JMEPEG (1998) 7:673-676

Application of Positron Annihilation Spectroscopy to Study Recovery and Recrystallization of Commercial Pure Al and Al-1wt% Mn Alloys

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(Submitted 20 December 1997; in revised form 4 May 1998)

Isochronal annealing of cold worked commercial pure aluminum (99.5%) and Al-1wt% Mn alloys was demonstrated between room temperature and 823 K. The stages of recovery and recrystallization were studied using microhardness and positron annihilation lifetime measurements. A positive correlation was established between the variation of the intensity of long lived component and Vickers microhardness with annealing temperature during the two stages of recovery, which were identified in both aluminum and Al(Mn). The retardation of the recovery stages and recrystallization in Al(Mn) alloys is interpreted in terms of precipitation of aluminum-manganese particles as well as manganese-vacancy interaction.

Keywords aluminum alloys, positron annihilation, recovery, recrystallization

1. Introduction

Commercial pure aluminum and aluminum-manganese alloys are main classes of non heat-treatable alloys. The initial strength of these alloys depends upon the hardening effect of elements such as manganese, silicon, iron, and magnesium, which can be used singly or in various combinations. Because these alloys are work hardenable, further strengthening is made possible by various degrees of cold working. In general these alloys are used when moderate strength combined with high ductility and excellent corrosion resistance is required (Ref 1).

Recovery, recrystallization, and grain growth are the main stages of annealing for cold-worked metal. The earliest change in structure and properties that occurs upon annealing a cold-worked structure is considered the beginning of recovery. As recovery proceeds, a sequence of structural changes emerge such as annihilation and rearrangement of dislocations, subgrain formation, and growth. However, hardness is not greatly sensitive to early stages of recovery. As long as there is a non-equilibrium concentration of point defects, the driving force for recrystallization is either the stored dislocation energy or grain boundary energy (Ref 1). After complete recrystallization, softening will occur, which is revealed through a sudden decrease of hardness. However, hardness measurement is not greatly sensitive to defect structural variations during early stages of recovery.

Positron is, conversely, known to be a probe of high sensitivity to local regions of lower-than-average electron density. This makes the positron annihilation technique (PAT) of particular advantage compared to other traditional tools (electrical resistivity or transmission electron microscopy) in resolving small vacancy clusters, voids, dislocation lines or loops, and jogs (Ref 2). Because of this defect specificity property, positron annihilation lifetime (PAL) is therefore able to differentiate between the earliest stages of vacancy clustering as well as the vacancy recovery stages during isochronal and isothermal annealing (Ref 3-7).

Several investigations have been performed using PAL for studying defects in aluminum and aluminum alloys after quenching (Ref 8), neutron (Ref 9), and electron (Ref 7) irradiation. The recovery processes (Ref 10,11) and precipitation phenomena (Ref 12-14) in various dilute aluminum alloys were explained by using PAL data results.

The isochronal defect recovery in direct chill cast and hot rolled aluminum-manganese (iron, silicon, and copper) industrial alloys, has been studied previously between room temperature (RT) and 893 K by using positron lifetime spectroscopy, where the recovery process was found to depend strongly on the prehistory of the alloy (Ref 15). However, the available literature dealing with recovery and recrystallization phenomena in cold worked aluminum and aluminum alloys is still limited (Ref 16).

Therefore, in the present work, the recovery stages in aluminum and Al-1 wt%Mn after cold rolling were investigated during isochronal annealing in the temperature interval (323 to 823 K) using PAL technique and microhardness measurements. The PAL results were interpreted by the two-state trapping model (Ref 17).

2. Experiment

Table 1 shows the chemical composition of the alloys used in this investigation in weight percent. The specimens were cold rolled (~67% reduction of thickness) then isochronal annealed in air for 1 h in the temperature range 323 to 823 K. The cooling of the samples to room temperature was performed slowly in the oven to avoid quenching effects. The positron lifetime measurements were carried out for the as received

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samples and the annealed samples at room temperature. Positron lifetime spectra were recorded using a plastic fast-fast lifetime spectrometer with a time resolution of 200 picoseconds (ps) for ^{60}Co. Spectra were recorded at a count rate 550 counts per second (cps) with 20 microcuries (μCi) ^{22}Na source deposited on kapton foil then sandwiched between two similar samples, each 1 mm thick. The lifetime spectra were analyzed into three components using the computer program PATFIT-88 (Riso National Laboratory, Roskilde, Denmark). The component characterized by intensity I_1 and lifetime τ_1 represent the positron annihilating in bulk and dislocation loops. The component with I_2 and τ_2 is for the positron trapped and annihilating in monovacancies or voids. The third component with I_3 = 1 to 3% and τ_3 = 2 to 2.6 nanoseconds (ns) might correspond to ortho positronium formed and annihilating at the surface and therefore, will not be discussed. The spectra were corrected for 3% contribution from the source and the variance of fit ranges from 0.99 to 1.2.

Microhardness measurements were performed for all samples using Shimadzu microhardness tester (Vickers) with applied load 50 g for 10 s. Ten readings were taken for each sample, and the standard deviation was calculated.

Table 1 Chemical composition of commercial pure aluminum and aluminum-manganese alloys

Material	Si	Fe	Cu	Mn	Mg	Zn	Ti	Al
Al-99.5%	0.23	0.36	0.05	0.05	0.05	0.05	0.03	bal
Al-Mn	0.22	0.51	0.08	1.10	0.40	0.01	0.013	bal

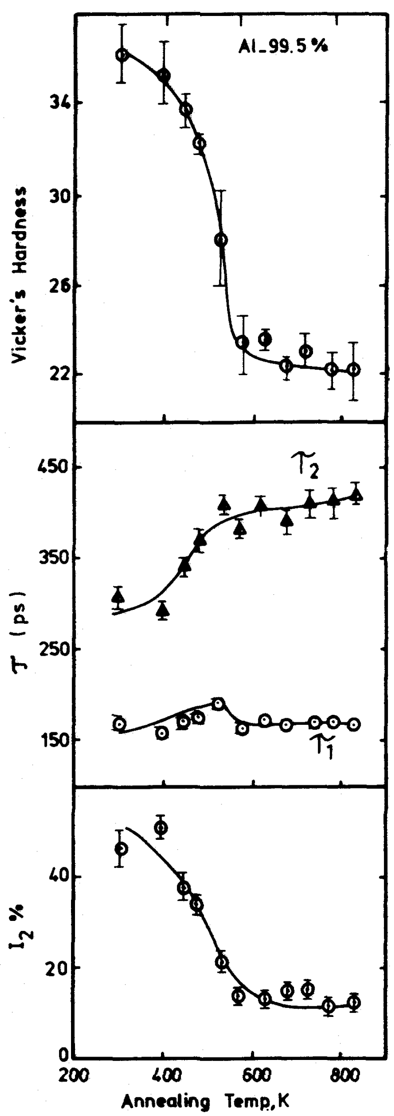

Fig. 1 Microhardness, lifetimes, and intensity, I_2, of positron annihilation versus annealing temperature in Al-99.5%

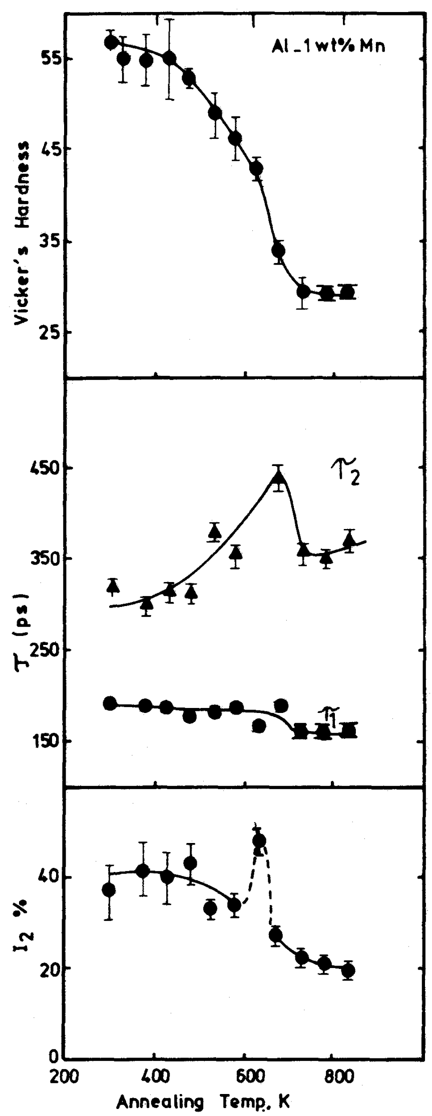

Fig. 2 Microhardness, lifetimes, and intensity, I_2, of positron annihilation versus annealing temperature in Al-1wt% Mn

3. Results and Discussion

Figures 1 and 2 show the temperature dependence of microhardness, life times, τ_1, τ_2 and intensity, I_2, for Al-99.5% and Al-1wt%Mn, respectively. In both samples, the τ_1 value of 190 ps is larger than the positron lifetime $\tau_b = (162 \pm 8)$ ps in bulk (Ref 18). It is therefore believed that the samples contain dislocation loops. Theoretical calculations predict a value of $\tau = 180$ ps for dislocation loops in aluminum (Ref 11). Hautojarvi et al. (Ref 16) found that the positron annihilation in dislocation loops in aluminum gave a lifetime of 230 ps, which supports the results of this study fairly well. These dislocations were found to anneal at 573 and 723 K in pure aluminum and in aluminum-manganese alloys, respectively. At higher temperatures τ_1 decreases to the bulk value of (160 ± 5) ps and remained essentially constant in both samples.

Theoretical calculations were done by Hautojarvi et al. (Ref 19) to establish correlation between the positron lifetime τ_2 in defects and their size. Accordingly the increase in τ_2 with temperature (Fig. 1 and 2) indicates the trapping of positrons to defects larger than monovacancies which would give $\tau_2 = 250$ ps in aluminum (Ref.19) corresponding to a mono vacancy size of ~2 Angstroms (Å). The variation of τ_2 can be explained by a growth of vacancy cluster which saturates at a value of 422 ps, corresponding to a cluster size of ~3Å in pure aluminum (Ref 19). In aluminum-manganese alloy, however, τ_2 continues to increase to a maximum of 488 ps, that is, cluster size of ~4Å, then decreases and saturates at a value of 369 ps. The increase of τ_2 with annealing temperature is combined with the decrease in τ_1 and can be interpreted due to the fact that migrating monovacancies, which are emitted from the loops are absorbed by the vacancy clusters, thus increasing their size and consequently their trapping (Ref 17) probability defined as $K = \mu c_t = I_2(1/\tau_1 + 1/\tau_2)$, where c_t is the trap concentration and μ is the specific trapping rate of positrons. K reaches a maximum at 400 and 600 K in pure aluminum and aluminum-manganese, respectively, as shown in Fig. 3 and 4.

Conversely, the behavior of the hardness with temperature is similar to that of the intensity of the long lifetime I_2 for the two alloys, that is, decreasing with increasing temperature followed by a saturation at 573 K for Al-99.5% and at 723 K for Al-1wt%Mn. The decrease of I_2 with temperature in both alloys means the reduction of lattice defect fractions and occurs at 573 and 723 K for Al-99.5% and Al-1wt%Mn, respectively. This indicates that the presence of manganese in aluminum matrix shifts the recrystallization temperature to higher temperatures, which is in agreement with other results (Ref 1,15) and can be due to precipitation of stable Al_6Mn particles in aluminum-manganese alloys. This process is slow due to the low diffusion coefficient of manganese in aluminum (Ref 15), but is considerably accelerated due to the presence of iron and silicon. The nucleation and growth of some metastable particles may therefore be responsible for the retardation of recrystallization in aluminum-manganese alloys (Ref 15).

According to the I_2 variation with annealing temperature, two stages can be suggested in the defect recovery occurring in Al-99.5% and Al-1wt%Mn alloys. The stages are 300 to 573 K and 573 to 823 K for Al-99.5% and 300 to 723 K and 723 to 823 K for Al-1wt%Mn. In the first stage I_2 decreases monotonically with annealing temperature indicating a decrease in the relative number of voids, which is associated with an increase in τ_2. This behavior can be explained by the migration of vacancies from dislocation and their subsequent clustering to form large size voids. In the second stage I_2 remains constant with increasing annealing temperature indicating complete recrystallization. The retardation of the recovery stages to higher temperatures observed in Al-1wt%Mn is due to retardation of the recrystallization temperature (Ref 20). In addition, the sudden decrease in τ_2 at annealing temperature 673 K in the aluminum-manganese alloy to a value of 369 ps, which is smaller than the corresponding saturation τ_2 value of 422 ps found in Al-99.5%,

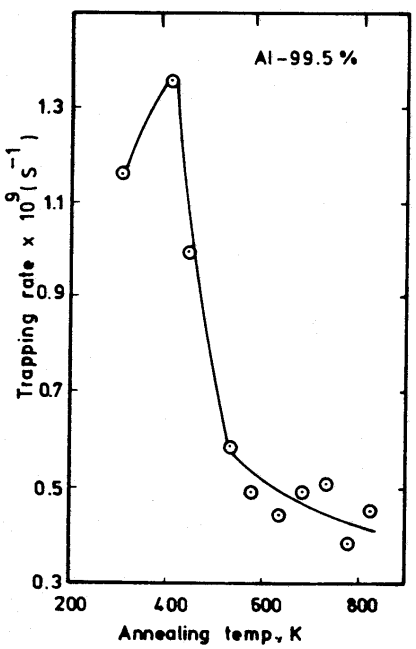

Fig. 3 Trapping rate of positron annihilation versus annealing temperature in Al-99.5%

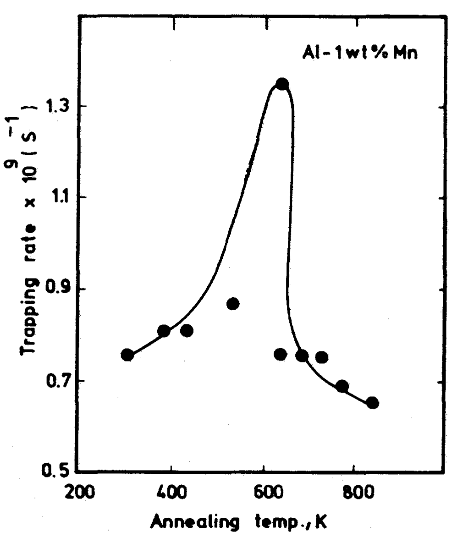

Fig. 4 Trapping rate of positron annihilation versus annealing temperature in Al-1wt%Mn

suggests that the precipitation of Al_6Mn particles may be the reason for the rapid reduction in the size of defects in the aluminum-manganese alloys. This leads to a more defect-free structure compared to Al-99.5%, in agreement with previous work (Ref 15).

The effect of manganese can also be deduced by comparing the variation of the trapping rate (K) of positron versus annealing temperature in Al-99.5% and Al-1wt%Mn, as shown in Fig. 3 and 4, respectively. It can be seen that a sharp increase to a maximum of 1.35×10^9 s^{-1} at ~400 K is observed for Al-99.5%, while slow increase to the same maximum is occurring at a temperature of 623 K in Al-1wt%Mn. Calculations of the difference between the activation energies for self diffusion and the diffusion of manganese in aluminum yielded previously a value of 0.39 eV for the binding energy of a manganese atom to a vacancy in aluminum (Ref 21). The results of the present work (not sufficient to estimate the activation energy) shown in Fig. 4 suggest that manganese-vacancy interactions are the reason for the slow increase of the trapping rate during the initial stages of recovery, compared with the corresponding results for Al-99.5% (Fig. 3). It is expected that the trapping rate will increase with increasing the size of a three-dimensional vacancy cluster. Thus it can be deduced that the manganese atoms enhance the nucleation of vacancy clusters in aluminum which was observed previously for this alloy (Ref 5). A similar enhancement of vacancy clustering by other solutes has been observed in positron annihilation studies in aluminum-lithium based alloys (Ref 10,11,14).

In addition, the manganese interaction with vacancies in aluminum will shift the trapping rate maximum from 400 K in Al-99.5% to 623 K in aluminum-manganese alloys.

4. Conclusions

- From the variation of positron lifetime parameters with annealing temperatures, dislocation loops, as well as vacancy clusters, were identified in both aluminum and Al(Mn) alloys. However, the presence of manganese atoms will enhance the nucleation of vacancy clusters in aluminum.
- The variation of positron trapping rate with annealing temperature, shows a slower increase in Al(Mn) than in aluminum with a maximum of 1.35×10^9 s^{-1} shifted to higher temperature, which can be explained by the manganese-vacancy interactions.
- A similar variation of the intensity I_2 of the long lived component is observed in aluminum and the Al(Mn) alloy leading to two stages of recovery. However, the recrystallization temperature in Al(Mn) is shifted to higher temperature compared to aluminum. Formation and precipitation of Al_6(MnFe) particles may be the reason for retardation of recrystallization in Al(Mn) as well as for the relatively faster size reduction of defects in the second annealing stage as observed from the variation of τ_2 with temperature.
- During the recovery stages, a positive correlation between I_2 and the Vickers microhardness was established. This indicates that the PAL techique is sensitive to precipitation phenomena.
- The hardending of Al and Al(Mn) alloy can be interpreted on the atomic level as taking place via the interaction of vacancies of 2-4Å with dislocations and precipitates.

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