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Early Stage Hardening in Al-Cu-Mg Alloys

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Abstract

The early stage age hardening behaviour in Al-Cu-Mg alloys with fixed Mg content (1.7at.%) and varying amounts of Cu has been studied. Two alloys that do and do not exhibit early stage hardening were selected for studying solute-dislocation interactions based on the Portevin Le Chatelier effect. The differences between the nature of serrations of these two alloys in the quenched as well as artificially aged stages are discussed. While the nature of serrations was found to be similar in the as-quenched alloys (rapidly hardening Al-0.8Cu-1.7Mg and non-rapidly hardening Al-0.2Cu-1.7Mg), it changed upon ageing in the rapidly hardening alloy. The possible causes for the observed differences in the nature of serrations are discussed.

1. Introduction

The phenomenon of initial rapid hardening (appreciable increase in hardening for short periods of artificial ageing at median temperatures e.g. 150°C) in certain Al-Cu-Mg alloys has drawn the attention of researchers worldwide and the causes for the observed effect is still being debated [1-4]. This behaviour is not observed in the ternary Al-1.7Cu-0.3Mg (at.%) alloy yet it is observed in the Al-1.1Cu-1.7Mg alloy [1,2], indicating its strong dependence on composition. Although, rapid hardening has been attributed to interaction of dislocations with GP/GPB zones [5], and because these features are present at both these ternary compositions [1], it is not clear what causes the rapid increase in hardening for such a short ageing time (a minute or less). Recent studies proposed alternative mechanisms to account for this effect [3,6,7]. Based on one- dimensional atom probe studies [8], it was proposed that interaction of dislocations with solute clusters may cause this effect. However, the presence of solute clusters was questioned when three dimensional atom probe studies failed to provide clear evidence for clustering [3]. By introducing a deformation step immediately following short time ageing, the same authors [3] were able to show that, the hardness increment due to work hardening was retained, and attributed this to enhanced solute dislocation interactions arising because of the presence of freshly introduced dislocations. Recently, Nagai et. al. [9] by using positron annihilation have reported the presence of Cu-Mg-vacancy complexes and have proposed that interactions of these complexes with dislocations is the cause for rapid initial hardening.

The above discussion shows that the phenomenon is composition dependent and that it may have its origin in solute-dislocation interactions. However, its dependence on composition and possible differences in solute-dislocation interactions between alloys that do and do not exhibit this phenomenon has not been made. An attempt has been made to ascertain these factors in the present study.

The Portevin Le Chatelier (PLC) effect, which manifests itself as serrations on a stress-strain curve, is used to study solute-dislocation interactions [10]. At low strain rates, the velocity of dislocations is comparable to the diffusivity of solute atoms. Whenever a dislocation is trapped (or locked) by solutes, an extra amount of stress is required for its unlocking. Following unlocking, the stress level drops immediately, giving rise to serrations. The objective of the present work is to see whether or not any differences could be seen in the PLC effect between alloys that do and do not exhibit the rapid hardening phenomenon.

2. Experimental Procedure

Alloys with nominal compositions Al-xCu-1.7Mg (x=0.2, 0.5, 0.8,1.0 and 1.1) (at.%) were solution treated in a salt bath for 1 h at 525°C, quenched rapidly into cold water and then aged in an oil bath maintained at 150°C. The Vickers hardness of these alloys in the as-quenched state and after ageing for various periods of time was measured using a 5 kg load. Two alloys that do and do not exhibit rapid hardening were selected for further verification. The microstructural analysis of these alloys was carried out with a Philips CM20 transmission electron microscope operating at 200 kV using the bright field imaging technique. Tensile testing was carried out at a strain rate of $0.2s^{-1}$ to study the solute-dislocation interactions by the PLC effect and to highlight any differences in serrations between the as-quenched and aged specimens.

3. Results and Discussion

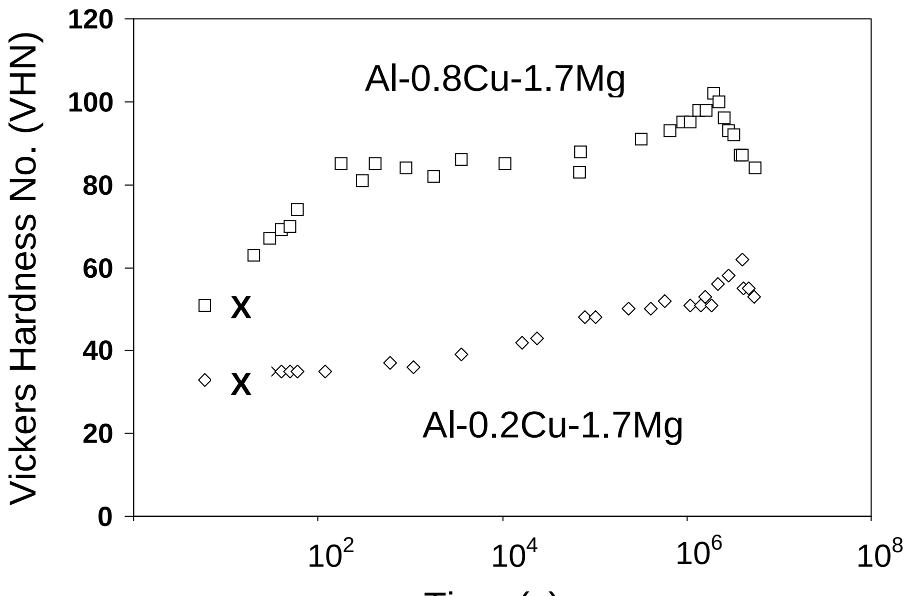

Figure 1: Age hardening behaviour of Al-0.2Cu-1.7Mg and Al-0.8Cu-1.7Mg alloys when artificially aged at 150°C after quenching from solution treatment. The hardness immediately after quenching is denoted by **X**.

Figure 1 shows the age hardening behaviour of Al-0.2Cu-1.7Mg and Al-0.8Cu-1.7Mg alloys. The hardness values shown correspond to an average of ten readings carried out for a particular ageing time. Although, a set of five alloys were artificially aged and their hardening behaviour studied, only the hardening behaviour of two alloys is shown in Figure 1. These alloys have been selected for further studies, as their early stage age hardening behaviour is markedly different. The alloy with Cu content 0.2% does not exhibit the rapid hardening phenomenon, but a gradual increase to peak hardness with continued artificial ageing. On the other hand the alloy with Cu content 0.8% exhibited the rapid hardening effect. For the rapid hardening alloy, there is a sharp increase in hardness for ageing time up to 300s. This is followed by a long period of ageing where the hardness is found to be constant. At longer ageing times a second hardening peak is observed at the end of this plateau. This hardness peak is similar to that observed in the lower Cu content alloy as well, and suggests that the origins of this hardness peak may be due to precipitation in both the alloys.

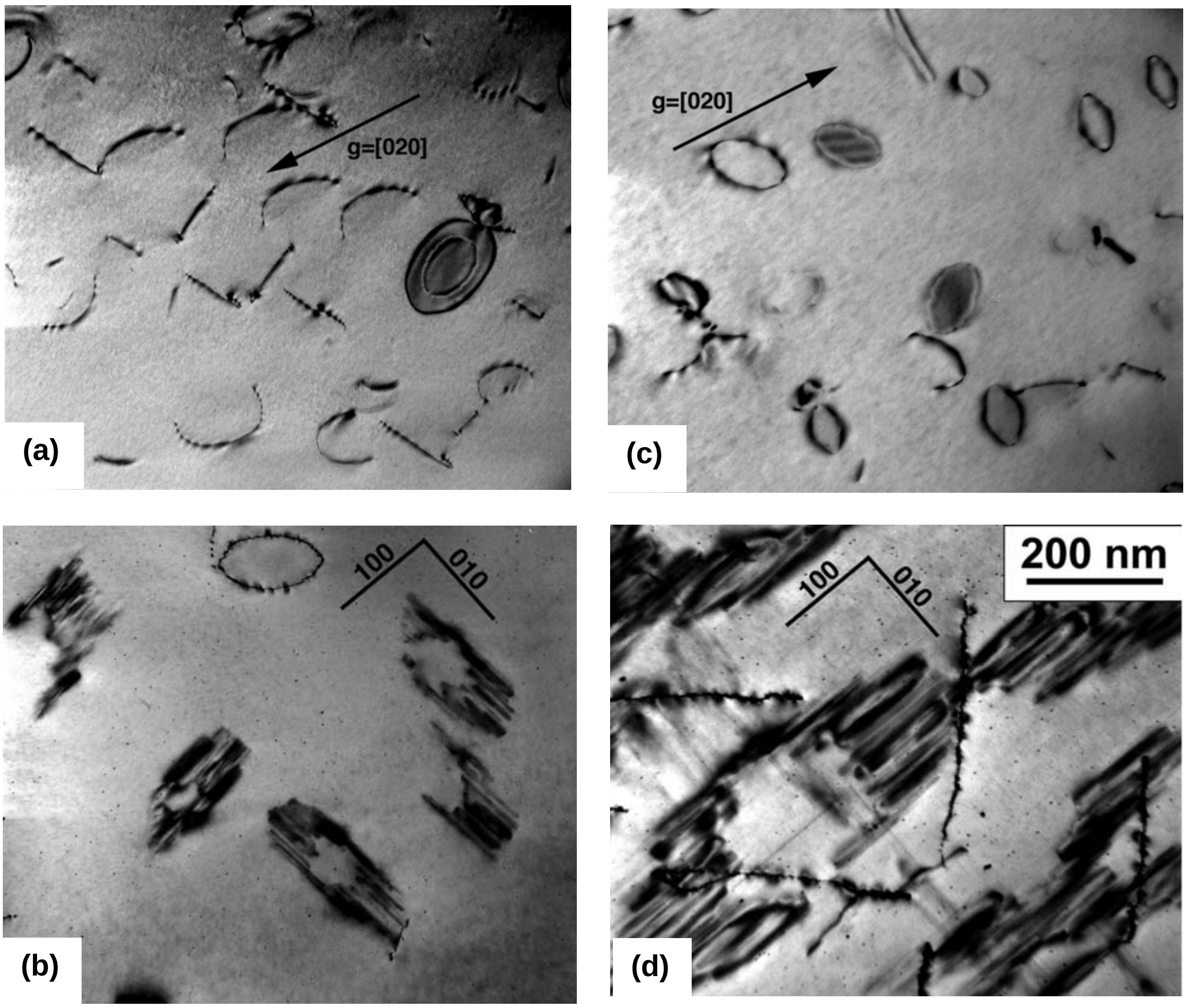

Figure 2: Bright field electron micrographs from Al-0.2Cu-1.7Mg (a&b) and Al-0.8Cu-1.7Mg (c&d) showing the microstructures after quenching (a&c) and after peak ageing (b&d).

It is appropriate to point out here that in an earlier study [11], Cu concentration was maintained at 1.1% and the content of Mg was varied. Rapid hardening was observed for Mg content either equal to or greater than 0.5%. No early stage hardening was observed in the Al-1.1Cu-0.2Mg alloy. The results of age hardening behaviour in the present study as well as the earlier study show that the rapid hardening effect is composition dependent, and that this dependency is not only on the mere presence of both Cu and Mg in Al, but a sufficient amount of each is necessary.

Figure 2 shows bright field transmission electron micrographs of 0.2%Cu (Figure 2a&b) and 0.8% Cu (Figure 2c&d) alloys after ageing at 150°C for 60s and to peak hardness. The electron micrographs from both alloys after 60s ageing do not show precipitates and exhibit mainly dislocation loops. Two different types of loops could be identified from the micrographs (Figure 2a&c). In the 0.8%Cu alloy, only one variety of loop without stacking fault contrast and having a Burgers vector of a<110>/2 was found. In addition to these loops, the 0.2%Cu alloy showed the presence of dislocation loops with stacking fault contrast and a characteristic Burgers vector of a<111>/3. The presence of these loops in the low Cu:Mg alloy points to the migration of vacancies to $\{111\}_{Al}$ planes. Considering that Mg has a higher affinity for vacancies than Cu and that Mg has a tendency to segregate to $\{111\}_{Al}$ planes [12], it may be inferred that the vacancies that contribute to the formation of loops on $\{111\}_{Al}$ planes were carried there by Mg atoms. The absence of such loops in the presence of higher amounts of Cu indicates that the solute-solute interactions between Cu and Mg prevented the segregation of Mg to $\{111\}_{Al}$ planes. Further, in general the loops of alloys with 0.8%Cu were bigger in size compared to the 0.2% Cu alloy. This may reflect either the retention of a higher number density of vacancies during the quenching process or the release of excess quenched-in vacancies due to enhanced Cu-Mg (solute-solute) interactions. As Mg has more affinity for vacancies than Cu, and since both alloys have the same amount of Mg, it is plausible that the latter mechanism may be operative. On continued ageing at 150°C for periods corresponding to peak hardness, both the alloys exhibited precipitation of the S-phase (Al_2CuMg) at the site of these dislocation loops. Figs. 2b&d clearly show the evidence for the presence of the S-phase.

The occurrence of solute rich precipitates at dislocations after prolonged ageing suggests that during early stage ageing (e.g., 60s), most of the solute atoms and quenched-in vacancies were still in solution with Al. As dislocation loops act as vacancy sinks, and solute atoms have preferential interactions with vacancies, a large fraction of solute atoms have been carried to these sinks by vacancies. This could be inferred from the occurrence of large precipitates (peak-aged condition) at the location of dislocation loops. The solute atoms that remain in solution with Al may act as barriers to the motion of dislocations during early ageing. Hence, solute-dislocation interactions were studied using the Portevin Le Chatelier (PLC) effect.

Figure 3 shows the serrations observed during tensile testing of the two alloys (0.2 and 0.8% Cu) immediately following quenching and after ageing for 60s at 150°C. The serrations of both the as-quenched alloys are similar. These serrations are referred to as "Unlocking" serrations. The frequency of serrations was uniform until fracture occurred. Such uniform serrations indicate that the dislocations encountered barriers that are more or less uniformly distributed. As the specimens have been studied immediately following solution treatment, it is to be expected that the solute atoms were distributed uniformly in the Al matrix.

Further, the stress-strain curve does not exhibit a well-defined yield point and points to a weak interaction between the solute atoms and dislocations.

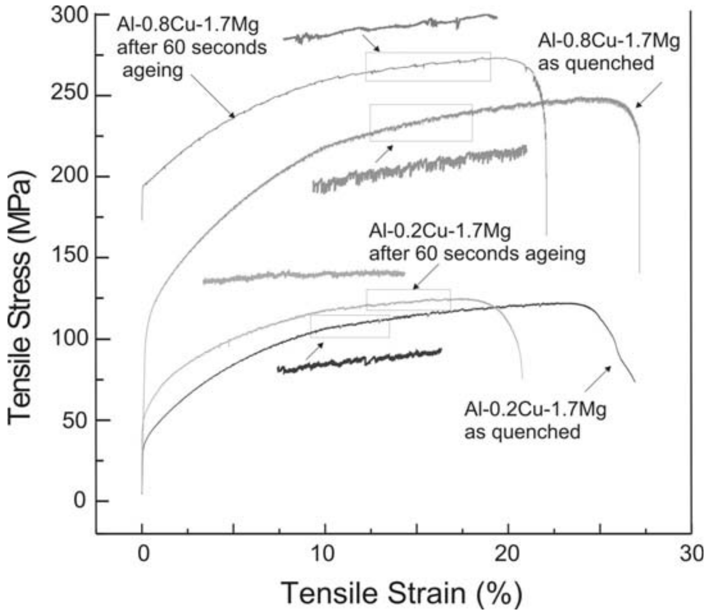

Figure 3: Tensile testing curves showing serrations due to PLC effect. Note the change in serrations of Al-0.8Cu-1.7Mg alloy after ageing.

In contrast to the solution treated specimens, the serrations of aged samples exhibited clear differences. The serrations are again uniform in the alloy with low Cu (Al-0.2Cu-1.7Mg). Interestingly, the serrations of the high Cu alloy (Al-0.8Cu-1.7Mg) are discrete indicating a higher stress was necessary for the unlocking of dislocations. When considered in conjunction with the microstructural evolution, the following observations could be made.

- It appears that in the low Cu alloy the interaction between Cu and Mg atoms was relatively weaker both before and after artificial ageing. (Only interactions between Cu and Mg are mentioned since rapid early hardening is not observed in the binary Al-Cu and Al-Mg alloys). The presence of loops with Burgers vector a<111>/3 is evidence that the interaction between Cu and Mg atoms is weak.
- In the low Cu alloy, no significant change takes place in the distribution of solute atoms even after artificial ageing for a short period of time. This is merely because solute atoms have to diffuse longer distances to form large enough solute aggregates that may act as effective barriers to dislocation motion.
- In the high Cu alloy, absence of loops with Burgers vector a<111>/3 suggests that the movement of Mg atoms is much more restricted. Also, probability of co-clustering of Mg and Cu could be higher due to shorter diffusion distances expected in the presence of higher solute content. This can give rise to larger solute aggregates that may be effective barriers for dislocation motion. However, it could be inferred from the nature of serrations that such aggregates/barriers are absent immediately after quenching and are similar to the low Cu containing alloy.

The serrations of the aged high Cu alloy indicate that the barriers are now more discrete and fewer. This suggests that rearrangement of solute atoms has now taken place. Dislocations moving in the matrix now face a stronger barrier when they come across these larger solute aggregates.

The bowing away mechanism (whereby the dislocation bows around the solute obstacles) is unimaginable in the presence of a high number density of solute aggregates, and as such any dislocation passing through has to cut through. As the cutting mechanism involves breaking and making of bonds, it may not be energetically favourable for breaking a Cu-Mg bond and forming Cu-Al and Mg-Al bonds given that these solute atoms have limited solid solubility in Al. Thus, in the presence of sufficiently larger solute aggregates, dislocations sweeping the matrix may be immobilised and this can lead to the observed rapid hardening effect.

4. Conclusions

Age hardening behaviour and solute-dislocation interaction during early stage ageing has been studied in Al-Cu-Mg alloys. The solute-dislocation interactions remain essentially the same immediately after quenching and after ageing for a short period of time in the Al-0.2Cu-1.7Mg alloy that does not exhibit the phenomenon of initial rapid hardening. On the other hand in the Al-0.8Cu-1.7Mg alloy, the serrations after ageing are markedly different when compared to the as-quenched state. The serrations in the aged alloy are spaced far apart and are larger. This is attributed to larger solute-aggregates that are hard to cut by a moving dislocation and are effective in pinning the dislocations. This may give rise to initial rapid hardening. However, such larger aggregates in the Al-0.2Cu-1.7Mg alloy are absent due to the lower solute content.

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