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A Study of Precipitation Phenomena in Aluminum Alloys by Positron Annihilation*

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In order to assess the sensitivity of positron annihilation (PA) to precipitation phenomena, the Doppler broadened PA lineshape was monitored as a function of isochronal annealing steps on a series of quenched aluminum alloys. A study of the Al-Cu system, including both PA and transmission electron microscopy, provided new information on the role of vacancies in the formation of Guinier-Preston zones and led to the conclusion that incoherent precipitate-matrix interfaces can provide efficient trapping sites for positrons. Further PA measurements on $AlMg_2Si$ and AlSi samples showed that the lineshape parameter changes coincided with microstructural changes during annealing deduced from other techniques and indicated that PA provides a sensitive monitor of these phenomena.

Earlier work has shown that positron annihilation (PA) responds to precipitation of solute atoms from solid solution [1]. In order to determine what changes in the alloy microstructure are sensed by PA, we undertook first a systematic study of AlCu by both PA and transmission electron microscopy (TEM). AlCu was chosen because it is the classical system in which Guinier-Preston (GP) zones form, and for which the evolution of the microstructure has been well documented [2]. Further PA measurements were done on $AlMg_2Si$, another system in which GP zones are thought to form but which is less well understood [3,4]; and on AlSi, in which incoherent precipitates are formed directly without first forming GP zones [3].

Measurements of the Doppler broadened PA lineshape for the samples were made during isochronal annealing sequences. The apparatus and experimental procedure are described in detail in reference [5]. The alloy samples were first annealed at 823 K in the solid solution region of the phase diagram and then rapidly quenched to 200 K, where vacancies in Al are not mobile. All Doppler broadening measurements were made at 85 K following annealing steps of 30 minute holding times at each temperature. Data were analyzed in terms of a lineshape parameter S, which is calculated by dividing the number of counts in a central region of the annihilation line, corresponding to about one third of FWHM, by the total counts in the line.

Figure 1 shows the variation in S-parameter with annealing temperature for pure Al quenched from 773 K and three AlCu alloy samples of different Cu content quenched from 823 K.

The main features of the curve for pure Al are the initial rise just above 200 K, where the vacancies become mobile and form small clusters, followed by a drop to a lower level due to the coarsening of the clusters

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into prismatic dislocation loops, and finally a return to the value characteristic of fully annealed Al as the loops anneal out [5].

The experimental results for the AlCu samples are discussed in detail elsewhere [6]. The main features are the drop occurring between 250 and 300 K, the decreasing value of S parameter with increasing Cu concentration in the temperature range 300-400 K, and the dip in the value of S between 450 and 650 K for the 2 and 4 wt.% samples. We note that S is lower for annealed Cu than for Al (28.1 compared to 33.5 for Al [6]), so that a drop in S below the Al value in an AlCu alloy indicates preferential sampling by the positrons of copper-rich regions. The lower values of S in the 300-400 K range are therefore due to the annihilation of positrons at Cu rich regions of the lattice. In the case of the AlCu 0.5 wt.% alloy we conclude that the Cu concentration is enhanced near the high density of dislocation loops that were observed by TEM. This is reasonable since substitutional Cu atoms are undersized in the Al lattice [7] and would therefore be attracted to the compressed areas of the lattice around the dislocation loops. The low value of S between 300-400 K for the 2 and 4 wt.% alloy suggests that here the positrons are being trapped at vacancies associated with the GP zones which begin to form at room temperature. This supports a recently proposed model for GP zones in AlCu based on extended x-ray absorption line structure measurements [8], which postulates a high density of vacancies in the Al matrix immediately next to the GP zones in order to accommodate lattice parameter differences.

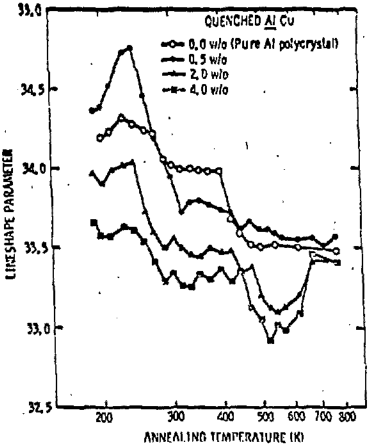

Fig. 1 PA lineshape parameter as a function of annealing temperature for pure Al and three AlCu alloys.

The abrupt decrease in S parameter between 450 K and 500 K for the AlCu 2 and 4 wt.% samples occurs at the temperature where the coherent θ" precipitate phase converts to the partially coherent θ' phase as shown by TEM [2,6]. When this occurs, misfit dislocations are formed at the edges of the disc shaped precipitates. The low value of the S parameter here again indicates annihilation of the positrons near Cu atoms in the lattice. We conclude that the positrons are being trapped at the misfit dislocations of the incoherent precipitate boundaries. Annealing at higher temperatures produces further coarsening of the precipitates until eventually they are so far apart that the positrons sample mainly precipitate-free regions of the lattice and the S parameter rises to a value close to that for annealed Al.

A similar annealing study was made using $AlMg_2Si$ samples with a composition of 1.0 wt.% Mg and 0.58 wt.% Si, corresponding to an atomic ratio of 2 Mg atoms per Si atom. Figure 2 shows the S parameter as a function of annealing temperature for these samples. The main features are a large rise at about 300 K, a drop at about 470 K and a second drop around 630 K. The evolution of the microstructure of this alloy is not as well understood as the AlCu system but is generally thought to be as follows [4,9]. First, small coherent GP zones are formed, which develop into needle-shaped zones in the <100> directions. At higher temperatures these convert to larger rod-shaped precipitates of the semi-coherent β' phase which has a hexagonal crystal structure. Finally, the precipitates develop into large incoherent platelets of the equilibrium β phase of Mg_2Si with the CaF_2 crystal structure.

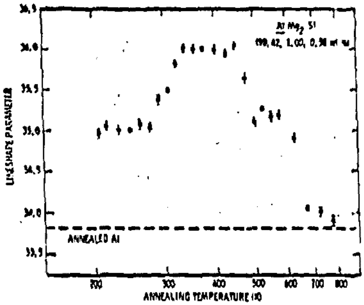

Fig. 2 PA lineshape parameter as a function of annealing temperature for the $AlMg_2Si$ sample.

Hardening of the alloy is believed to be due to the formation of the GP zones and since hardening has been observed to occur already at room temperature [10], it is reasonable to associate the rise in the S parameter at 300 K with the initial formation of these zones. It should be noted that the value of S in the plateau region of the curve where Al zones are present is high for the $AlMg_2Si$ alloy whereas it was low for the AlCu alloy. In both cases the positrons are sampling impurity rich regions in the lattice but the different electronic structures of the impurity elements give rise to the different effect on the value of S for these two alloys.

The first drop in the S parameter occurs near the temperature where the rod shaped β' phase precipitates have been seen to form and the final drop correlates with the formation of the large incoherent β-phase platelets [4,9]. Again, the final return to the value for annealed pure Al is probably due to the sampling by the positrons of mainly the precipitate-free regions between the large platelets.

We have also studied precipitation in an AlSi alloy sample. This system is different from the previous alloys in that no GP zones have been observed [11]. The precipitate phase is pure Si with a diamond cubic crystal structure. Previously reported studies of precipitation in this system [11, 12, 13] indicate that post-quench annealing at temperatures between 270 K and 340 K results in formation of clusters of vacancies and Si atoms too small to be resolved by TEM, but which act as nuclei for growth of Si precipitates at higher temperatures. These small clusters were found to nucleate only at temperatures below about 340 K in AlSi 1.2 wt.% samples [11], suggesting that they are unstable at higher temperatures. Residual resistivity measurements showed that precipitation of the Si occurs around 450 K [14] and annealing at 473 K results in the growth of the Si precipitates to a size large enough to be observed by TEM [15].

This picture is consistent with the results obtained from our PA measurements on the AlSi 1.0 at.% alloy shown in Figure 3. The S parameter shows little change up to 340 K but a large decrease between 340 K and 440 K. The decrease appears to be due to the dissolution of the small clusters of vacancies and Si atoms. This occurs at a higher temperature than the drop in S parameter associated with the dissolution of small vacancy clusters in pure Al, because of the stabilizing effect of the Si atoms on the clusters in the alloy. In the temperature range 440 K to 620 K where the Si precipitates are growing we observe an increase in the value of S. The return to the value of S for annealed Al at higher temperatures occurs when the precipitates have become so large that the positrons sample mainly the precipitate free regions of the material.

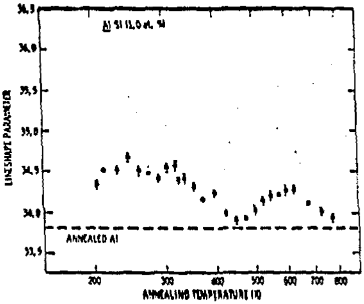

Fig. 3 PA lineshape parameter as a function of annealing temperature for the AlSi sample.

In summary, we have shown that changes in the PA lineshape parameter observed during annealing of three different types of Al alloys can be correlated with microstructural changes determined by other techniques. The PA measurements on the AlCu system, which were more comprehensive than the work on the other alloys, provide new evidence for the role of vacancies in the precipitation process.

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