
Proceedings of the 32nd Polish Seminar on Positron Annihilation, Jarnołtówek 2000

POSITRON ANNIHILATION IN PURE AND HYDROGENATED PALLADIUM DEFORMED BY UNIAXIAL TENSION

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Angular distributions of the positron annihilation quanta were measured for pure and hydrogenated palladium samples deformed by uniaxial tension up to different deformation degrees. The relative difference curves, and the values of the R parameter characterizing the dimensions of defects trapping the positrons, were determined. It was found that for relative elongations ranging from 1.9 to 3.5%, in both the pure and hydrogenated palladium, the positrons are trapped in defects larger than vacancy clusters. The values of the R parameter for pure palladium samples elongated up to 0.39, 0.77, and 1.5% indicate a dominant role of smaller defects, like the vacancy clusters.

PACS numbers: 78.70.Bj

1. Introduction

It is well known that metals are deformable, first elastically and then plastically, under action of external forces. During the elastic deformation, the ordering of the crystal lattice remains unchanged, only the interatomic distances along some crystallographic directions are changing, depending on the direction of external forces. The conventional elasticity limit for metals corresponds to the strain (deformation degree) not larger than 0.05%.

Plastic deformation requires the dislocation movement by gliding or climbing [1]. During the climbing process the edge or mixed dislocation is moving by adjoining (or releasing) vacancies to (or from) the extra half-plane. This way the edge and mixed dislocations are the sinks (or sources) for vacancies. The dislocation movement can be hampered or even blocked by various obstacles, e.g. other dislocations, impurity atoms, or grain boundaries.

In context of the present study it is worthy drawing attention to the interaction of dislocations with impurity atoms and grain boundaries. The very basis of the first of them is the mutual attraction of the stresses of opposite signs. Thus, the impurity atom inducing a compressive field will be attracted to the region of edge dislocation with a stretching field, i.e. under the extra half-plane, and vice

versa. Adjoining of an impurity atom to the dislocation diminishes the deformation energy and this is the driving force for such displacements. Especially important is the interaction of dislocations with interstitial atoms, in particular with fast diffusing hydrogen atoms, whose distribution in palladium can be easily rearranged even at room temperature [2].

Grain boundaries violate the continuity of both the crystallographic and slip planes, thus slowing down the dislocation movement, which in turn results in the piling up of the dislocations at the grain boundary. Piling up of the dislocations causes a local pressure on the neighbouring grains, which may lead to the generation of new dislocations in them or even to the changes of their shape.

Static tensile tests belong to the most widely used methods for determining the strength of materials. Nevertheless, due to the complexity of the problem and a great number of hardly controllable factors influencing the results of such tests, their interpretation on the atomic level is not unambiguous until now.

The purpose of the present study is to investigate the kinetics of the defects, formed in the pure and hydrogenated palladium samples during the static uniaxial tensile test, by measurements of the angular distributions of the positron annihilation quanta.

2. Experimental

The samples used in the experiments were two strips of 4N palladium with dimensions $18 \times 5 \times 1$ mm³. Before the experiments both of them were tempered at 1123 K and then slowly cooled, at the rate of 1 K min⁻¹, down to the room temperature. After cooling, one of the samples was electrolytically charged with hydrogen in 5% water solution of H₂SO₄, up to the concentration of 0.06 at.H/at.Pd.

For each of the samples the angular correlation curves of positron annihilation quanta were determined experimentally by means of a standard scintillation spectrometer described in detail in [3]. The positron source (²²Na) of the activity of about 10 mCi was used. All the measurements were performed at room temperature under normal pressure. Besides the undeformed samples, the curves of the angular correlation of the positron annihilation quanta have also been determined for both the pure and hydrogenated samples after static uniaxial elongation up to 0.39, 0.77, 1.5, 1.9, 2.7, and 3.5%.

3. Results and discussion

The most convenient way of a distinct visualization of the changes in the angular correlation of annihilation radiation (ACAR) curves for pure and hydrogenated palladium samples, caused by elongation, is the presentation of the experimental results in the form of relative difference curves, calculated according to the formula

$$\delta N = \frac{N_n(\Theta) - N_m(\Theta)}{N_m(\Theta = 0)}, \quad (1)$$

where $N_n(\Theta)$ and $N_m(\Theta)$ denote the number of coincidence counts at angle Θ for deformed and undeformed sample, respectively. $N_m(\Theta = 0)$ is the number

of coincidence counts for undeformed pure (not hydrogenated) sample at angle $\Theta = 0$.

The relative difference curves $\delta N = f(\Theta)$ determined for pure and hydrogenated palladium samples extended up to six different elongation degrees are shown in Figs. 1–6. The continuous line in all the figures represents the relative

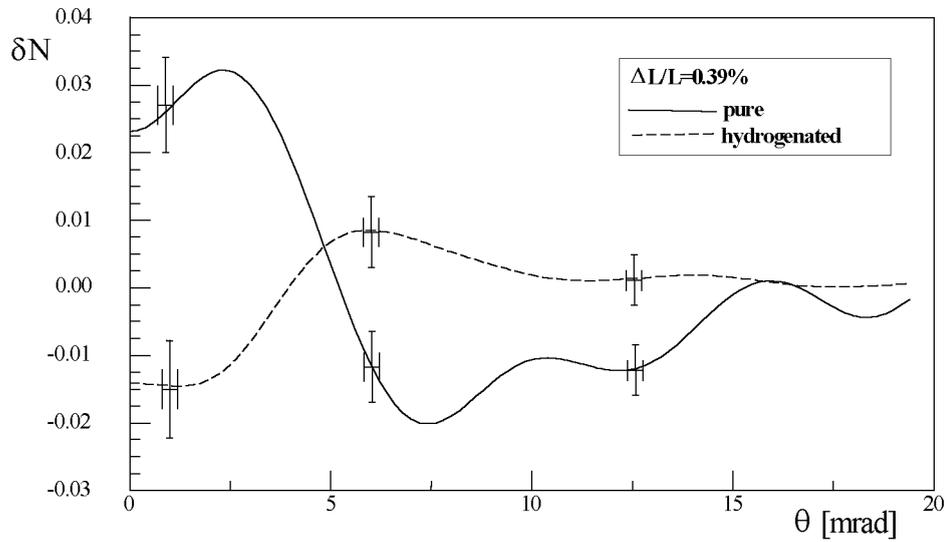


Fig. 1. Relative difference curves for pure and hydrogenated palladium samples elongated up to 0.39%.

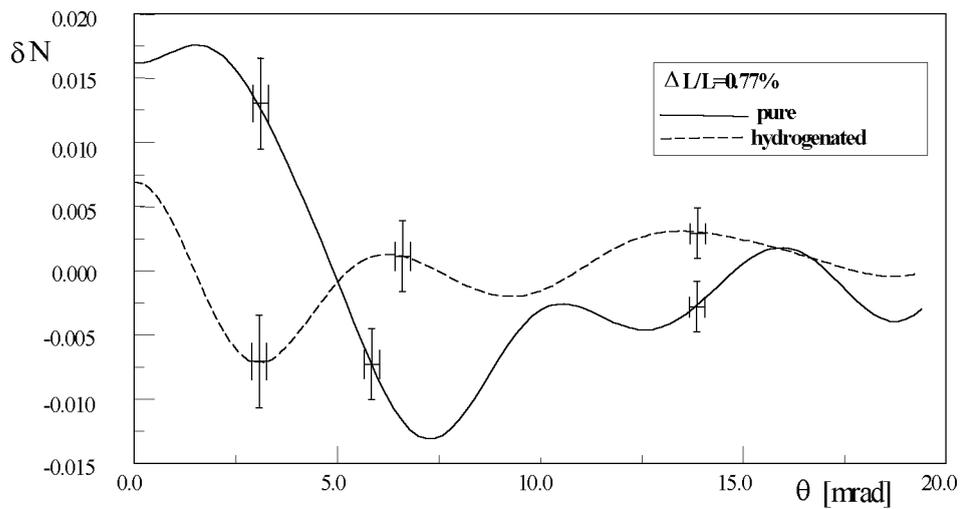


Fig. 2. Relative difference curves for pure and hydrogenated palladium samples elongated up to 0.77%.

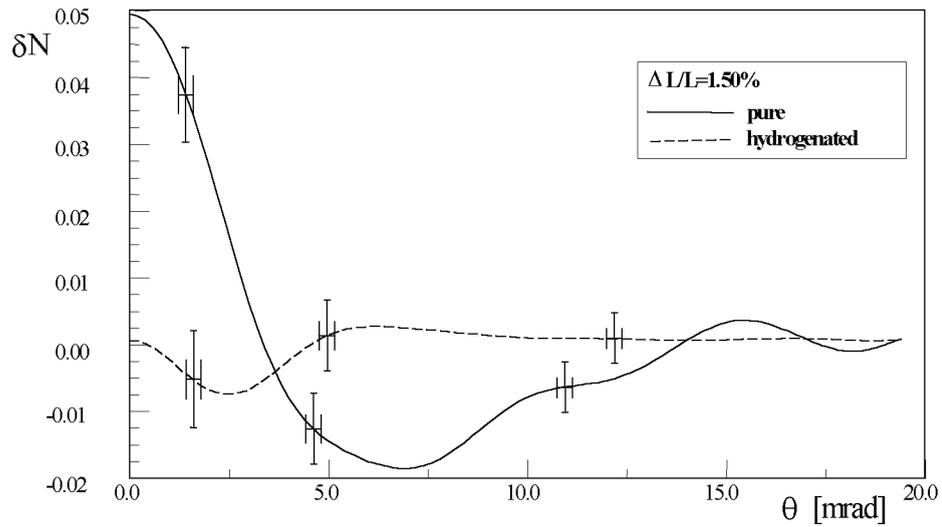


Fig. 3. Relative difference curves for pure and hydrogenated palladium samples elongated up to 1.5%.

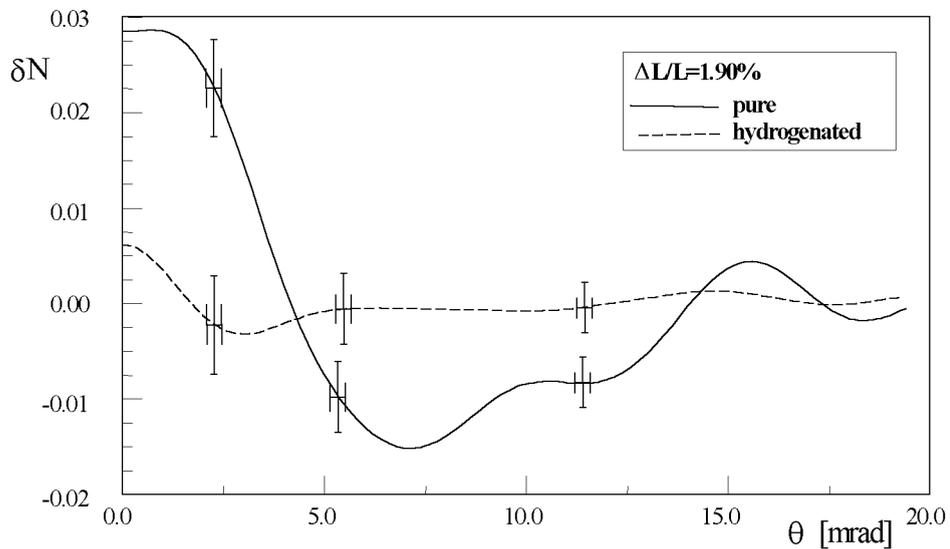


Fig. 4. Relative difference curves for pure and hydrogenated palladium samples elongated up to 1.9%.

difference curve for pure sample and broken line — the relative difference curve for hydrogenated sample.

Simple considerations lead to the conclusion that the positive value of relative difference δN in some interval of the values of the momentum component means that in a given sample the probability of positron annihilation with elec-

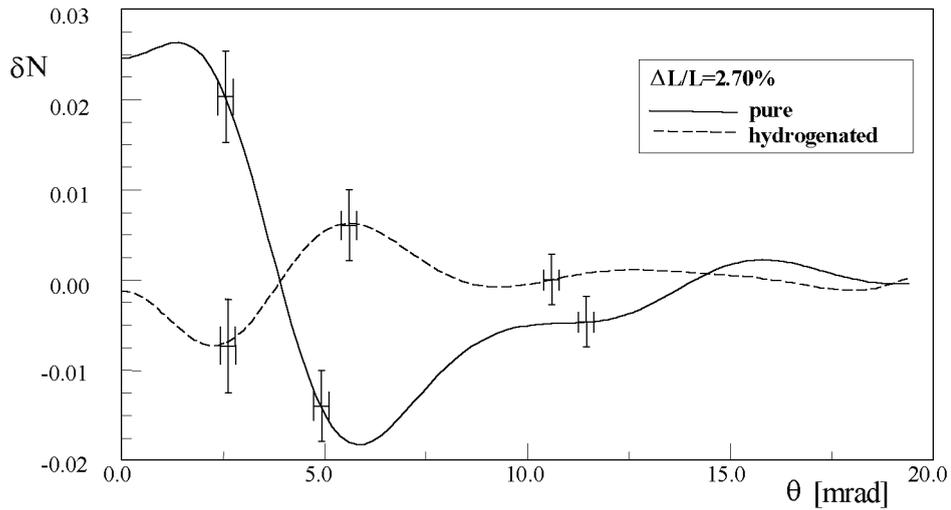


Fig. 5. Relative difference curves for pure and hydrogenated palladium samples elongated up to 2.7%.

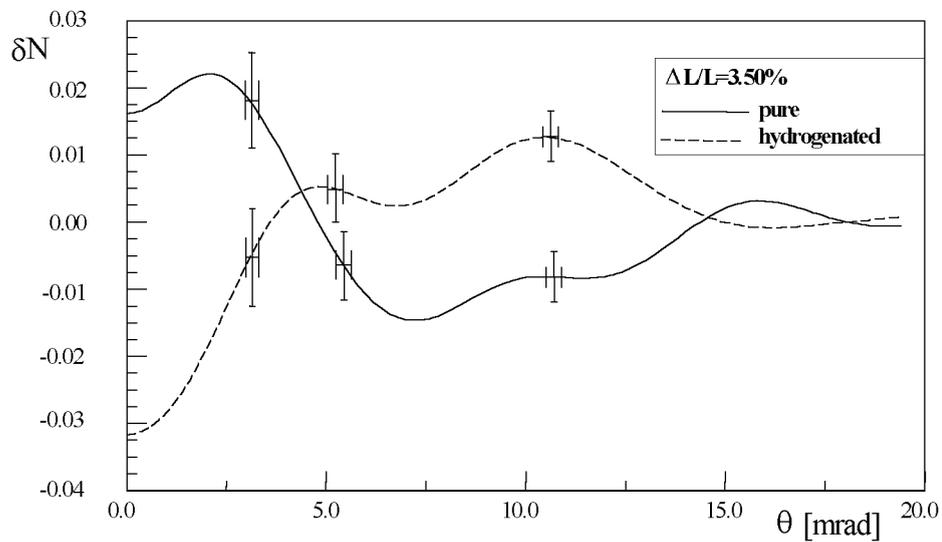


Fig. 6. Relative difference curves for pure and hydrogenated palladium samples elongated up to 3.5%.

trons having momentum component falling into this interval is greater than that in the reference sample.

From the shapes of the relative difference curves for pure (not hydrogenated) palladium sample (continuous lines in Figs. 1–6) it follows that at each relative elongation the probability of annihilation of positrons from the trapped state ($0 \leq \theta \leq 4$) is greater than that in undeformed sample. The increase in δN at relative elongations of 0.39 and 0.77% (Figs. 1 and 2) may be connected with

microdeformations at the grain boundaries as well as with formation of vacancy clusters due to the sample elongation. The greatest increase in the probability of annihilation from the trapped state is observed for samples elongated by 1.5% (Fig. 3). Probably, this result leads from the mutual interaction of dislocations with vacancies to the releasing of vacancies from dislocations followed by formation of vacancy clusters. Further increase in the relative elongation of pure palladium sample, starting from 1.9% to 3.5% (Figs. 5 and 6) causes a diminishing in the values of relative differences δN . The lack of distinct differences in the course of δN curves in this interval of elongations suggests that the configuration of defects generated during the deformation does not change substantially. Mutual interaction of dislocations probably leads to the formation of larger defects trapping the positrons.

In the case of hydrogenated palladium sample, the changes in the shape of δN curves at relative elongations ranging from 0.39 to 2.7% are very weak. The evident reason for such behaviour is the presence of hydrogen in the sample. Hydrogen atoms in deformed samples, besides natural for them interstitial positions, may be trapped in crystal lattice defects (empty spaces in the lattice). Hydrogen atoms may be trapped not only in the vacancies and vacancy clusters, but also at dislocations and grain boundaries [2]. The size of grains is much smaller than positron diffusion length. It is well known that in palladium hydrogen atoms are very mobile and tend to conglomerate. Therefore, it can be assumed that the absence of distinct changes in the δN curves could be explained by filling the defects generated by deformation by hydrogen atoms, thus making the defects inaccessible for positrons. Consequently, the number of positrons annihilating from the trapped state in the pure undeformed and hydrogenated deformed palladium is nearly the same.

For hydrogenated palladium sample elongated by 3.5% (Fig. 6, broken line) distinct change in the δN curve is observed. This is most probably connected with dislocation enhanced movements leading to their multiplication, accompanied by generation of vacancies, which in turn may be occupied by both the vacancies and hydrogen atoms. The vacancy–hydrogen pairs are mobile and form clusters [4]. As the number of hydrogen atoms in the sample is constant, the net concentration of defects accessible for positrons increases, thus the probability of trapping the positrons increases, too.

Additionally, Fig. 7 represents the changes of the parameter S as a function of the elongation of samples for the pure sample (continuous line) and the hydrogenated sample (dashed line). Earlier performed discussion of the course of relative difference δN (Figs. 1–6) is corroborated in presented in Fig. 7 curves.

The observed changes in the shape of the ACAR curves may be also characterized by means of parameter R , introduced by Triftshäuser [5] and discussed in detail in previous paper [6]. The changes in the value of the R parameter are connected with changes in the dimensions of defects trapping the positrons. The values of the R parameter were calculated according to the formula

$$R = \frac{H_n - H_0}{A_n - A_0}, \quad (2)$$

where H_m and H_0 denote the area under the central part of the ACAR curve

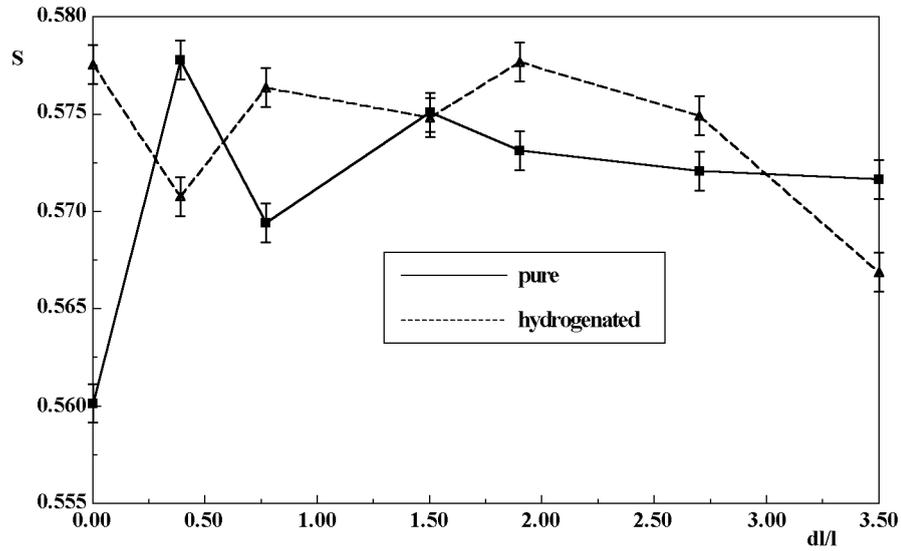


Fig. 7. The S -parameter as a function of relative elongation of palladium samples.

(for $0 \leq \Theta \leq 4$), measured at a given elongation and for undeformed sample, respectively, whereas A_n and A_0 represent the corresponding area in the tail part of ACAR (for $8 \leq \Theta \leq 12$). The calculated values of R parameter are presented in Table.

As it follows from Table, the values of R parameter for pure palladium sample at $\Delta L/L > 1.5\%$ are distinctly higher than those at smaller elongation are. This suggests that for weakly deformed samples ($\Delta L/L < 1.5\%$) the positron trap-

TABLE

Values of the R parameter for pure and hydrogenated palladium samples at different relative elongations.

$\frac{\Delta L}{L}$ 100%	R	
	pure sample	hydrogenated sample
0.39	0.2 ± 0.02	8.4 ± 0.1
0.77	1.1 ± 0.02	9.5 ± 0.1
1.50	2.3 ± 0.02	7.5 ± 0.1
1.90	9.3 ± 0.02	8.8 ± 0.1
2.70	9.0 ± 0.02	5.3 ± 0.1
3.50	6.2 ± 0.02	4.4 ± 0.1

ping defects have small dimensions — maybe they are the vacancy clusters. For $\Delta L/L \geq 1.9\%$ the positron trapping defects are much larger — their dimensions are comparable with those for hydrogenated sample.

Values of the R parameter for hydrogenated sample systematically decrease with increase in the relative elongation of the sample. It may be assumed that the diffusing hydrogen atoms occupy the defects thus preventing the trapping of positrons at these defects. Simultaneously, the sizes of the conglomerates of the vacancy–hydrogen pairs decrease with increase in the relative elongation of the sample.

The validity of the above consideration could be checked by measurements of the positron lifetime spectra or by other techniques. The results of the positron lifetime measurements will be presented in [7].

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