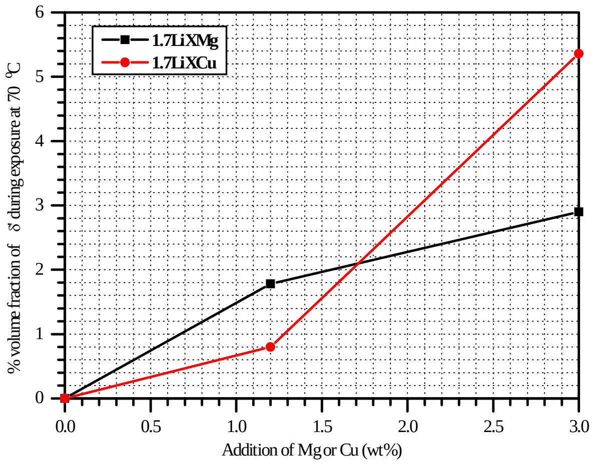

Figure 4: Volume fraction of δ´ produced by 1000h exposure at 70°C in alloys with different magnesium and copper concentrations.

A similar DSC procedure to calculate the volume fraction of exposure δ´ was used for 1.7Li and the 1.7Li1.2Cu alloys and these are also plotted on Figure 4. In the case of the

1.7Li3.0Cu alloy the DSC plot was complicated by the formation of both GP_{cu} zones and δ´ during exposure. The dissolution peak from the zones overlaps with the dissolution peak from the δ´ (Figure 5). However, by ageing a binary Al-3.0%Cu alloy for 24h at 150°C followed by 1000h at 70°C the ΔH contribution from the GP_{cu} zones could be estimated, thus enabling the volume fraction of exposure δ´ to be calculated in the 1.7Li3.0Cu alloy; this value is again plotted on Figure 4. It can be seen from Figure4 that the volume fraction of δ´ is slightly lower in the 1.7Li1.2Cu alloy compared to the 1.7Li1.2Mg alloy, but at the 3.0%Cu level the volume fraction of δ´ is significantly larger than that in the 1.7Li3.0Mg alloy.

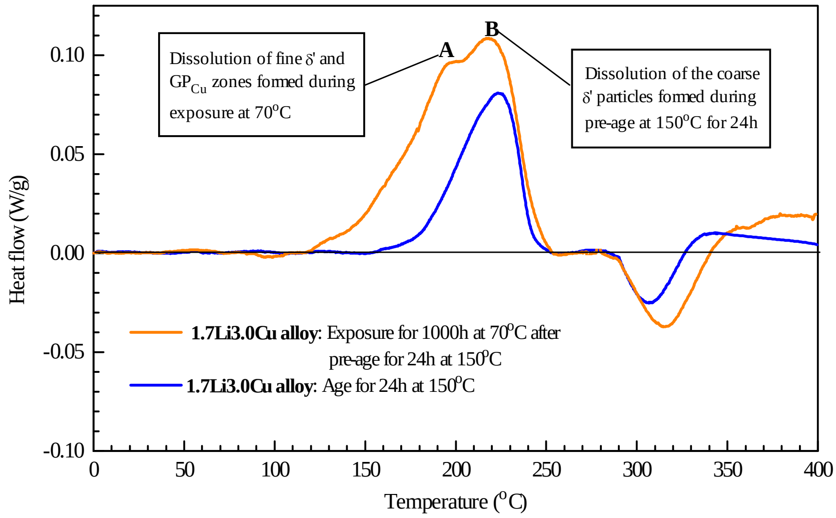

Figure 5: Effect of 1000h exposure at 70°C on the DSC characteristics of the 1.7Li3.0Cu alloy.

5. Electrical Resistivity Measurements

Changes in electrical resistivity have also been monitored during the hold at 70°C. In a binary Al-1.7Li alloy the resistivity increases very slightly during exposure (Figure 6) indicating the formation of either a very small volume fraction of fine (≤ 1nm) δ´ phase or an ordering precursor to δ´, both of which scatter the conduction electrons causing the resistivity to increase. The amount of δ´ formed is not sufficient to affect the proof stress or fracture energy after a 1000h exposure at 70°C (Figures. 1-2). The addition of 1.2 and 3.0% Mg eliminated the resistivity increase; only a smooth decreasing resistivity was observed. This indicated the removal of significant amounts of lithium from solid solution during exposure. This is consistent with the DSC measurements reported in Figure 3 which showed magnesium additions increasing the volume fraction of δ´ precipitated during exposure.

The addition of 1.2% Cu produced electrical resistivity results similar to those from the 1.2% Mg addition, again indicating the removal of significant amounts of lithium from solid solution during exposure (Figure 6). In the 1.7Li3.0Cu alloy a resistivity increase was observed during the early stages of exposure at 70°C. TEM showed this to be caused by the formation of very fine GP_{cu} zones prior to δ´ precipitation.

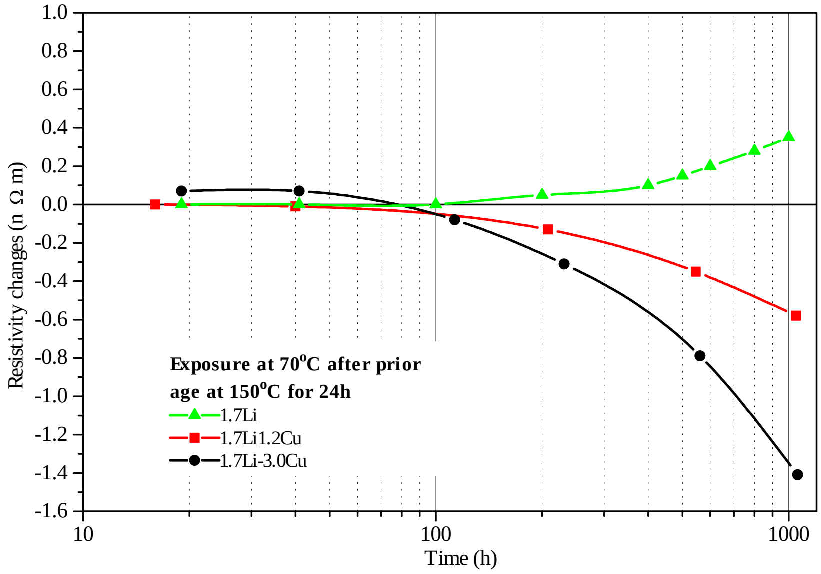

Figure 6: Isothermal resistivity changes in 1.7LiXCu alloys during exposure at 70°C after the standard age at 150°C.

6. Discussion

A 1000h exposure at 70°C of a binary Al-1.7Li alloy produces very little additional δ´ precipitation (Figure 6) and has no effect on the mechanical properties (Figures. 1-2). The presence of separate additions of magnesium or copper increases the amount of δ´ that is precipitated (Figure 4). This increased amount of exposure δ´ causes the proof stress to increase and the fracture energy to decrease. The effect of copper on exposure precipitation is considerably greater than that of magnesium (Figures. 1-2).

Increasing the concentration of magnesium from 0 to 3.0% increases the volume fraction of exposure δ´ because magnesium causes the movement of the α/δ´ solvus boundary to give an increased supersaturation of lithium in aluminium. There is evidence in the literature for such a shift; Baumann and Williams [11] quote an increase in the α/δ´ solvus temperature of 20 deg/wt%Mg, and Valentine and Sanders [12] a value of 5 deg/wt%Mg. Unpublished work by the present authors has produced a value of 7 deg/wt%Mg.

Increasing the concentration of copper in Al-1.7Li from 0 to 3.0% increases the volume fraction of δ´ precipitated during a 1000h exposure at 70°C from 0 to 0.055. This increase is not related to movement of the α/δ´ solvus boundary as copper additions do not affect significantly the position of the boundary [11]. In the 1.7LiXCu alloys exposed at 70°C, in addition to δ´ forming, GP_{cu} zones are also produced (Figure 6). However, it is known that GP zones in Al-Cu and Al-Cu-Mg alloys when aged at 150°C and exposed at 70°C produce little change in properties during the exposure period. The observed changes must therefore be largely due to the additional δ´ phase that is forming.

Recent work [13] on a 1.6Li3.2Cu alloy aged at 200°C has shown that the δ´ phase nucleates and grows on the flanks of GP_{cu} zones. We too, have observed this effect in the 1.7Li3.0Cu alloy aged at the lower temperature of 100°C. It is therefore likely that similar nucleation events take place at 70°C. Note that GP_{cu} zones formed at 70°C are very small making it difficult to observe the effect at this temperature. The result of this would be to provide nucleation sites for the δ´ thus causing the enhanced δ´ precipitation during exposure (Figure 4) and a more defined bimodal δ' distribution (Figure 5). This explanation is supported further by Figure 6 and TEM observations which shows GP_{cu} zones form before δ' precipitation during exposure.

7. Conclusions

- All alloys in this study have been given a standard age of 24h at 150°C followed by exposure for up to 1000h at 70°C. Under these conditions binary Al-1.7Li alloys do not change their mechanical properties during exposure.
- Addition of magnesium to Al-1.7Li alloys results in small increases in proof stress and a decrease in fracture energy. This is caused by an increase in the volume fraction of δ' that precipitates during exposure, resulting from a shift of the α/δ' solvus boundary.
- Addition of copper to Al-1.7Li alloys results in larger changes of mechanical properties during exposure when compared to Al1.7LiXMg alloys. This again is caused by an increased volume fraction of exposure δ' but in this case it is the result of GP_{cu} zones acting as heterogeneous nucleation sites for the δ' phase.

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