PALS determination of defect density within friction stir welded joints of aluminium alloys

J Kansy¹, K Mroczka² and J Dutkiewicz²

¹ Institute of Material Science, Silesian University, Bankowa 12, 40-008 Katowice, Poland
² Institute of Technology, Pedagogical University of Cracow, Podchorążych 2, 30-084 Kraków, Poland

E-mail: kansy@us.edu.pl

Abstract. Positron annihilation spectroscopy is employed to investigate the density of defects in samples of aluminium alloys (2017 A and 6013) welded using the Friction Stir Welding method. The vacancy and dislocation densities were determined at the weld junction as a function of various parameters and conditions: Travel and rotational speed of welding tool, cooling of the surface of the welded material and the compositions of the welded alloys. The 3-state trapping model used in the computer analysis allowed to separate a vacancy component from a component related to dislocations. The determined lifetime of positron trapped by dislocation was much shorter than its experimental values referred to in literature, however, it is closer to the theoretical predictions.

1. Introduction

Conventional welding methods of the materials can be applied only to a few aluminium alloys due to many unfavourable phenomena occurring in the melt during the welding process e.g. oxidation of the weld, fracture due to contraction, structure degradation in the heat-affected zone as a result of overheating. An alternative method of joining metallic materials of flat shapes is the Friction Stir Welding (FSW) technology worked out at The Welding Institute, Cambridge. The basic principle of the method is making the joint in the solid state (materials are not melted). Friction between the welding tool and the materials causes a local increase in temperature up to the plastifying point. Simultaneously, the tool, due to its shape and movement (rotation with simultaneous movement along the welding line), causes mixing of
the plastified materials and thus the formation of the joint. The FSW welding process is presented schematically in figure 1. The structure of the FSW welds is complicated because the welding proceeds very dynamically and the material flow around the tool is strongly oriented [1].

The processes occurring during the formation of the structure of FSW joints are not fully discovered which is clearly visible in the studies of structure and mechanical properties of welds (e.g. distribution of microhardness) [2,3].

The aim of this work was to determine the changes in the defect structure at the region of FSW joint as a function of various parameters and conditions of welding: Travel and rotational speed of the welding tool, the cooling of the surface of the welded material and the compositions of welded alloys. It is expected that the results allowed to steer the conditions of the welding process towards the optimization of the structure of the friction stir welded joints.

2. Materials
Examined aluminium alloys 2017A and 6013 of composition given in table 1 were welded in the Institute of Welding, Gliwice, PL using a conventional friction stir butt welding.

| Additional alloying elements [%] | Fe   | Si   | Cu   | Zn   | Ti   | Mn   | Mg   | Ni   | Sn   | Cr   |
|---------------------------------|------|------|------|------|------|------|------|------|------|------|
| 2017 A                          | 0.23 | 0.42 | 3.94 | 0.07 | 0.03 | 0.55 | 0.57 | 0.01 | -    | 0.02 |
| 6013                            | 0.28 | 0.81 | 1.1  | 0.02 | 0.04 | 0.71 | 1.04 | -    | 0.08 | 0.08 |

In the aluminium alloys of the series 2017A a grain size of about 5 μm was found within the weld nugget. Grain refinement comparing to the original material took place due to a dynamic recrystallization. The observation performed by means of transmission electron microscopy showed higher dislocation density than in an annealed material.

A complex dislocation structure was found in the FSW welds of the 7075 alloy that contained screw dislocations, dislocation loops and dislocations interacting with precipitates [4,5]. Besides, in the structure of the weld nugget of 2017A alloy some helical dislocations were found, which indicates a process of structure formation with considerable influence of vacancies. Figure 2 shows the structure of the weld nugget of the FSW joint corresponding to the 6013 alloy with higher density of dislocations and also some dislocations interacting with the precipitates. On the basis of the TEM findings the following conclusion can be made: regardless of the type of the alloy higher density of defects in the crystal lattice for the FSW welds is observed. Fast cooling of the material during the welding should increase the density of defects.

3. The PALS experiment and its numerical analysis
The PAL spectra were measured at room temperature with a conventional fast-fast spectrometer of time resolution FWHM = 300 ps (value from fitting of Na22 lifetime spectra). The positron source of activity 400 kBq, covered by 5 μm Ni foil, was placed between two pieces of the investigated sample.
at the weld line. Thickness of the metal sheets was about 5 mm. Since the range of a positron emitted from Na\textsuperscript{22} in aluminium is only about 0.1 mm, PALS measurements were performed first with the positron source placed between the top sides (faces of weld) of the sample pieces and for the second time between their opposite sides. For each sample and its corresponding surface orientation a series consisting on 6 to 12 lifetime spectra was recorded. A single spectrum collected 1.2 million of counts. The peak to background ratio was 15700 : 4.4 and the calibration coefficient 6.37 ps/channel.

The experimental data were analyzed with the LT-9-2 software \cite{6}, which enables to fit many spectra simultaneously during the same fitting process. The simultaneous fitting reduces the number of free parameters used in the fitting, because some of the parameters are common for various spectra.

The spectrometer time resolution function and the source contribution were estimated on the basis of 33 PAL spectra (of total statistics 140 \times 10^6 counts) measured for pure well annealed silicon. The source correction for the Si spectra consisted of two source components of lifetimes 99 ps and 1.4 ns and intensities 33.6 and 0.4\%, respectively.

Numerical analysis of PAL spectra for the investigated samples was based on the assumption that each spectrum contained three sample components (besides the source components) related to the positron annihilation in the bulk material and to two kinds of defects. Therefore, for theoretical description of the spectra, the well known three-state trapping model \cite{7,8} was employed. The fitting procedure searched directly for values of the model parameters, i.e. the positron lifetimes in bulk ($\tau_b$), the lifetimes in two types of defects ($\tau_1, \tau_2$) and the positron trapping rates into the defects ($\kappa_1, \kappa_2$). During the analysis the lifetimes originating from the positron source were fixed at values determined previously with help of the Si sample. However, because of possible different positron scattering coefficients for Si and Al the component contributions were free. The found contributions were similar to those for Si, i.e. 33.1\% (for 99 ps) and 0.3\% (for 1.4 ns).

4. Results and discussion

At the beginning, the values of the lifetimes $\tau_b$, $\tau_1$ and $\tau_2$ were searched in a series of a preliminary fitting. The values that were found, turned out to be the same, within the range of error, for both investigated materials. The determined bulk lifetime $166 \pm 2$ ps was close to the measured value 163 ps \cite{9} and to the calculated value 168 ps for pure aluminium \cite{10}. The lifetime $\tau_1$ was found to be $253 \pm 3$ ps. Because this is similar to the measured positron lifetime 248 ps \cite{9} and to its theoretical value 254 ps \cite{10} for monovacancies in Al, we also assigned $\tau_1$ to positron annihilations in monovacancies in the investigated alloys. The determined value of $\tau_2$ ($190 \pm 3$ ps) was assigned to positron annihilation in dislocations. According to some theoretical calculations, the positron lifetime in dislocation lines in Al is only by 9\% longer than in bulk Al \cite{11} or even less (1.7\% of $\tau_b$) \cite{12}. On the other hand, the measured positron lifetime in dislocations (in deformed Al and dilute Al-Si, Al-Mg and Al-Cu alloys) by Hashimoto et al. \cite{13-15} are much longer (about 235 ps). It seems that the big discrepancies between the theoretical predictions and the experimental results can be explained by a complexity of the component assigned by the authors to positron annihilations in dislocations. The authors decomposed their analyzed spectra into two components only, of lifetimes $\tau_1$ and $\tau_2$, related to bulk material and to dislocations. Most probably $\tau_2$ indicates the trapping of positrons at more than one type of defects. Indeed, when we attempted to employ the 2-state trapping model in our analysis, the lifetime related with defects was found to be $232 \pm 2$ ps, which is very close to value 235 ps determined by Hashimoto et al. In our opinion, the 3-state trapping model, used by us, allowed to separate the dislocation component from a component originated from vacancies. Maybe the separation is not yet complete so $\tau_2$ could indicate resulting positron annihilation in the dislocation lines and in defects associated with them (e.g. jogs).

\footnote{The trapping model is introduced into the code of the program as its option. More details about this option and its operating one can find in the program manual.}
During the main fitting, the lifetimes $\tau_b, \tau_1$ and $\tau_2$ were fixed at the previously found values — 166, 253 and 190 ps, respectively. In this way only two model parameters were searched, i.e. $\kappa_1$ and $\kappa_2$. It is well known that the trapping rate of positron by defects is proportional to the defect concentration

$$\kappa = \mu c$$

(1)

where $\mu$ is the trapping coefficient characteristic for the structure of a defect. Its unit depends on the unit chosen for $c$. In this paper the defect concentration $c$ will be expressed either as the number density of vacancies or as the density of dislocation lines (m$^{-2}$).

With equation (1) we recalculated the found values of $\kappa_1$ and $\kappa_2$ into the respective defect concentrations. In this calculations we used $\mu_V = 4.5 \times 10^{14}$ s$^{-1}$ as a trapping constant for monovacancies in Al [9].

A problem arose with the estimation of the trapping constant $\mu_d$ for dislocations. In literature, there is a big scatter of the experimental values for $\mu_d$ in Al, ranging from $0.066 \times 10^{-4}$ to $2 \times 10^{-4}$ m$^2$s$^{-1}$ [16]. Moreover, $\mu_d$ can depend on the content of impurities in Al and even on the dislocation density [13-15]. However, we supposed, that the dependence of $\mu_d$ on the dislocation density observed in those works may result from the method of lifetime spectrum analysis which did not separate the positron trapping at dislocations from the trapping to vacancies. We hope that the separation of those contributions during our analysis makes $\mu_d$ less dependent (or even independent) on the dislocation density. For a rough estimation of the density of dislocation lines, we decided to use a constant value for $\mu_d$ equal to $1 \times 10^{-4}$ m$^2$s$^{-1}$.

The estimated values of monovacancy concentrations ($c_V$) as well as the density of dislocation lines ($c_d$) in the vicinity of the weld line as a function of the travel speed ($v_t$) of the welding tool are shown in figure 3. Both $c_V$ and $c_d$ decrease when $v_t$ increases. It turned out that, in the tested ranges of $v_t$, the variations of $c_V$ and $c_d$ can be well fitted by a logarithmic function

$$c = A \ln(v_t) + B$$

(2)

with fit correlation coefficients $R^2$ better than 0.995.

The observed dependencies can be easily explained. The motion of the welding tool releases a big amount of heat to the material...
and its temperature increases. At high temperature the lattice defects, produced during the welding process, are partially annealed. Of course, the higher temperature, the higher recovery of the material.

In order to investigate the effect of the cooling rate on the density of defects in the welding region, a sample of the 6013 alloy was welded once with cooling its top surface with solid CO₂ and for the second time without any cooling. In both cases the travel speed and the rotational speed of the welding tool were the same. Figure 4 compares the defect densities at the weld junction on the top surface of the samples determined by PALS measurements. It is seen that the recovery of the sample is less efficient in the case of the cooled sample.

5. Conclusions
The analysis of the PAL spectra of the investigated samples with the 3-state trapping model allowed to separate the contributions of different kinds of defects (vacancies and dislocations), therefore such determined positron lifetimes at the dislocation lines are lower than the experimental lifetimes usually referred in literature, simultaneously the values are closer to the theoretical predictions.

The PALS measurements enabled to monitor the density of defects in friction stir welded materials in relation to the alloy’s composition and the welding parameters such as the travel and rotational speeds of the welding tool and the efficiency for cooling the samples.

During the motion of the welding tool, the process of structural defect production competes with that of the material recovery. The densities of vacancies and dislocations at the welded part of the studied samples decrease linearly with the logarithm of the welding tool speed.

Acknowledgement
Partial financial support of the Ministry of Science and Higher Education of Poland under the grant number 3 T08A 035 30 is acknowledged. FSW joints were made in the Institute of Welding, Gliwice, Poland.

References
[1] Chan Z W, Pasang T and Qi Y 2008 Materials Science and Engineering A 474 312
[2] Peel M, Steuwer A, Preuss M and Withers P J 2003 Acta Materialia 51 4791
[3] Cavaliere P, Nobile R, Panella F W and Squillance A 2006 International Journal of Machine Tools and Manufacture 46 588
[4] Mroczka K, Dutkiewicz J, Lityńska-Dobrzyńska L and Pietras A 2008 Archives of Material Science and Engineering 33 93
[5] Mroczka K, Dutkiewicz J and Pietras A 2008 Journal of Microscopy 237 521
[6] Kansy J 1996 Nucl. Instr. Meth. A 374 235
[7] Bergersen B and Stott M J, 1969 Solid State Commun. 7 1023
[8] Connors D C and West R N 1969 Phys. Lett. A 30 24
[9] Schaefer H-E 1987 Phys. Stat. Sol. (a) 102 47
[10] Puska M J and Nieminen R M 1983 J. Phys. F: Metal Phys. 13 333
[11] Shen J Q, Lung C W, and Wang K L 1986 Phys. Stat. Sol. (b) 134 97
[12] Martin J W and Paetsch R 1972 J. Phys. F: Metal Phys. 2 997
[13] Hashimoto E, Iwami M and Ueda Y 1993 J. Phys.: Condens. Matter 5 L145
[14] Hashimoto E, Iwami M and Ueda Y 1994 J. Phys.: Condens. Matter 6 1611
[15] Hashimoto E, Iwami M and Ueda Y 1995 J. Phys.: Condens. Matter 7 9935
[16] Jensen K O, Eldrup M, Singh B N, Linderøth S and Bentzon M D 1988 J. Phys. F: Metal Phys. 18 1091