A PAT STUDY OF INFLUENCES OF PRECIPITATION ON DEFECTS AND ELECTRONIC DENSITY IN Al-Li-Cu-Mg-Zr ALLOYS①

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ABSTRACT The positron annihilation lifetime spectra of an Al-Li-Cu-Mg-Zr alloy in the conditions of under-ageing, peak-ageing and over-ageing were measured. The results showed that the precipitation and growth of δ′ (Al_3Li) phase particles increased the bulk electronic density, while the precipitation of S' (Al_2CuMg) phase particles decreased the bulk electronic density. A large number of vacancies were recovered during the peak-ageing process, which made the number of defects decreased. However, the number of defects increased after the over-ageing process, which was related to the precipitation of S' phase particles.

Key words positron lifetime spectra Al-Li-Cu-Mg-Zr alloy ageing precipitation defects bulk electronic density

1 INTRODUCTION

Because of their high specific strength and excellent processability, the Al-Li-Cu-Mg-Zr alloys will find extensive applications in the aerospace industry. The mechanical properties of those alloys are controlled by the precipitates δ′(Al_3Li) and S'(Al_2CuMg)[1], but the micromechanisms are still unknown up to date. In this paper, the positron annihilation technology (Hereafter PAT), which is very sensitive to micro-defects, was used to study the influences of precipitation on defects and bulk electronic density in an Al-Li-Cu-Mg-Zr, aiming at revealing the micromechanisms of precipitation strengthening.

2 MATERIALS AND EXPERIMENTAL

The composition of the studied alloy is listed in Table 1. The preparation procedures of the alloy are as follows: melting in a vacuum induction furnace → refining and casting in argon atmosphere → two-step homogenizing treatment (763 K, 18 h + 793 K, 2 h) $\xrightarrow{\text{oxide scale removal}}$ heat preservation (723 K, 2 h) → extruding to dia. 30 mm bars → heat preservation (723 K, 1 h) → hot rolling to 4 mm thick sheets. Samples were cut from the sheets. After 798 K, 1 h solution treatment, cold-water quenching, and artificial ageings (463 K, 2 h, 16 h or 48 h), three sets of samples were obtained, i.e. under-aged, peak-aged and over-aged, which were marked Ⅰ, Ⅱ and Ⅲ respectively. Then 0.8 mm thick films were made, whose surfaces were polished using metallographic abrasive paper.

Table 1 Composition of samples (%)

Li	Cu	Mg	Zr	Fe+ Si	Al
2.49	1.22	0.61	0.06	< 0.4	bal.

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TEM observations[1] on differently aged samples show that there were present fine spherical δ' (Al_3Li) phase particles with smaller than 10 nm diameter in sample Ⅰ (aged at 463 K for 2 h), the δ' phase particles became much coarser (d = 30 nm) in sample Ⅱ (aged at 463 K for 16 h), and as the ageing time increased, there were S' phase particles preferentially nucleate at the grain boundaries, when the time reached 48 h, there were already present a large number of uniformly distributed S' phase bars at the grain boundaries and in the grain interiors while the δ' phase particles did not coarsen obviously and retained about 30 nm in diameter.

The positron lifetime spectra were measured at ambient temperature (25 ℃) with a standard quick-quick coincidence spectrometer made by the ORTEC company. The ^{22}Na positron source with kapton film as substrate was sandwiched between two identical samples. The two detecting heads at both ends were in antiparallel arrangement. The resolution of the apparatus (FWHM) was determined to be 260 ps, and the track increment was 12.7 ps. The total counts per spectrum reached 10^6. The POSIFITEXTEND program[2] was used to analyze the spectra.

3 RESULTS AND DISCUSSION

3.1 Parameters of positron lifetime spectra

The characteristic parameters were achieved by three-lifetime fitting of the measured positron lifetime spectra and deducting the source constituent (350 ps, 10.5%). In the characteristic parameters the third lifetime $\tau_a \approx 1300$ ps and its intensity $I'_a \approx 0.72\%$, which are contributed to the annihilation of the positrons on the surfaces of the source and the samples, and can be omitted. Re-normalize the first two lifetimes I'_1 and I'_2 and mark them as I_1 and I_2. According to the standard two-state trapping model[3], the annihilation ratio of the positron in the matrix of the alloy, i.e. λ_b can be obtained as follows:

$$\lambda_b = I_1\tau_1^{-1} + I_2\tau_2^{-1} \qquad (1)$$

Table 2 gives the characteristic parameters of positron lifetime spectra of the three kinds of samples, where τ_2 corresponds to I_2 and τ_1 to $(1-I_2)$. Table 2 also gives the values of τ_2^{-1} and $\tau_b (= \lambda_b^{-1})$. It is generally considered that λ_b, τ_2^{-1} and I_2 are related with the density of free electrons, the localized electronic density at the defects and the concentration of defects, respectively.

3.2 Influence of precipitations of δ' and S' phases on bulk electronic density

The variation of the λ_b reflects the variation of the bulk electronic density. By comparing the λ_b values of the three samples in different ageing conditions, it is clear that at the initial stage of ageing, the value of λ_b is relatively small, and it reaches the largest in the peak-ageing condition, further increasing the ageing time to the over-ageing condition will only reduce the value of λ_b.

According to Ref. [3], the bulk lifetimes of positrons in lithium, aluminium and copper are τ_b^{Li} = 293 ps, τ_b^{Al} = 166 ps and τ_b^{Cu} = 122 ps, respectively. When they form alloys, an individual copper atom contributes the most to the bulk electronic density, Al the next, and Li the least. Because the mole percent of Li atoms in the matrix is smaller than that in the δ' (Al_3Li) phase, as the δ' phase particles grow from under-ageing to peak-ageing, the Li atoms in the matrix are consumed continuously, thus increasing the bulk electronic density. At the same time, because

Table 2 Characteristic parameters of positron lifetime spectra for the three samples

Sample No.	τ_1/ps	τ_2/ps	I_2/%	λ_b/ns^{-1}	τ_2^{-1}/ns^{-1}	τ_b/ps
Ⅰ	167 ± 6	286 ± 9	34 ± 5	5.14	3.50	194.5
Ⅱ	170 ± 3	323 ± 11	19 ± 3	5.35	3.10	186.8
Ⅲ	168 ± 3	319 ± 9	24 ± 3	5.28	3.13	189.5

the mole percent of the Cu atoms in the matrix is smaller than that in the $S'(Al_2CuMg)$ phase, as the S' phase particles precipitate at the grain boundaries and in the grain interiors from peak-ageing to over-ageing, the Cu atoms in the matrix are consumed continuously while the δ' phase particles do not coarsen appreciably retaining the concentration of the Li atoms in the matrix almost unchanged, thus reducing the bulk electronic density.

The higher the bulk electronic density, the stronger the cohesive forces of the metallic bonds. Therefore, it is clear that the coarsening of δ' phase particles will help stiffen the metallic bonds and improve the tensile strength, while the precipitation of S' phase particles will soften the metallic bonds and worsen the tensile strength.

3.3 Influence of precipitations of δ' and S' phases on defects

It can be seen from Table 2 that as far as the value of τ_2^{-1} which reflects the electronic density at the defects and the value of I_2 which reflects the concentration of defects are concerned, sample I ranks the first, sample III the second, and sample II the last.

The internal defects in the Al-Li-Cu-Mg-Zr alloy after solution treatment followed by artificial ageing include thermal vacancies, grain boundaries and phase boundaries[4], and microvoids encircled by 3~4 δ' phase particles as well.

The value of I_2 decreased from under-ageing to peak-ageing, which shows that the as-quenched thermal vacancies can be recovered gradually by artificial ageing, and that with increasing ageing time the number of vacancy-type defects recovered in peak-aged sample II is larger than that in under-aged sample I, and the ratio of difficult-to-move defects such as grain boundaries and phase boundaries among unrecovered defects is relatively increased. And what is more, because of the growth of δ' phase particles, the open spaces of microvoids encircled by the δ' phase particles are enlarged, thus the value of τ_2^{-1} is reduced considerably. The reduction of I_2 from 34% in sample I to 19% in sample II shows that a large number of the thermal vacancies have been recovered by peak-ageing. Therefore, it can be regarded that in the under-ageing condition the thermal vacancies in the Al-Li-Cu-Mg-Zr are primary traps for positrons, while in the peak-ageing condition the microvoids are the primary traps for positrons. From peak-ageing to over-ageing the δ' phase particles do not grow appreciably, but I_2 increases from 19% to 24% and τ_2^{-1} increases slightly, which may be attributed to the precipitation and growth of the S' phase particles. Ref. [5] pointed out that the S' phase particles can act as the traps for positrons. If this is true, then the trapping rate of the S' traps for positrons must be equal to the difference of the trapping rates of sample III and sample II (see Table 3).

On the basis of the standard two-state trapping model, the trapping rate of traps for positrons can be expressed as

$$k = I_2(\tau_1^{-1} - \tau_2^{-1}) \quad (2)$$

For the extensive spherical defects such as vacancies and microvoids, the trapping rate relates to the size and number of the traps as follows[6]:

$$k = 4\pi D_+ rN \quad (3)$$

where D_+ ($= 10^{-4}\ m^2 \cdot s^{-1}$) is the diffusion constant of Al in the alloy[5], r the radius of traps, and N the number of traps in unit volume.

For the long and thin traps such as S' phase particles, the trapping rate can be expressed by[6]

$$k = -4\pi D_+ LN/\ln(\pi r^2 LN) \quad (4)$$

where L is the length of the traps, r the width of the cross-sections of the traps.

For estimation, take the average radius of thermal vacancies $r \approx 1$ nm; consider the size of microvoids encircled by δ' particles and that of the δ' particles of the same order of magnitude, $r = 15$ nm; take the typical values of the dimensions of S' phase particles[5] $r \approx 100$ nm (which are about the same as those obtained by TEM observations[1]). By means of eqs. (2) ~ (4), the numbers of defects in unit volume in

Table 3 Positron trapping rates and numbers of defects

Sample No.	Defect type	k / s^{-1}	r / nm	L / nm	N / m^{-3}
I	Thermal vacancies	8.47×10^{8}	1		6.7×10^{20}
II	Microvoids	5.29×10^{8}	15		2.8×10^{19}
III	Microvoids and S' phase particles	6.76×10^{8}			
III	S' phase particles	1.47×10^{8}	5	100	1.1×10^{19}

sample I and sample II and the increment of defects resulting from the precipitation of S' phase in sample III were calculated, and the results are shown in Table 3.

From the calculated results, it can be known that the number of defects in under-aged sample I is one order of magnitude larger than that in peak-aged sample II, in the over-aged sample III the number of S' phase particles is about the same as that of microvoids formed due to the coarsening of δ' phase particles in the peak-aged sample II.

4 CONCLUSIONS

(1) The precipitation of δ' phase particles will increase the bulk electronic density and help stiffen the metallic bonds, thus improving the tensile strength of the alloy, while the precipitation of S' phase particles will reduce the bulk electronic density and soften the metallic bonds, thus worsening the tensile strength.

(2) A large number of thermal vacancies can be covered by peak-ageing, in this condition the microvoids encircled by the δ' phase particles are the main defect type, and the number of defects is one order of magnitude smaller than that in the under-ageing condition. The number of traps for positron increases in the over-ageing condition, which is caused by the precipitation of S' phase particles.

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