JMEPEG (2003) 12:95-98

Application of Positron Annihilation Spectroscopy to Study the Recovery of Commercial Pure Al and Al-0.96 wt.% Si Alloys

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(Submitted 16 July 2002)

Isochronal annealing of quenched commercial pure Al (99.95%) and Al-0.96 wt.% Si was investigated between room temperature (RT) and 550 °C. The annealing of defects was studied using positron lifetime and Doppler broadening (DB) techniques. The retardation of annealing defects in the two alloys is interpreted in terms of precipitation of impurities and Si atoms, as well as defect properties (size and concentration) in the framework of the two state trapping model.

Keywords Al alloys, Al-0.96 wt.% Si alloys, annealing defects, positron annihilation lifetime spectroscopy (PALS)

1. Introduction

Positron annihilation lifetime spectroscopy (PALS) is a specific technique for the detection of vacancy-like defects in metals and alloys.[1] The combination of this technique with Doppler broadening of annihilation radiation (DBAR) provides additional information on the nearest-neighbor atoms of the defect.[2] DBAR experiments with one or two detectors provide information on the momentum distribution of the annihilation electrons.[3]

Recently, positron annihilation spectroscopy (PAS) studies have been completed on binary Al alloys to measure the activation energies for the migration of solute atom-vacancy complexes,[4,5] as well as to detect vacancy-rich clusters in Al-Mn, Al-Zn-Mg, and Al-Cu-Zn alloys after quenching or after aging at relatively low temperature.[6-9] In addition, the identification of Guinier-Preston (GP) zones in aluminum alloys that are either vacancy free or contain vacancies within their structure has been investigated by positron annihilation (PA).[10] On the other hand, the Doppler broadening (DB) technique is applied to evaluate the microhardness variations during isochronal annealing of Al and Al-Mn alloys,[11] and the positron annihilation lifetime spectroscopy (PALS) technique is used to study the recrystallization and grain growth of sintered Al powder alloy.[12]

As discussed in the previous review, little attention has been given to the study of isochronal defect recovery in Al(Si) alloys. The aim of this work is to study the recovery of lattice defects in commercial pure Al (99.5%) and Al-0.96 wt.% Si alloy during the isochronal annealing from room temperature (RT) to 550 °C after quenching to –196 °C using DBAR and PALS techniques.

2. Experimental Details

The materials used in this investigation were quenched commercial pure Al (99.5%) and Al-0.96 wt.% Si alloys. The samples were purchased from the military factory 63 for nonferrous industries, Cairo, Egypt. These specimens were cut from a commercial rod in the form of discs, 20 mm in diameter and 3 mm thick. One of the disc surfaces was polished to a mirror. The samples were annealed at 500 °C and then quenched to –196 °C. Next, isochronal annealing was performed in steps of 30 °C with a hold time of 30 min at each annealing step from RT to 550 °C, at which the temperature stabilized at the level of ±1 °C. The cooling of the samples to RT was performed slowly in the furnace.

A fast-fast coincidence spectrometer was used for measuring PALS. The positron source used in this investigation was ~20 μCi of ^{22}Na deposited on a thin kapton foil (7 μm) and sandwiched between two identical pieces of each sample. The time calibration was found to be 50 ps per channel, and the time resolution (full-width at half-maximum [FWHM]) was established to be 230 ps for ^{60}Co. The positron lifetime spectra were recorded at RT with integral counts not less than 10^6 counts for each sample. The lifetime spectra were analyzed into two components using the PATFIT program.[13] The component characterized by lifetime τ_1 and intensity I_1 represents positron annihilating in monovacancies and dislocation loops. The components with τ_2 and I_2 characterize positrons trapped and annihilating in three-dimensional vacancies and vacancy clusters. From the data analysis of positron experiments, the average lifetime τ_{av} of the two main components $\tau_{av} = \tau_1 I_1 + \tau_2 I_2$, as well as the bulk lifetime $\tau_b = I_1/\tau_1 + I_2/\tau_2$ and trapping rate $\kappa = I_2/I_1(1/\tau_b - 1/\tau_2)$, were deduced on the basis of the two state trapping model.[1]

The Doppler broadening annihilation line shape S-parameters were measured using a hyperpure germanium detector. The measured FWHM was established to be 1.2 keV at 662 keV of ^{137}Cs. The energy dispersion of the equipment was 49 eV per channel. The number of channels included in the annihilation peak area was 300. The positron source used was ~15 μCi of ^{22}Na deposited on kapton foil, and sandwiched

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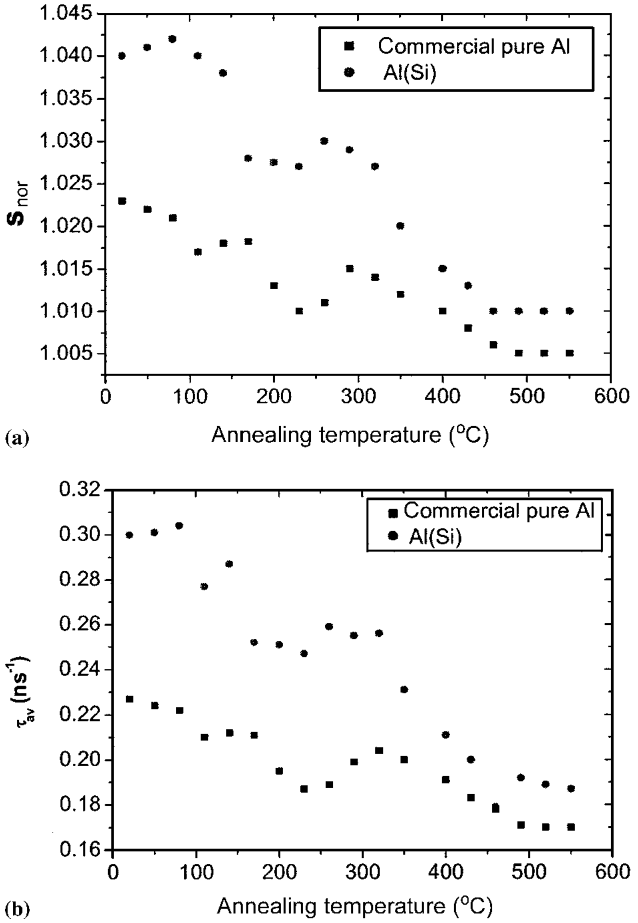

Fig. 1 The variation of **(a)** S_{nor} and **(b)** τ_{av} with annealing temperature in commercial pure Al and Al (Si) alloys

between two identical pieces of the sample. The total number of counts in the measured spectrum was ~10^7. The S-parameters were measured as the number of counts lying within an energy interval of 1.3 keV centered at the peak of the annihilation line. The parameter S_{nor} normalized can be determined from the ratio of S/S_{ref}. S_{ref} was obtained by measuring the line shape distribution using annealed samples of both commercial pure Al and Al(Si) alloys.

3. Results and Discussion

Figure 1(a) and (b) shows the variation of line shape S_{nor} and average lifetime τ_{av} as a function of annealing temperature from RT to 550 °C for commercial pure Al (99.5%) and Al(Si) alloys. The lifetime components τ_1, τ_2, and I_2 are shown in Fig. 2 and 3. For comparison, τ_b is evaluated using the two state trapping model.[1] It is obvious that the S_{nor}, τ_{av}, τ_1, τ_2, and I_2 values in Al(Si) are higher than the values obtained in commercial pure Al because of the formation of small size vacancy clusters. In addition, growth of Si precipitates in Al(Si) alloy results from the rapid quenching of alloys to –196 °C. These results are in agreement with those previously reported for precipitation in quenched Al(Si) alloy.[10] The behavior can be divided into three stages, as illustrated in the figures.

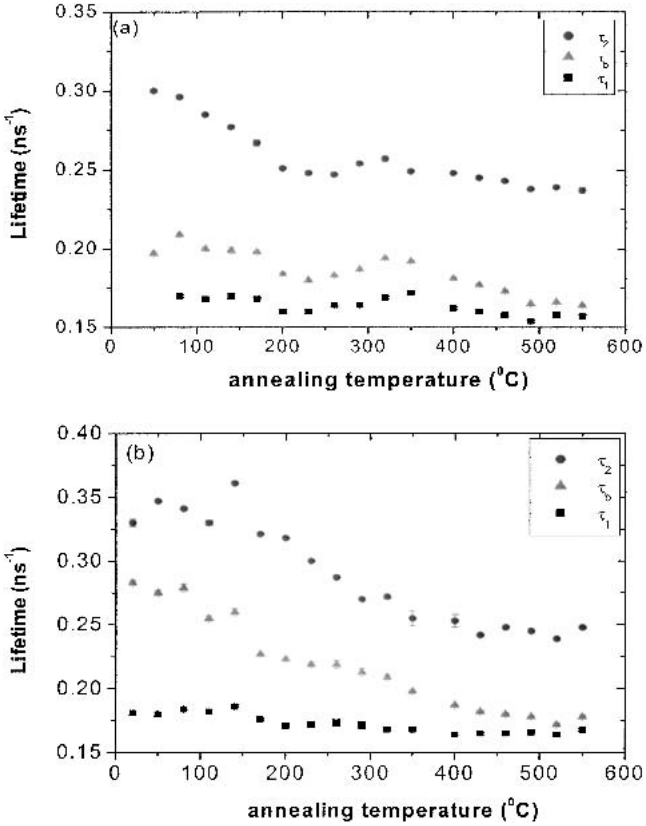

Fig. 2 The variation of lifetime **(a)** in commercial pure Al and **(b)** in Al(Si) alloys with the annealing temperature

The first stage is taken from RT to 200 °C. At the beginning of this stage, little change is observed in S_{nor}, τ_{av}, τ_1, and τ_2 in both alloys up to 80 °C because of the movement of some impurities, which hinders the dislocations and consequently leads to a retardation of recovery. S_{nor} is found to be 1.04 in Al(Si) alloy compared with 1.02 for commercial pure Al. τ_{av} has a value 225 ps in commercial pure Al, which is close to the value of 228 ps that characterizes saturation trapping of positrons at dislocations in pure Al.[9] However, τ_{av} has values around 300 ps in Al(Si) alloy, indicating the formation of small vacancy clusters caused by the presence of Si in Al. The longer lifetime component τ_2 has a value of 290 ps in commercial pure Al, indicating positron trapping at divacancies,[14] whereas an increased τ_2 of 340 ps in Al(Si) corresponds to migration of small clusters and formation of clusters of five vacancies.[3] In commercial pure Al, the τ_1 value is 171 ps; this value is 5 ps above the bulk value of 166 ps,[14] indicating the trapping of the positron at dislocation loops, and increases to reach 180 ps in Al(Si) ascribed to the positron trapped in dislocation loops and small impurity vacancy clusters of two-dimensional character. Above 80 °C, the effect of impurities will decrease owing to their solubility, leading to a decrease in S_{nor}, τ_{av}, τ_1, and τ_2 in both alloys, indicating the beginning of recovery. Note that the annealing of defects occurred at 200 °C; i.e., at $T > 0.33T_m$ in commercial pure Al. This is apparent because S_{nor} approaches the bulk value of 1.010, which is in good agreement with earlier results,[11] reflecting the decrease in defect concentration and change in the defect formation or the atomic configuration around vacancies.[6] On the other hand, τ_{av} decreased to 190 ps in agreement with the value obtained in Ref. 9. Although τ_2 shortened to 250 ps corresponds to the lifetime characteristics for monovacancies in pure Al,[14] τ_1 reached the value of defect-free Al of 166 ps. However, in this stage, I_2 decreased from 80-35%, which means that defect concentration was reduced in both alloys.

The second stage is taken from 200-400 °C. In commercial pure Al, no obvious changes are observed in S_{nor} and lifetime components (τ_{av}, τ_1, τ_2 , I_2), indicating stability in size and concentration of defects. However, in Al(Si) alloy, the S_{nor}, τ_{av}, τ_1, and I_2 values do not change up to the annealing temperature of 320 °C, indicating a saturation in defects concentration caused by the Si content. This saturation is followed by a reduction in S_{nor}, τ_{av}, and I_2, indicating the dissolution of the small vacancy clusters and Si atoms leading to a beginning of annealing defects that are nearly completed at 400 °C; i.e., at $T > 0.5T_m$ of Al. This was apparent when S_{nor} reached 1.01 and τ_{av} reached 211 ps, which is 21 ps over the value of 190 ps obtained in commercial pure Al. On the other hand, τ_2 continued to decrease from 340-250 ps, suggesting the breakup of vacancy clusters and the formation of higher order Si-vacancy complexes such as monovacancies with small size. However, τ_1 reached to the value of defect-free Al (166 ps), and I_2 had a value of 35%. We observed that the annealing of the defect occurred faster (at 200 °C) in commercial pure Al, and shifted to higher temperature (at 400 °C) in Al(Si) alloy because of the stabilizing effect of the Si atoms on the clusters in Al(Si) alloy.[10]

In the third stage from 400-550 °C, a slight decrease in S_{nor}, τ_{av}, τ_1, and τ_2 is observed at the beginning of this stage in commercial pure Al, followed by a saturation until the end of the stage, indicating that the dislocations defects disappear. However, in Al(Si) alloy, S_{nor}, τ_{av}, and τ_2 have the same trend as commercial pure Al, but with higher values, indicating a retention of defects caused by the stabilizing effect of Si atoms on clusters in Al(Si) alloy.[10] On the other hand, I_2 rapidly decreased in both alloys with the same percentage from 35-15%, reflecting that some vacancies were lost to sink during migration,[8] leading to the annealing of defects with the same concentration in both alloys.

Two observations can be made when the measured τ_1 and the calculated τ_b from the two state trapping model are compared.[1] First, a remarkable deviation exists in the first stage of recovery, which increases in Al(Si) alloy and indicates the trapping of positrons at more than one type of defect. Second, the calculated values are close to each other during the annealing temperature (indicating the annealing of defects), which is reached faster in commercial pure Al and shifts to higher temperature in Al(Si) alloy.

To clearly illustrate the above-described results, we plotted the relation between the trapping rate κ and annealing temperature for commercial pure Al and Al(Si) alloy (Fig. 4). κ gives an indication of the number of defects in alloys. The results clearly indicate that κ has a positive correlation with I_2. At RT, κ is found to be 1.825×10^9 s^{-1} and 2.014×10^9 s^{-1} in commercial pure Al and Al(Si), respectively. However, κ is decreased rapidly in commercial pure Al to reach the value of 0.716×10^9 s^{-1} at 200 °C, and reaches almost to the same value in Al(Si) alloy at 400 °C. Above this annealing temperature, κ decreases to 0.300×10^9 s^{-1} in the two alloys, indicating that they annealed with the same number of defects. This confirms the above suggestion that some vacancies were lost to sink during migration in both alloys.

4. Conclusions

- The combined use of positron lifetime and Doppler broadening techniques together with the two state trapping model was sensitive to the annealing of defects in commercial pure Al and Al(Si) alloys.

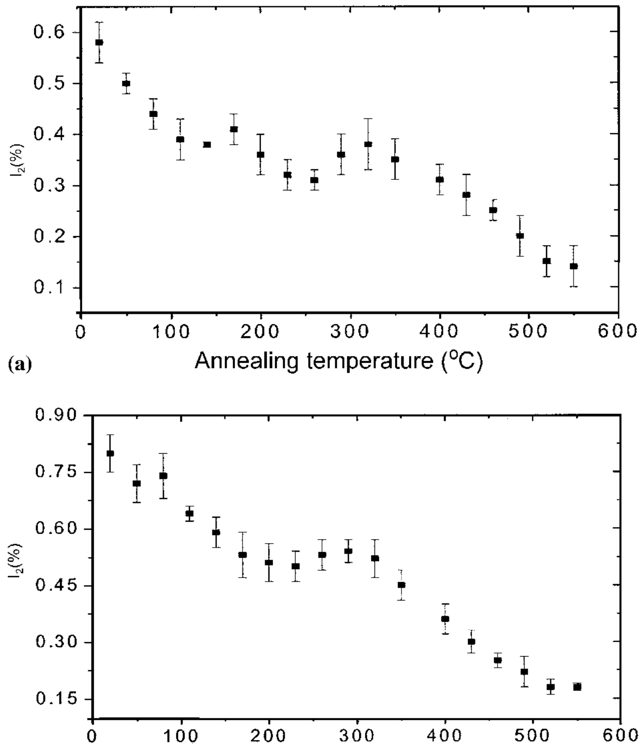

Fig. 3 The variation of I_2 in **(a)** commercial pure Al and **(b)** Al(Si) alloys with the annealing temperature

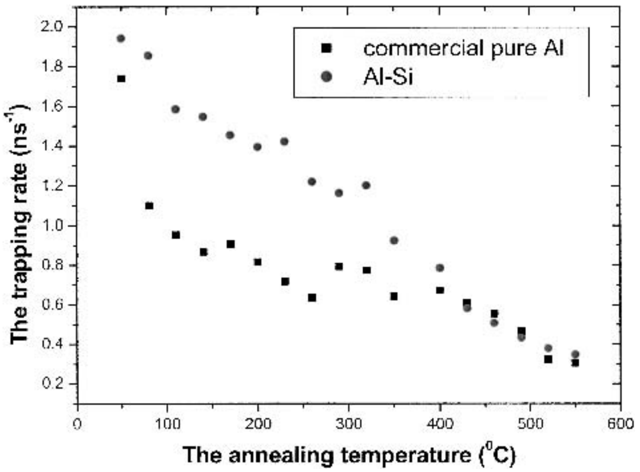

Fig. 4 The variation of trapping rate with annealing temperature in commercial pure Al and Al(Si) alloys

- During the stages of recovery, a positive correlation was observed between S_{nor} and τ_{av}, as well as between I_2 and κ.
- The annealing of defects in Al(Si) alloy were retarded to 400 °C compared with that in commercial pure Al, which appeared at 200 °C. These defects were due to the presence of vacancy-Si complexes, as well as the stabilizing effect of Si atoms on clusters in Al(Si) alloy.
- In Al(Si) alloy, τ_1 approached the bulk lifetime of ~166 ps at the last stage of recovery, whereas in commercial pure Al, τ_1 reached the bulk value at the second stage.
- Through the last stage of recovery, some vacancies in Al(Si) alloy were lost to sink during migration, leading to annealing of defects with the same concentration, which were apparent from the I_2 and κ values.

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