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LONG-TIME AGING CHARACTERISTICS OF Al-MG-Si COMMERCAIL ALLOY

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ABSTRACT

The influence of long-time aging on hardening precipitates of Al-Mg-Si commercial alloy has been investigated. Samples were heat treated at different aging temperatures ranging from 120 to 200°C. Incase of prolonged aging (from 0.5-200 hours) the hardness vs aging time curve revealed two peaks, a primary peak and a secondary peak during the aging phenomena. An activation energy of 1.37eV was measured in temperature range of 140-180°C in the present study. Transmission electron microscopy study has identified a low density of fine precipitates during the early stage of aging. However, upon prolonged aging, the precipitation hardening process is associated with high density of Mg_2Si precipitates.

INTRODUCTION

Al-Mg-Si alloys are pseudo-binary alloys. These are known to be age-hardenable and studies have been undertaken to understand their aging behavior[1-8]. The combination of properties, such as tensile strength, ductility, formability, weld ability, together with a high corrosion resistance, and an attractive surface appearance render the Al-Mg-Si system very useful for extruded product[9]. These alloys can be safely used at relatively low temperature, but they start loosing their strength when exposed to the high temperature (above 200°C), primarily due to the coarsening of the hardening precipitates[2]. The influence of heat-treatment on the structure of the Mg_2Si precipitates was first studied by Thomas[10]. He discovered that the precipitates have needle-shaped morphology that continuously changes according to the aging conditions. The sequence of precipitates formation is as follows:

Supersaturated Solid solution → Coherent GP zone of needle shape β′ →

Semi-coherent β″ rods → Incoherent β Mg_2Si plates

The formation of GP zone, β′ and β″ precipitates give rise to an appreciable change in the hardness and other properties of the Al-Mg-Si alloys. It is argued that age-hardening in the Al-Mg-Si alloy is controlled by the migration of mobile Mg-Si vacancy cluster, and the activation energy of the process varies from initial to final stage of aging. The activation energy is affected by the concentration of quenched-in vacancies contained in Mg-Si cluster [6-7].

In this paper we report the effect of long-time aging behavior in Al-Mg-Si alloy as a function of time and temperature. Its effect on hardness and microstructure has also been studied.

MATERIALS AND EXPERIMENTAL

The commercial Al-Mg-Si alloy samples 10 x10 x 1mm in size, having 0.54wt%Mg and 0.34wt%Si, were prepared from a long extruded tube, after sectioning. The samples were homogenized at 420°C for 1 hour and were solution-annealed at 547°C for 1 hour followed by rapid quenching in brine, as it exhibits better cooling rate characteristics[11]. The quenched samples were then aged for a pre-determined time and temperature, in the range from 0.5-200 hours and 120-200°C, respectively.

Hardnesses of the aged samples were measured on polished surface, using a Vickers′s hardness tester at a load of 5kg. To minimize the factor of error, each reported hardness-value is the average of five readings. TEM samples were prepared by mechanical grinding of 3mm diameter disc from a thickness of 1 to 0.15mm. The samples were then electrolytically polished in Fischione twin-jet electro-polisher, using an electrolyte of 10% percholoric acid in methanol at a temperature of -15°C. Thin foils were studied under JEOL-200CX transmission electron microscope (TEM) at an operating voltage of 160 keV.

EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows the isothermal linear-log plot of Vicker′s hardness versus aging-time. Initially, the aging was planned at temperatures of 140°C, 160°C and 180°C.

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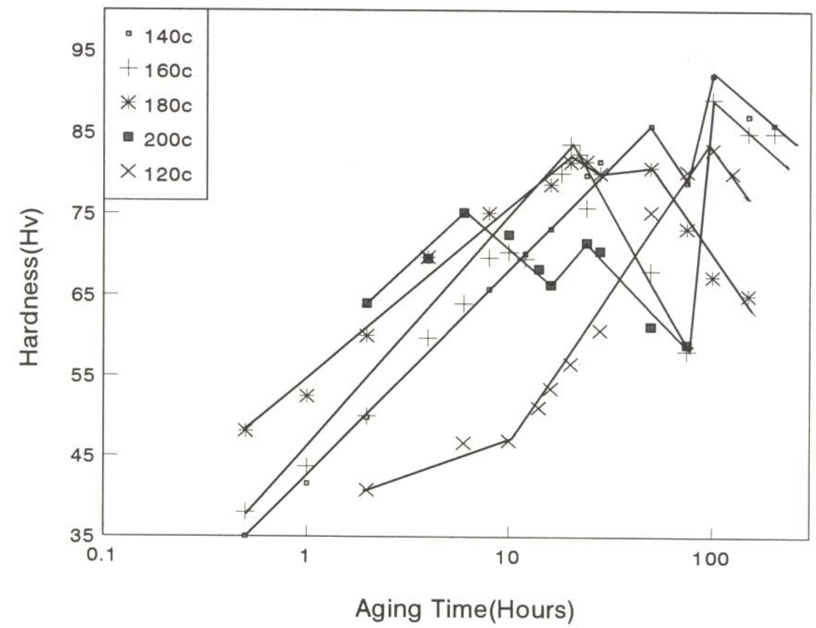

Figure - 1: Hardness (Hv) as function of log aging time (Hours) shows the appearance of primary and secondary peaks.

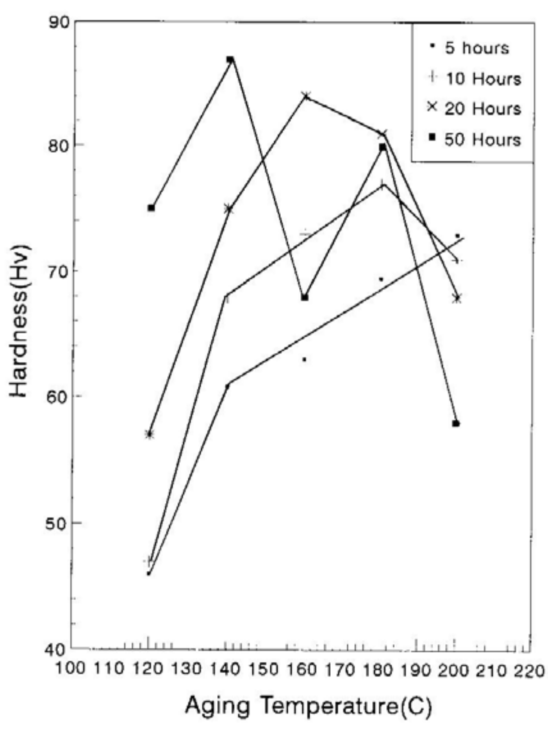

Figure - 2: Hardness (Hv) as function of aging temperature (^{o}C) for fixed time.

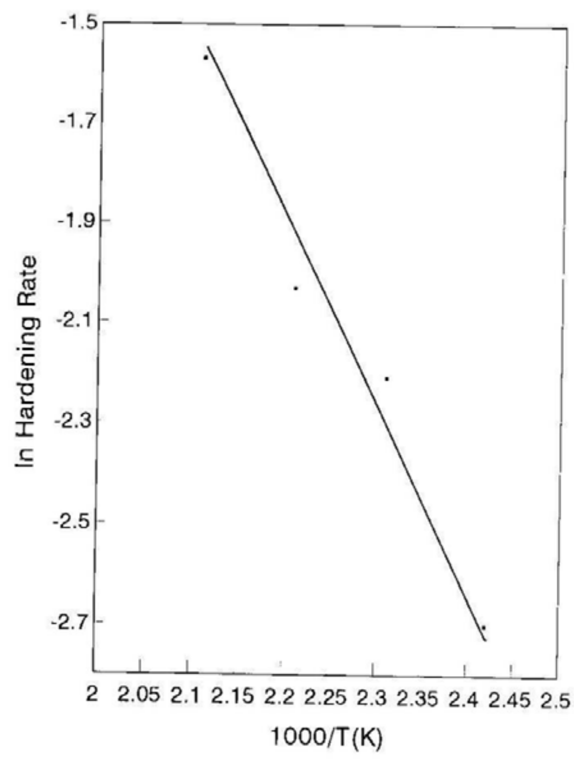

Figure - 3: Arrhenius type of plot of log hardening rte as a function of absolute aging temperature.

The plots show that there is a linear increase in hardness during the early stage of the aging process. After this stage, the hardness drops sharply and then rises again when the alloy is aged further for a longer time. This means, each curve has two hardness peaks, a primary and a secondary peak. The aging curve at 140°C shows the formation of the primary peak around 50 hours, after which the hardness drops and then rises sharply above 100 hours and the secondary peak develops. Similarly, the aging curve at 160°C shows the primary peak around 20 hours of aging and the secondary peak above 100 hours, in analogy to the aging curve at 140°C. Although the difference of time between the two curves to achieve the primary peak is very large, there is only a slight difference in the hardness values. This indicates that growth-rate of precipitate is slower in case of lower temperature.

The sample aged at a temperature of 180°C shows the primary peak after 15 hours, the hardness then slightly decreases and then secondary peak develops above 50 hours. The drop in hardness after the primary peak is not very significant at a temperature of 180°C; however, it follows a trend similar to that at 140°C and 160°C aging temperatures. At higher aging temperature, it takes a shorter time to complete the whole process, as such, primary and secondary peaks for hardness are not developed properly. Following these observations the aging process was repeated at of 120°C and 200°C. The aging phenomena at a temperature of 120°C occurred with two different slopes. The rise in hardness in the first 10 hours is very slow, after which it rises in a normal way as is observed for 140°C, 160°C and 180°C. This further indicates that high activation energy is required for precipitation at low temperatures. In this case, the primary peak was observed after 100 hours of aging. The sample was stopped from further aging after the appearance of primary peak at 120°C, as it was expected to depict a trend similar to that at 140°C, 160°C and 180°C aging temperatures. The aging temperature of 200°C did show the primary and secondary peaks; however, the results at this temperature are not reproducible. Probably reaction kinetics is so high that it is difficult to follow the aging mechanism. Thus it may be concluded that the higher the aging temperature, the lower is the time at which primary and secondary peaks for hardness appears. Furthermore, this aging at 200°C or above is not recommended for these alloys.

Figure 2 shows the plot of Vicker's hardness as a function of aging temperature for fixed time-intervals. Better hardening tendency is shown in the temperature range of 160-180°C and in 10-20 hours after that, the hardness decreases continuously as the aging temperature increases.

The kinetics of age-hardening has been analyzed by employing Arrhenius type of equation of the form:

$$H = H_0 e^{-Q/RT}$$

where H is the hardening rate, H_0 is the structural constant, Q is the activation energy, R is the gas constant and T is the absolute temperature.

Figure 3 is the Arrhenius plot of log hardening-rate as a function of the reciprocal of the absolute aging temperature. The slope of the straight line in figure 3 gives an activation energy of 1.37eV. The aging phenomenon observed in the initial stage of aging in the present study is in agreement with the results of Ismail and Bouchra[7] reported for a similar alloy (Al-Mg-Si). They measured activation energy of 0.95 eV for their alloy, which appears to have better hardening tendency and as a result took shorter time to achieve the maximum peak hardness. Moreover, their peak hardness values are relatively high, which is perhaps the effect of compositional difference of the alloy used in the two studies (0.98wt%Si, 0.78wt%Mg, 0.31wt%Mn and 0.06wt%Fe[7]). However, Ismail and Bouchra have proposed that the low activation energy is probably due to the formation of complexes between quenched-in vacancies and solute atoms. Yanagwa and co-workers have proposed that activation energy for the aging process varies from 0.7-1.1eV for the early stage of aging to 1.3-1.5eV for the later stage. The activation energy is dependent upon the concentration of alloying elements, such as Si and Mg which is a source of precipitates. The value of 1.37eV recorded in the present study is consistent with the result of Ismail and Bouchra and Yanagawa *et al*. It is to be noted that activation energy of 1.43eV and 1.13 eV has also been reported by Hunsikker[14] for diffusion of Si and Mg in aluminum, respectively.

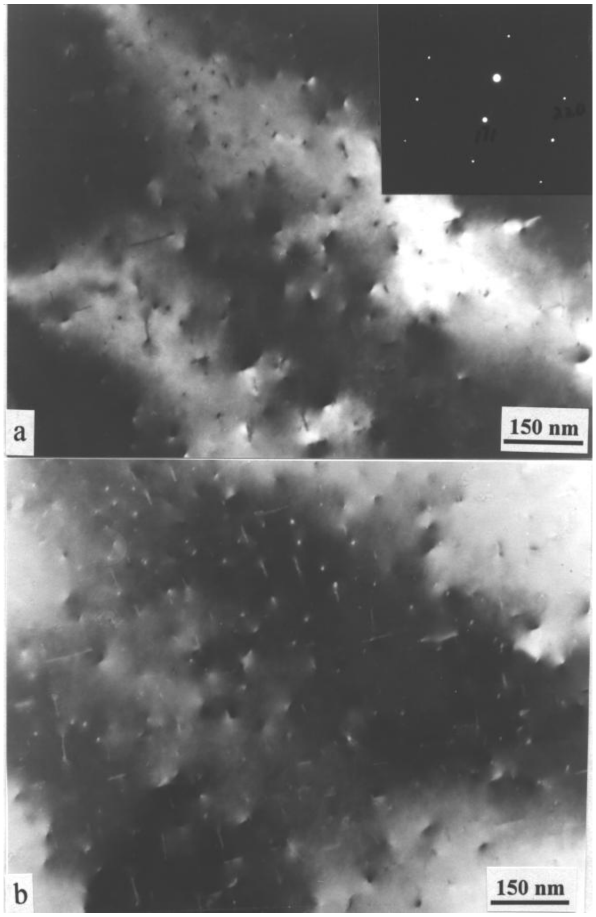

Figure - 4: TEM micrographs show the features of Mg_2Si hardening precipitates in the initial stage of aging process (primary peak) at 160°C after 20 hours. a. Bright Field, b. Dark Field. Diffraction pattern (Zone = [112]) is placed over the micrograph

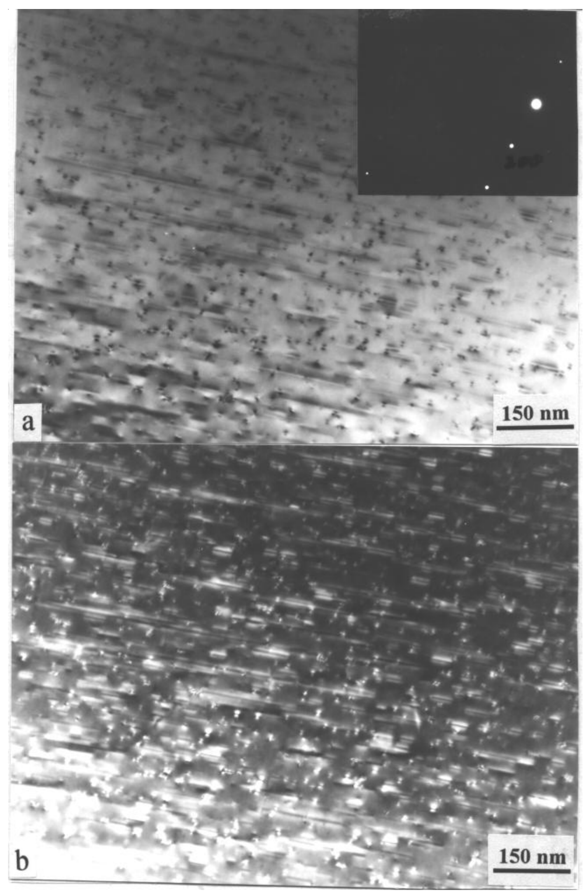

Figure - 5: TEM micrographs show the features of Mg_2Si hardening precipitates at the final stage of aging (secondary peak) at 160°C after 100 hours. a. Bright Field, b. Dark Field. Diffraction Pattern (Zone = [012]) is placed over the micrograph

It is to be noted that, during the whole aging process discussed above, the hardness has higher values incase of secondary peaks as compared to that of the primary peaks (figure 1). The reason for this effect is supported by TEM observation in figure 4 and 5. Figure 4 reveals the feature of needle-shaped β' hardening precipitates at the initial stage of aging. These are related to the primary peak in figure 1. As the aging of the Al-Mg-Si alloy is continuous over longer times, a high density of rod shaped β'' precipitates have appeared, which are dispersed throughout the matrix and the curve approaches to the secondary peak. This is evident in the TEM micrograph, figure 5. The change in the type of Mg_2Si precipitates and increase in their density may be the possible reasons for the rise of hardness at the secondary peak. This type of precipitate β'' was also reported earlier[12-13]. The two peaks (primary and secondary) determined during the aging process are in agreement with the observation reported by Takeda and co-workers[8]. According to them, the peak formed in the later stage of aging is independent of the first peak. This is probably due to the change in the precipitation morphology, as it is observed in the present study. Moreover it was suggested that needle-like Mg_2Si precipitates play a leading role in the aging process. Contrary to this, it is noticed that β'' rods are dominant; however, these β'' rods precipitates have formed after 100 hours of aging. This contradiction may perhaps be attributed to the different chemical composition of the alloy used in this study. The variation in alloy-concentration may be responsible for variation in the aging time interval and in the morphology of the hardening precipitates.

It is established[1] that the precipitation-process in the supersaturated solid-solution can take place in two distinct stages, which may be termed as pre-precipitation and precipitation stages. Pre-precipitation stage is characterized by clustering or segregation of solute atoms in the matrix, known as GP zones. The initial stage of aging (primary peak) can be characterized as pre-precipitation stage, where the clustering of the solute atom is more frequent and the growth of the precipitate is less, as is evident in figure 4. In this context, it will be worth explaining that the shape of the precipitate particles and the nature of the interface, between the precipitate and the matrix are of interest, as these have a great influence on the mechanical properties of the resulting two-phase structure. The interface between the precipitate particles and the matrix can only be fully coherent when it is smaller in area and any misfit must then be accommodated by the elastic strain. If the interface becomes large in area, the strain-energy can be reduced by introduction of structural dislocation lying in the interface. Such an interface is not fully coherent[15]. Similar conditions have been discussed [16-17] for an initially coherent precipitate to become non-coherent, as it grows. It is proposed that an initially coherent precipitate will become non-coherent when it is so large that the elastic energy due to the difference in atomic volume of matrix and the precipitates becomes greater than that of the surface energy.

The same situation of non-coherency might have existed during the growth of the Mg_2Si precipitates, which has led to the drop in hardness after achieving the primary peak, figure 1, in Al-Mg-Si alloy. As the aging continues, the growing precipitate changes its type (morphology) from β' needle to β'' rods, which ultimately appear as a secondary peak with a higher hardness value in the aging process.

CONCLUSION

In this work, Al-0.54%Mg-0.34%Si alloy was studied under a wide range of aging parameters and the following conclusions have been drawn:

1. The alloy under discussion has less hardening tendency and the desired properties are only obtained if it is aged at 160-180°C for 10-20 hours

2. The alloy, during long-time aging, exhibits primary and secondary hardening peaks.

3. In the initial stage of aging, the growth of the Mg_2Si hardening precipitates was very slow; however, as the aging continues, a high density of Mg_2Si, β'' rods appeared. An activation energy Q = 1.37eV measured for the initial stage of aging is consistent with the aging-process reported in the literature.

4. The drop in hardness after achieving the primary peak is probably due to the interface structural misfit (non-coherence) between the precipitate particle and the matrix as it grows.

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