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Positron annihilation spectroscopy as a tool to develop self healing in aluminium alloys

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Positron lifetime and Doppler broadening spectroscopy have been applied to probe the free volume generation (vacancies, dislocations and nano-cracks) during plastic deformation of a commercial aluminium AA2024 (T3) alloy. Aim of the total program is to study how solute atoms can be driven to the areas where initial cracking may occur in order to prevent the failure of the specimen. The phenomenon of closing the nano-crack is called Self Healing, and can provide extra strength and ductility to the alloy under some loading conditions. Plastic deformation of over-aged aluminum alloy at room temperature increases the average positron lifetime from initial value of 190 ps to 203 ps. The low momentum parameter S increases in agreement with the increase of open volume defects. The elastic deformation of the sample does not have a recordable effect on the positron annihilation data. It is also shown that the induced damage does not recover after loading the sample, i.e. the AA2024 in the T3 state is non self healing material, as expected, providing important first state result in the research of self healing Al alloys.

In solid metals self healing behaviour will be difficult to arrange as in general the atoms are insufficiently mobile to travel substantial distances to do their repairing action. However, there are two metal systems which maybe more promising: supersaturated aluminium alloys, which age harden naturally, and high temperature metals used close to their melting point. Supersaturated Al alloy subjected to a mechanical load may perform a healing phenomenon by aggregating solute atoms in the areas where strain has induced open volume defects [1–3]. The scope of this work is to study the self healing of Al-Cu-Mg alloy by generating deformation damage with controlled stress and strain levels and record the size and the chemical environment of strain-induced defects by positron annihilation spectroscopy. Other groups have shown that laboratory Al alloys in supersaturated solution exhibit positron lifetime changes during aging at room temperature indicating vacancy diffusion, which is essential for the self healing concept [4, 5]. We present the damage development in a commercial AA2024 in its fully hardened and most frequently used state. It provides the reference behaviour from which to judge future alloy modifications aimed at inducing self healing behaviour.

We are using positron annihilation spectroscopy (PAS) in order to study the native defects in Al alloy. A positron is the antiparticle of an electron, and it can get trapped by defects without positive charge, i.e. by vacancies or other open volume defects. Positrons can also get trapped by negative ions or by solute aggregates with higher positron affinity than the host atoms. A measurable positron signal is arising

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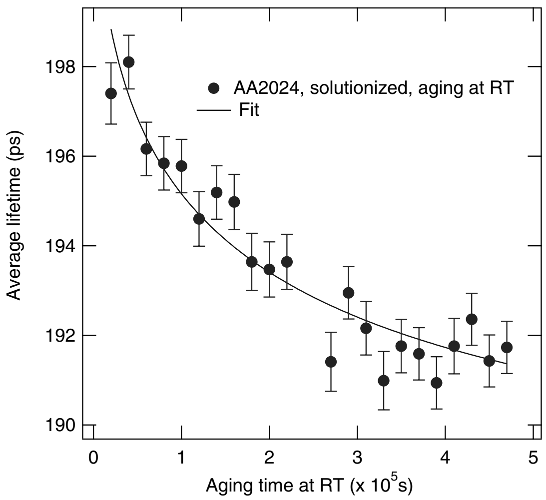

Fig. 1 Average positron lifetime during aging at room temperature after a solution treatment (492 °C for 10 min) and quench. The solid line is a fit.

from the annihilation of positron-electron pair where the mass of both particles is converted to two 511 keV γ-quanta. The broadening of 511 keV annihilation photo peak is often characterized by S and W parameters [6], where the S parameter describes annihilations with low momentum electrons and the W parameter is sensitive to annihilations with high momentum electrons. By using radioactive sources like ^{22}Na the lifetime of a positron can be measured since in the decay of ^{22}Na one 1.28 MeV photon is emitted almost simultaneously with the positron. By measuring the time interval between the birth and an annihilation photon the positron lifetime can be monitored.

For determining basic stress-strain curves for aluminum we cut samples parallel to the direction of rolling from rolled commercial 1 mm thick AA2024 sheet in the hardened (T3) state. Used Al alloy contains Cu (3.8-4.9 at.%), Mg (1.2-1.8 at.%), Mn (0.3-0.9 at.%), Fe (0.5 at.% max.), Si (0.5 at.%), Zn (0.25 at.%) and other constituents with concentrations of 0.15 at. % at maximum. The sample shape and tension test were done according to ASTM B557 standard. In order to perform aging measurements samples were solutionized in a furnace with circulating N_2 gas (492 °C for 10 min) and quenched into water (20 °C) and stored in LN_2 before test. All measurements were done at room temperature with a ^{22}Na source between the samples. The source was sealed between two kapton foils with a thickness of 8 µm. For lifetime measurements we use plastic scintillators and photomultiplier tubes, which were coupled to a fast digitizer (2 GS/s) [7]. After background reduction and source correction the spectra were analyzed with POSFIT-program [8] using a single Gaussian resolution function with a FWHM = 260 ps. For the Doppler broadening spectroscopy we use two HPGe detectors for detecting both annihilation photons in coincidence, which gives good peak-to-background ratio ($\sim 10^5$) with an energy resolution of 1 keV [9].

Figure 1 shows the average positron lifetime in the AA2024 (T3) sample during aging at RT after the solutionizing treatment at 492 °C and a quench into water. In contrast to the other samples in this work, the material was not mechanically loaded. The solid line in Fig. 1 is a fit to the function ($\sim \tau_0 + (\tau_{inf} - \tau_0)exp((-t/t_0)^{\beta})$) with similar parameters as described in Ref. [5]. The fit gives an initial lifetime τ_0 of 205±1 ps and a final value of τ_{inf}=188 ±1 ps with a time constant t_0 of 1.4±0.1 $\times 10^5$s and β=0.41. The more detailed analysis is beyond the scope of this work, but we emphasize that this is an important indication of vacancy and atom diffusion at room temperature, which holds promise for achieving self healing behaviour in this alloy.

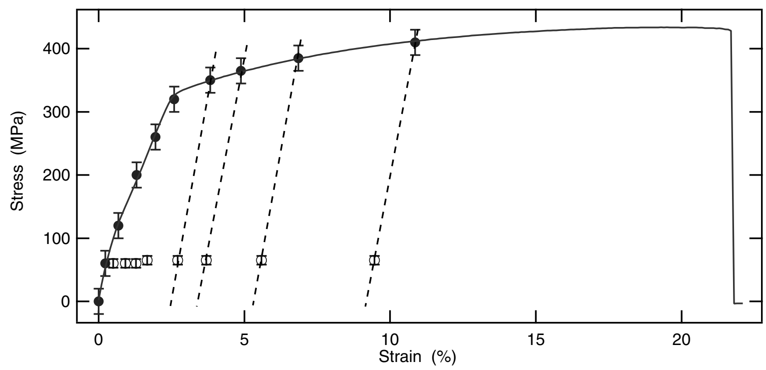

Fig. 2 Basic stress-strain curve of used Al2024 alloy (solid line). Symbols (filled and opened) denote points where positron signal of Fig. 3 have been measured. Dashed lines connect the points where the sample has been unloaded from high stress value to the lower constant level.

Figure 2 shows in a compact way the loading cycles of the sample. In the experiment the strain was increased stepwise (initially 1%, at higher strain levels bigger steps) and subsequently unloaded to a constant stress level. The measurement at full load reveals the damage development with increasing deformation, which includes the elastic and plastic component of the strain. The measurements at constant stress level reveal the damage at fixed level of elastic straining and provides a better indication of the damage density development because the contribution of elastic deformation is minor in relation to full load condition.

Figure 3 shows the evolution of the positron annihilation parameters during an applied load. The points are divided into two groups, where one denotes the measured positron signal during full load (filled circles) and another during unloading the sample (open circles). The intention is to describe the variation in the positron signal between different loading states of the sample (see Fig. 2). The free volume generation during mechanical load is clearly seen in Fig. 3. Both S parameter and average positron lifetime increase with increasing total strain of the sample, while the corresponding W parameter decreases. The parameters seem to be unaffected for strain values below $\sim$ 2.3 %. Referring again to Fig. 2, there is indeed a pronounced difference in the stress-strain curve at this strain value. This is due to the bigger yield of the material in the region where more dislocations are formed, and the fraction of positrons annihilating in them is increasing with increasing plastic deformation having a positron lifetime of $\sim$ 235 ps at the saturation [10].

There is also an interesting behavior of the parameters according to the plastic and elastic deformation of the samples. On the basis of Fig. 3 it can be said that releasing the load has no effect on the positron parameters whereas the increased plastic load will, in particularly, increase S parameter; open and filled circles increase side by side indicating that plastic deformation has generated damage, which does not recover even if the load has been removed. This shows that positrons are sensitive only to plastic deformation i.e. generation of dislocations and other open volume defects, while the detection of the elastic deformation is obscured not only by a small increase in dislocation density at low strain values ($<$ 2.3%), but also by experimental scattering and stability of set up. However, recognizing the non recovering plastic deformation in a controlled stress-strain test by positron annihilation technique is an important step to perform the detailed study of self healing in metal alloys.

Instead of using heat treatments to trigger the precipitation mechanism we study the possibilities to control solute atom clustering by applying mechanical load. The studied AA2024 (T3) alloy exhibits normal stress-strain behavior with regions of elastic and plastic deformation. The results of the combined tension test and the positron measurements during mechanical load can be summarized as follows: i) The

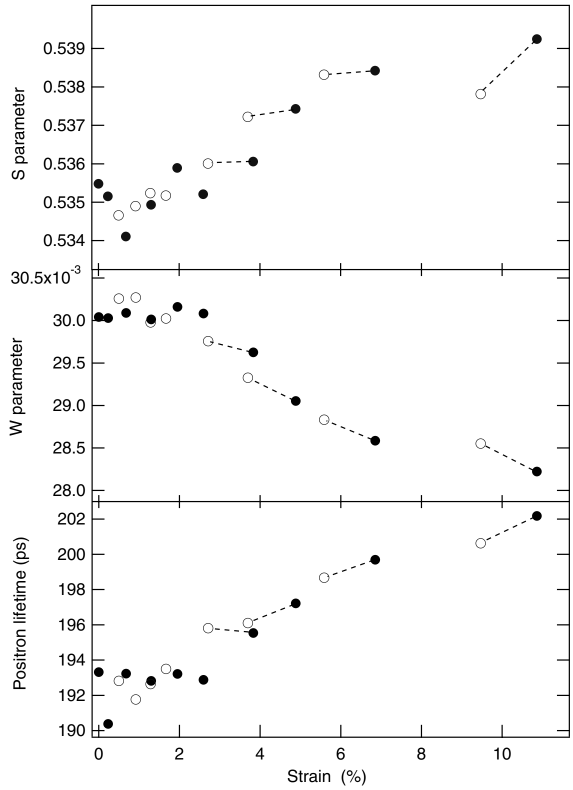

Fig. 3 Positron lifetime, S and W parameter as a function of the total strain of the AA2024 sample in T3 state. The S parameter is calculated from the momentum values between 0-3$\times 10^{-3}$$m_0c$ and the W parameter from values between 10-30$\times 10^{-3}$$m_0c$. Dashed lines connect the points of full load state and constant elastic strain (compare to Fig. 2). Estimated error is 0.5 ps for lifetime measurement and 1×10^{-3} and 5×10^{-4} for S and W parameter, respectively.

positron lifetime increases with increasing plastic deformation ii) The positron signal is more sensitive to the increase in dislocation density than the stretching of the lattice iii) There is hardly any difference in positron signal in the elastic region of the sample after plastic deformation. This means that the damage introduced by mechanically loading the Al-Mg-Cu sample does not repair automatically, i.e. the material is non self healing. This is, of course, an expected result as the material is an untreated fully precipitated Al alloy, but is, nevertheless, an important result because it provides information about nano-scale damage generation in metal alloys with carefully controlled stress and strain levels.

References

[1] R.N. Lumley, I.J. Polmear, and A.J. Morton, Mater. Sci. Technol. **19**, 1483 (2003).
[2] J. M. Silcock, J. Inst. Met. **89**, 203 (1960/61).
[3] K. Hono, Acta Mater. **47**, 3127 (1999).
[4] M. Biasini, G. Ferro, P. Folegati, and G. Riontino, Phys. Rev. B **63**, 092202 (2001)
[5] A. Somoza, A. Dupasquier, I. J. Polmear, P. Folegati, and R. Ferragut, Phys. Rev. B **61**, 14454 (2000).
[6] H. Krause-Rehberg and H.S. Leipner, Positron Annihilation in Semiconductors (Springer-Verlag, Berlin, 1999).
[7] J. Nissilä, K. Rytsölä, R. Aavikko, A. Laakso, K. Saarinen, and P. Hautojärvi, Nucl. Instrum. Methods A **538**, 778 (2005).
[8] P. Kirkegaard, N. J. Pedersen, and M. Eldrup, a program from Risø National Laboratory (1989).
[9] M. Alatalo et al., Phys. Rev. B **51**, 4176 (1995); M. Alatalo et al., Phys. Rev. B **54**, 2397 (1996).
[10] G. Dlubek, P. Lademann, H. Krause, S. Krause, and R. Unger, Scripta Mater. **39**, 893 (1998).

