Positron Annihilation in Aluminium with Varying Grain Size

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Positron lifetime and lineshape parameter S measurements are reported for polycrystalline aluminium with different grain size. The results show that the grain boundaries can act as positron traps. The percentage of positrons trapped and annihilated on the surface of the grains depends on the grain size.

Introduction

It is well known that positron annihilation spectroscopy is a valuable tool for the study of lattice defects such as vacancies, vacancy clusters, voids and dislocations. Grain boundaries are also expected to act as trapping sites for positrons, since they are regions of substantial dislocation densities ($N_d \sim 10^8 - 10^{10}$ cm^{-2}). However, with a few exceptions [1—8], their influence on the positron annihilation parameters and the trapping mechanisms for grain boundaries has not yet been clarified.

Lynn et al. [1] have measured the positron lifetime in copper polycrystals that had three mean grain sizes and they suggest that grain boundaries can act as trapping sites for positrons. Using their results, Leighly [2] concluded that there is a linear relationship between the grain boundary surface area per unit volume and the mean positron lifetime. Our results in aluminium [3] showed that the parameter W_p increases with increasing number of grains per unit volume. Room temperature measurements with a fine-grained Zn-Al alloy [4] and more recently with a Zn-Al-Mg alloy [6] suggest that grain boundaries in these alloys can act as traps for positrons. In [5] a simple model for the positron trapping at the grain boundaries has been developed. Applying this model to the experimental results in [4], Hidalgo and Diego showed that there is a linear relationship between any linear annihilation parameter and the inverse grain size.

The purpose of this work is to study the dependence of annihilation parameters on the number of grains for polycrystalline aluminium.

Experimental procedure

The aluminium samples (99.998 grade) in the form of discs 20 mm in diameter and 3 mm thick were used. Different grains were prepared by giving the samples different deformation and thermal treatments (Table 1). The number of grains n was estimated by the intercept method using optical microscopy. After the thermal treatment, the samples were furnace cooled to eliminate the nonequilibrium concentration of vacancies, since vacancies also act as positron trapping sites.

Positron lifetime measurements were carried out at room temperature using a conventional fast-fast coincidence system. Two identical samples were sandwitched with a 12 μCi ^{22}Na positron source. The time resolution of the system was 280 ps (FWHM). Prior to all measurements the samples were chemically polished in a solution of 40 % KON and rinsed in water and afterwards in ethyl alcohol and dried in

Table 1

Sample number	Deformation ε, %	Thermal treatment	n/cm^2
1	5	40 min at	45
2	0	500°C and	65
3	8	2 h at	93
4	27	200°C	140

air flow. The measured lifetime spectra were analyzed as a sum of exponential lifetime components by means of *positronfit extended* computer program. After the source-background corrections, in all cases, two components enabled a satisfactory fit. From the analyzed lifetimes τ_i and the relative intensities I_i we calculate also the average positron lifetime $\bar{\tau} = \sum_i \tau_i I_i$ which is rather intensitive ($< \pm 2$ ps) to uncertainties in the analysing procedure. This parameter reflects reliably the changes of the defect structure which is of extreme importance in analysing small changes in the lifetime distribution. It must be pointed out, however, that the decomposed parameters offer much more information on the defect configuration than the average lifetime alone.

In addition to the positron lifetime we also measured the Doppler broadening of the annihilation line. The Doppler spectrometer resolution, measured by the FWHM of the 514 keV ^{85}Sr gamma-line was 1.17 keV. The energy spectra were treated to extract the lineshape parameter S.

Results and discussion

The results of measurements are presented in Table 2. For comparison, the characteristic lifetime values for different defects are also shown.

The main features of the experimental results are the increasing S (Fig. 1) and I_2 and the decreasing τ_2. The average positron lifetime $\bar{\tau}$ shows a tendency towards an insignificant increase. The experimental results suggest the presence of two kinds of defects that act as traps for positrons in the studied specimens. The only possible explanation for the appearance of the long lifetime τ_2 is the existence of small vacancy clusters. If we assume that τ_1 is due to positron annihilation in the bulk and in the dislocations (in pure aluminium vacancies are mobile at RT) and τ_2 is connected with vacancy clusters, applying a simple trapping model [12], which takes into account two

Table 2

Results from the analysis of positron lifetime spectra

Sample number	τ_1, ps	τ_2, ps	I_2, %	$\bar{\tau}$, ps	K_{cl}/K_d
1	178±2	449±19	16±1	223±2	0.47±0.1
2	175±4	417±33	18±2	221±2	0.65±0.1
3	175±1	418± 4	20±0.1	226±2	0.70±0.1
4	172±1	376± 1	30±0.3	237±2	0.95±0.1

Bulk	τ=166 ps [9]
Monovacancy	τ=246—253 ps [9, 10]
Dislocation	τ=250 ps [9]
Vacancy clusters	
N= 2	τ=273 ps [10]
N= 4	τ=329 ps [10]
N= 6	τ=351 ps [10]
N=13	τ=422 ps [10]
Pores (radius R=25 nm)	τ=450±10 ps [11]

types of defects, the positron trapping rate into dislocations K_d and vacancy clusters K_{cl} can be obtained from the equations

$$K_d = \frac{\tau_1(\lambda_f - I_2\lambda_{cl}) - I_1}{\tau_d - \tau_1} \quad (1)$$

$$K_{cl} = \frac{I_2}{I_1}(\lambda_f - \lambda_{cl} + K_d), \quad (2)$$

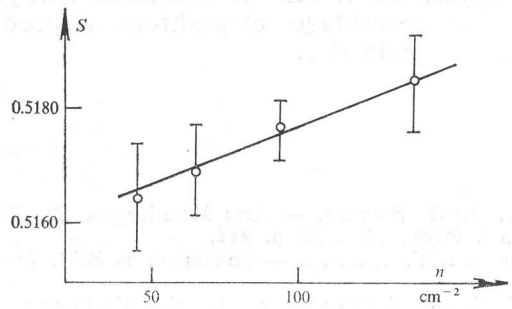

Fig. 1. *S*-parameter as a function of grain number/cm^2

where $\lambda_f = \tau_f^{-1}$, $\lambda_{cl} = \tau_{cl}^{-1}$ ($\tau_{cl} = \tau_2$). For all samples the ratio K_{cl}/K_d increases with the number of grains n. This means that the concentration of vacancy clusters increases. Based on theoretical calculations [13] we estimated that the observed clusters consist of about eight (sample *4*) to fifteen (sample *1*) vacancies, i. e. their mean diameter is of the order of 0.3—0.4 nm.

Denoting the specific trapping rate for dislocations and vacancy clusters as μ_d and μ_{cl}, respectively, the positron trapping rates K_d and K_{cl} become

$$K_d = \mu_d C_d \quad (3)$$

$$K_{cl} = \mu_{cl} C_{cl}, \quad (4)$$

where C_d and C_{cl} are the concentration of dislocations and vacancy clusters. Equation, (3) and (4) directly give the defect concentration in the lattice. There are no experimental data on the specific trapping rate μ_{cl} as a function of the cluster size. The theoretical calculations in [14] show that in the case of a small vacancy cluster μ_{cl} is proportional to the number N of vacancies in the cluster. If we denote $K_{cl} = \mu_v C_{cl}^*$ where μ_v is the specific trapping rate into monovacancies, we get a quantity C_{cl}^*, giving the effective concentration of the vacancies, which are bound within the vacancy agglomerates. The actual cluster concentration thus becomes $C_{cl} = C_{cl}^*/N$. In our estimates we took $\mu_d = 0.15$ cm^2 s^{-1} [15] and $\mu_v = 1.2\times10^{15}$ s^{-1} [16] and received for $C_d = (1.3$ to $2.2)\times10^{10}$ cm^{-2} and for $C_{cl}^* = (1$ to $2.6)\times10^{-6}$.

According to Nieminen and Manninen (p. 145 in ref. 16) the average positron diffusion length before annihilation is $L = \sqrt{6D_+\tau}$, where D_+ is the diffusion constant and τ is the lifetime against annihilation. For aluminium $D_+ = 0.4$ cm^2 s^{-1} [16], $\tau = 10^{-10}$ s and one finds $L = 1.55\times10^{-5}$ cm. If a mean grain size l is small enough ($l \lesssim L$) [8] an appreciable fraction of the positrons can reach the grain boundaries. One can see from our results that the annihilation parameter S and I_2 depend on the number of grains, but it is impossible to know if the obtained trapping signals are originated only at grain boundaries, or if, on the contrary, they also come up from positron annihila-

tion at other trapping sites (dislocations, vacancies, etc.), distributed within the crystalline grains. Bearing in mind the decrease of I_2 observed with increasing grain size, it can be suggested that in our samples the grain boundaries act as traps for positrons.

Conclusion

From this positron experiment it can be concluded that grain boundaries can act as traps for positrons. The percentage of positrons trapped and annihilated on the surface of grains depends on grain size.

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Аннигиляция позитронов в образцах алюминия с различными размерами зерен

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(Резюме)

В образцах поликристаллического алюминия с различными размерами зерен измеряли S параметр и времена жизни позитронов. Установлено, что позитроны аннигилируют частично в объеме матрицы и на дислокациях и вакансионных кластерах. Размер кластеров порядка 0,3—0,4 nm. Результаты и приведенные оценки указывают на наличие эффекта, связанного с аннигиляцией позитронов в субграницах.