

Application of Doppler Broadening of Annihilation Radiation Technique to Evaluate the Microhardness Variations during Isochronal Annealing of Al and Al (Mn) Alloys

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Doppler broadening measurements have been carried out to study the isochronal annealing of cold-worked commercial pure Al (99.5%) and Al-1 wt.% Mn alloys. The deduced line shape and wing parameters are investigated in the range from room temperature to 823 K and correlated with the corresponding microhardness measurements. The vacancy migration and the effect of the precipitated Al_6 Mn in Al (Mn) alloys could be probed as a function of annealing temperature. Three stages of microstructures can be distinguished in both Al and Al (Mn) alloys, which are recovery, partial recrystallization, and complete recrystallization. The line shape parameter-wing parameter (*S-W*) map indicates the same behavior in both alloys at high temperature. However, at low temperatures, Al (Mn) shows different behavior from the linear trajectory Al alloy.

Keywords Al and Al (Mn) alloys, Doppler broadening, positron annihilation, microhardness, recovery, recrystallization, *S-W* map

1. Introduction

There are many nondestructive methods for the investigation of defects in crystals. One of these methods is the Doppler broadening of annihilation radiation (DBAR) technique, which has been used successfully in metals.[1,2,3] A positron-annihilation Doppler broadening spectrum, which is the energy distribution of the positron 511 keV annihilation line, provides momentum distribution information about the electron with which the positron annihilates.[4] In well-annealed metal samples, the positron annihilates in a delocalized state in the defect-free lattice. In deformed samples, the positron can be trapped at various defects and annihilates from different localized states.[4,5]

The effect of positron trapping can be understood in terms of the overlap of the positron wave function with the conduction and core electrons in the solid.[6] In vacancy-type defects, the average electron density is lower than in the bulk of the material due to the relative absence of core electrons. As a consequence, a positron trapped in a defect has a higher probability to annihilate with a less energetic conduction electron. This results in a narrowing of the annihilation peak. This probability decreases as recovery and recrystallization proceed during annealing of the specimens; hence, the core electrons participate to a greater extent than the conduction electrons.

From the 511 keV annihilation distribution radiation, the line shape parameter S is defined as the ratio of the area C of a fixed central part of the peak to the total area A_0 of the peak, as illustrated in Fig. 1. The parameter S corresponds to the positron annihilation with low momentum valence electrons.[5] The wing parameter W that is defined as the ratio of the wing areas $A + E$ to the total area A_0 corresponds to positron annihilation with high momentum core electrons. The positrons have a preference to occupy the open-volume defects such as vacancies, clusters of vacancies, or voids in the system.[3] In the relatively large open-volume defects, the annihilation process takes place mainly with the low momentum valence electrons, which results in a narrowing of the peak, giving rise to a large S and a small W value. With decreasing defect volume, the positron will become more enclosed, which causes a stronger interaction with the core electrons leading to a broadening of the annihilation peak. This results in a smaller S and a larger W value, which means that S increases and W decreases with the volume size of the defect.

In the present work, the DBAR technique has been used to study the recovery stages after cold-worked and isochronal annealing in commercial pure Al and in Al-1 wt.% Mn alloy as a function of annealing temperature. Previous work on these alloys has been done by Hood and Schultz,[7] who studied the recovery of quenched single crystals of Al and Al 1.5×10^{-2} at.% Mn by the DBAR technique. They concluded that S exhibits a strong positive temperature dependence from the trapped state for both studying samples. However, the probability of positron annihilation with core electrons (W) did not show any changes for the two materials, which could not be interpreted owing to the limited temperature used in this investigation. Accordingly, in the present work, the S and W parameters deduced for Al and Al (Mn) have been investigated as a function of isochronal annealing from room temperature to 823 K. In addition, the results of the line shape parameters are also compared to the microhardness measurements.

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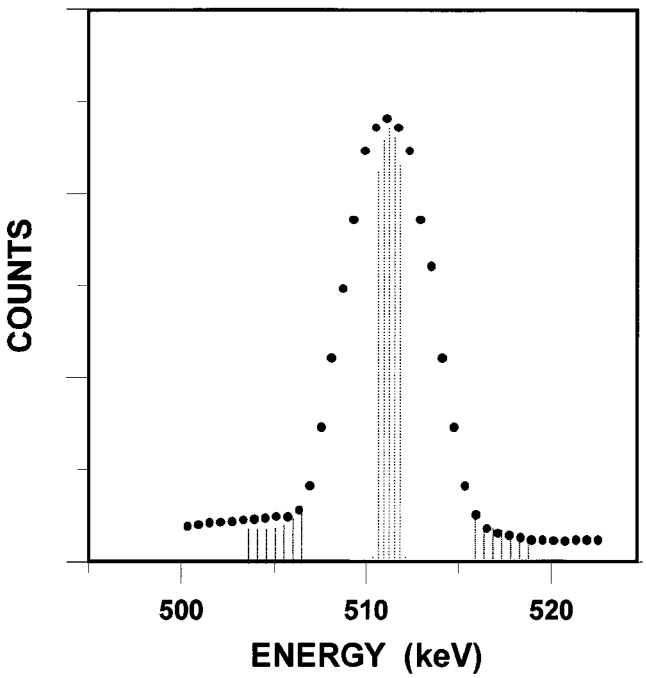

Fig. 1 Line shape parameters S and W of the 511 keV annihilation gamma ray. $S = C/A_0$, and $W = (A + E)/A_0$. A_0: total integral area under the photopeak ($A_0 = A + B + C + D + E$)

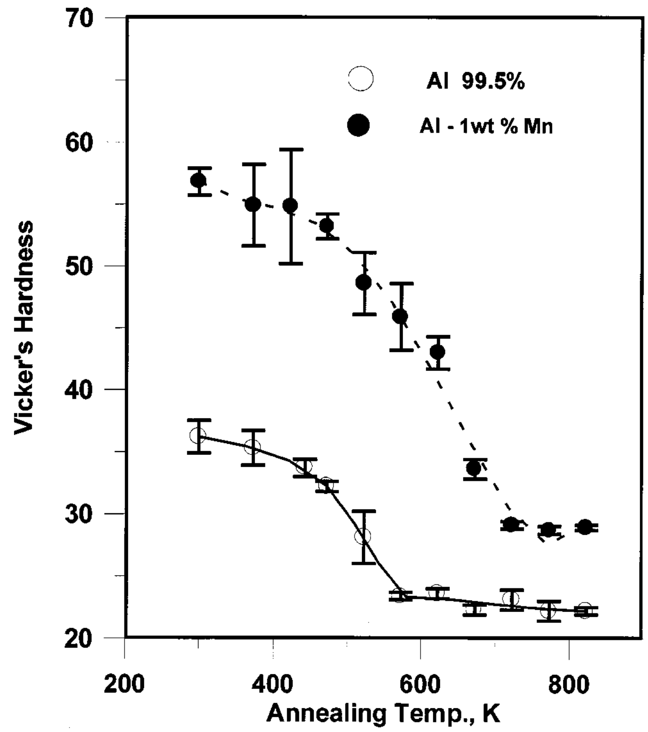

Fig. 2 Vicker's hardness vs annealing temperature

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

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

2. Experimental

The samples that have been investigated were commercial aluminum 99.5% and Al-1 wt.% Mn. Their chemical composition is shown in Table 1. These specimens were cold rolled to a reduction of thickness of ~67% and isochronal annealed for 1 hour in the temperature range 300 to 823 K.

The Doppler broadening setup consists of a hyper pure germanium (Canberra, USA) connected to a 575 Ortec (ORTEK, USA) amplifier and fed to a 444 Ortec bias amplifier. The measured full-width at half-maximum at 662 keV ^{137}Cs was established to be 1.0 keV, which is comparable with the variation of electron momentum usually of the order of ~1.5 keV. The energy dispersion of the equipment was 41 eV/ch. The number of channels included in the annihilation peak area was 320. The positron source used in this investigation was ~20 μci of ^{22}Na deposited on kapton foil (Netherlands Company) and sandwiched between two layers of the sample. The total number of counts in the measured spectrum was $\sim 10^8$. The S parameter (Fig. 1) was measured as the number of counts lying within an energy interval of 1.4 keV centered at the peak of the annihilation line. To eliminate the effect of annihilation with valance electrons,[7,8] the wing parameter W was calculated as the sum of counts lying within an energy interval of 1.7 to 3.4 keV further from the peak center on both sides of the peak. The parameters S_{nor} and W_{nor} normalized can be determined from the ratios of S/S_{ref} and W/W_{ref}, respectively. In order to determine these ratios, the S_{ref} and W_{ref} were obtained by measuring the line shape distribution using annealed samples of commercial Al and Al (Mn). The Vickers microhardness, Hv, has been measured at room temperature using a load 50 g for 10 s. Ten readings were taken for each sample, and the standard deviation was calculated. The measured values of Vickers hardness (Fig. 2) ranged from 36.2 $\pm$ 1.3 to 22.2 $\pm$ 1.3 and 56.8 $\pm$ 1.1 to 28.9 $\pm$ 0. 2 for Al (99.5%) and Al (Mn) alloys, respectively. These results are comparable with those obtained before[9] for the same alloys (the hardness of the recrystallized and cold-worked specimens was changed from 23 to 44 and from 28 to 55 for Al (99.5%) and Al (Mn), respectively).

3. Results and Discussions

The relationships between microhardness (Hv), the line shape S_{nor}, and the wing W_{nor} parameters with the annealing temperature for Al (99.5%) and Al (Mn) alloys are shown in Figs. 2, 3, and 4, respectively. It can be noted that S_{nor} at room temperature is lower for Al (Mn) than for Al, which is in agreement with the results obtained before by Hood and Schultz[7] for single crystals of Al and Al-1.5 $\times$ 10^{-2} at.% Mn. From the present results, we assume that during deformation (cold rolling), a higher density of dislocations is expected in Al (Mn) than Al, such that much of the annihilation signal

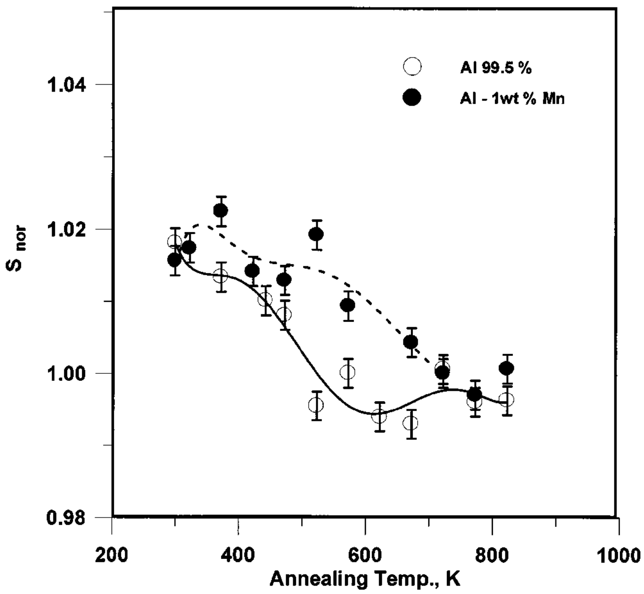

Fig. 3 *S* parameter as a function of annealing temperature

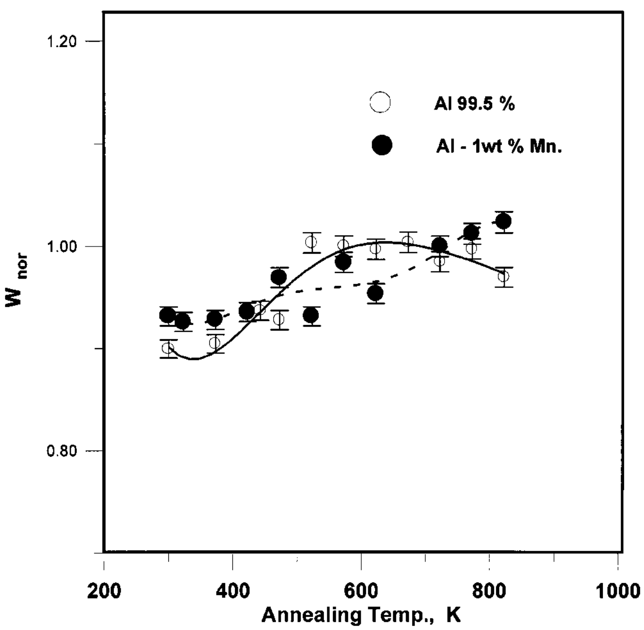

Fig. 4 *W* parameter as a function of annealing temperature

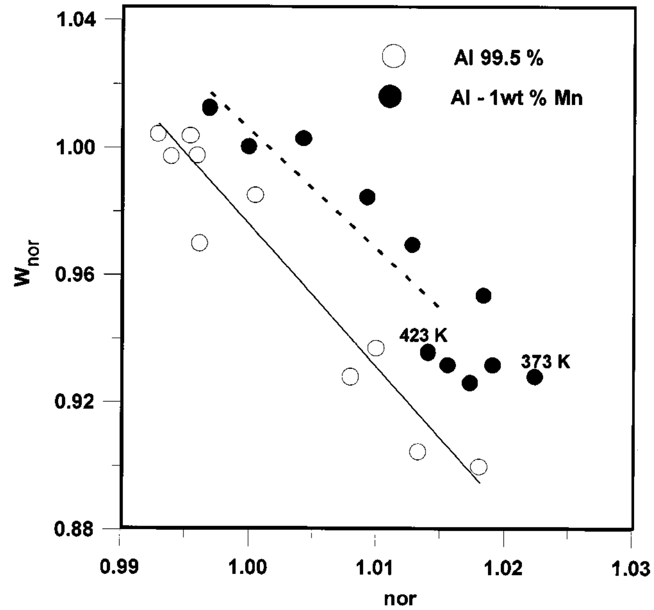

Fig. 5 *S-W* map of Al and Al (Mn)

represents positron-dislocation interactions. This situation would probably lead to smaller trapped-state parameter changes due to some degree of Mn-dislocation interactions than those that would be anticipated for dislocation traps alone, as in Al. With annealing temperature, the increase of S_{nor} for Al (Mn) compared with that of Al alloy is suggested to be due to some degree of Mn-vacancy interactions.

From the figures, three stages can be distinguished in both alloys. The first stage exhibits a slight decrease of Hv and S_{nor} and almost no change in W_{nor}, which can be related to the recovery process in these alloys. It ranged from 300 to 423 K for Al (99.5%) and from 300 to 473 K for Al (Mn). The second stage is characterized by rapid decrease of Hv and S_{nor} and a rapid increase of W_{nor}. This stage is attributed to partial recrystallization for both alloys and starts at 523 and 573 K for Al and Al (Mn), respectively. However, the third stage, recognized by a saturation of Hv, S_{nor}, and W_{nor}, indicates complete recrystallization for both alloys. It starts at 573 and 673 K for Al and Al (Mn), respectively. One can observe that at complete recrystallization (means free of defects) S_{nor} and W_{nor} approach the bulk value, which are 0.996, 1 for S_{nor} and 0.997, 1.02 for W_{nor}. This approach was previously observed for Al-Ag alloy.[10]

It appears that stages I and II for S_{nor} and W_{nor} are altered to higher temperatures than that for Al. Thus, the presence of Mn in Al matrix retarded the recovery and recrystallization processes. These results are in agreement with other ones[11,12] and can be attributed to the precipitation of stable Al_6Mn particles in Al (Mn) alloys. These processes are slow due to the low diffusion coefficient of Mn in Al[12] but are considerably accelerated due to the presence of Fe and Si. The nucleation and growth of some Al_6Mn metastable particles therefore may be responsible for the retardation of recovery and recrystallization in Al (Mn) alloys.[12]

Calculations by Hodges[13] and the measurements[14,15,16] demonstrate that both vacancies and dislocations in metals are able to trap positrons and thereby cause them to annihilate with a different electron distribution than in a defect-free lattice. Hood and Schultz[7] expected that the rise of *S* is due to an increasing size of the three-dimensional vacancy cluster, which nearly increases linearly with the number of vacancies in the defect.

Figure 5 illustrates the S_{nor} and W_{nor} plots for Al and Al (Mn) alloys with temperature *T* as a running parameter. The combined use of S_{nor} and W_{nor} allows a better view of types of defects, as demonstrated before by Van Veen *et al.*[8] In general, the points in the plot follow a linear trajectory for Al alloy, which indicates that one type of defect is thermally generated.

In the case of Al (Mn) alloy, the trend is similar for temperatures higher than 423 K. However, for Al (Mn) alloy, in the temperature range 300 to 423 K, the slope of trajectory changes, which may indicate the occurrence of another defect type (Mn-dislocation interactions).

4. Conclusions

The obtained results indicate the following.

- The behaviors of Hv and *S* are similar, while *W* shows a reverse behavior. The variation of Hv and *S* with temperature in Al and Al (Mn) samples indicates the presence of three stages, recovery, partial recrystallization, and complete recrystallization.
- The strong interaction between Mn atoms and dislocations in Al leads to a significant retardation of both recovery and recrystallization with temperature.
- When the two alloys are recrystallized *S*, *W* approaches the Al bulk value.
- The *S*-*W* map has revealed the presence of one type of defect in commercial Al, as indicated by Fig. 5. However, in Al (Mn) alloy, a deviation from the trajectory behavior is observed at low temperature, indicating another type of defect (Mn-dislocation interaction).

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