Presentation of the DSIP method for study of recrystallization process in subsurface zone induced by sandblasting

Jerzy Dryzek\textsuperscript{1,2}

\textsuperscript{1} Institute of Nuclear Physics PAN, ul. Radzikowskiego 152, 31-342 Kraków, Poland
\textsuperscript{2} Institute of Physics, Opole University, ul. Oleska 48, 45-052 Opole, Poland

E-mail: Jerzy.Dryzek@ifj.edu.pl

Abstract. In the paper we demonstrate the application of the positron annihilation experimental technique based on the scan of the positron implantation profile for observation of the recrystallization process in the subsurface zone (SZ). The SZ was created in the copper sample whose surface was exposed to a sandblasting treatment and then annealed at different temperatures. Application of $^{68}$Ge/$^{68}$Ga positron source allows us to scan the depth of c.a. 150 $\mu$m. It was found that recrystallization goes faster close to the surface than in deeper regions. The complete rebuilding of the microstructure takes place at temperature of 600 °C. Some changes in the defect structure begin at temperature of 300 °C.

1. Introduction

The scan of the defect depth profile using positron annihilation technique is usually possible applying the slow positron beam. In that case, only the close to surface regions at the depth up to a few micrometers can be observed. Nevertheless, in some cases the defect profiles are extended even to the depth of a few hundreds micrometers [1, 2]. In order to implant positrons at such a depth one may use a several MeV relativistic positron beam. Unfortunately, no beam is available at present for such studies.

To perform the detection of such depth profiles one can use the conventional positron techniques combining them with the sequenced etching method. After removing the sample material layer-by-layer deeper regions of the sample can be studied, for instance, by conventional positron lifetime spectroscopy. Numerous papers, devoted to the studies, of subsurface regions, or subsurface zone (SZ) created in the friction treatment, proved the usefulness of this method [3]. But this is a destructive technique, which cannot be applied to solution of some problems.

To solve this inconvenience the author proposed to employ the positron implantation profile which is always present when positrons from a radioactive source are injected into condensed matter. Before annihilation the positrons traverse a relatively large distance, which depends on the implant energy and material properties, and this fact can be applied to scan the defect profiles. Fortunately, positrons which have nonthermal energy are not trapped by the open volume defects, thus the implantation profile is limited only by the implant energy, density and to a minor degree by the atomic number of the implanted matter.

In general terms, the implantation profile is described by the near-exponential decay function. Thus, scanning that depth using, for instance, the HpGe detector and measuring the value of the $S$-parameter, defined as the ratio of the central part area to the total area under the annihilation line, one

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can get information on the defect depth distribution below the surface. The high sensitivity of the $S$-parameter to the defect type and concentration is worth noticing. The HpGe detector must be located behind the lead or tungsten shields with a narrow slit and the sample with the positron source is precisely moved perpendicular to the slit. The scheme of the setup is depicted in figure 1. We proposed to call the presented experimental method the depth scanning of the implantation profile (DSIP), and more details one can find in Ref. [4].

The positron implantation profile should more or less coincide with the defect depth profile. In the previous paper of the author that experimental technique was used for study of pure aluminium samples, whose surface was sandblasted [4]. The $^{22}$Na source was applied because in the aluminium the reciprocal value of the linear absorption coefficient for positrons from that source is $93.4(2.3) \, \mu m$ [5]. This allowed us to detect the SZ, which was extended to the depth of c.a. 250 $\mu m$. The present studies are performed for copper. The SZ may be extended to a similar depth but the reciprocal value of the linear absorption coefficient for positrons emitted from the $^{22}$Na source in copper is $26(2) \, \mu m$. Thus, the $^{58}$Ge/$^{68}$Ga positron emitter is employed. In that case, the end-point of positron energy spectrum is c.a. 1.9 MeV and in copper the reciprocal value of the linear absorption coefficient is equal to $133.5(8.3) \, \mu m$ [5]. We expected that the defect profile could be scanned to the depth of c.a. 150 $\mu m$. The aim of the performed studies was also the examination of the thermal stability of the SZ created by sandblasting.

![Figure 1](image)

**Figure 1.** The scheme of the DSIP apparatus used for scanning of the defect depth distribution in the SZ. Positrons emitted from the source are implanted into reference and treated samples, which surrounded the source. The samples and the source form a sandwich, which can move perpendicularly to the slit. The HpGe detector detects only one of two annihilation quanta. From the annihilation line, the $S$-parameter, sensitive to the open volume defects, is extracted as the function of the sandwich position.

2. **Sample preparation**

The sample in the shape of 0.2 cm thick plate of size $1 \times 1$ cm$^2$ was made from the oxygen-free-high-conductivity (OFHC) copper purchased from Nilaco. Before the sandblasting treatment the sample was annealed at temperature of 900 $^\circ$C under air pressure in a stream of nitrogen gas during 1 hour and then slowly cooled to room temperature. This allowed obtaining the virgin sample with residual defects. Before the treatment, the surface was etched for cleaning using 1:3 nitric acid and distilled water. In the measured positron lifetime spectrum for such a sample only the single-lifetime
component equal to 120(1) ps was found. This value coincides well with the data for pure copper reported by other authors (see in Ref. [6]). This indicated that initial, virgin sample is almost defect free. Then the surface of the sample was sandblasted by silicon carbide particles of diameter less than 0.5 mm. The pressure of the compressed air used for blasting was c.a. 6.5 bar.

In the experiment, the sandblasted sample was exposed to successive isochronal annealing and then the measurements of the positron lifetime spectrum and the S-parameter measurements in the DSIP apparatus were performed. Temperature step was 100 °C, starting from 200 °C. Samples were annealed for 1 hr at each temperature in the atmosphere of inert gas (N₂) and then they were cooled to the room temperature. All measurements were performed at this temperature.

![Figure 2. The dependency of the positron mean lifetime on the annealing temperature for sandblasted copper sample.](image)

3. Results and discussion

During the sandblasting treatment the region close to the surface was highly deformed. Then the stored energy is released in processes called recovery and recrystallization initiated by temperature increase. The recovery refers to changes in the properties of a deformed material which occur prior to recrystallization; these changes are such as to partially restore the properties to their values before deformation. During recovery the stored energy of the material is lowered by dislocation movement, i.e. annihilation and rearrangement of dislocations. The recrystallization involves the formation of new strain, and defects free grains in certain parts of the sample and subsequent growth of these to consume the deformed or recovered microstructure.

In the positron lifetime spectra two lifetime components were resolved, the value of the second one was equal to 230(5) ps with intensity c.a. 35%. This indicates that in the sample after sandblasting the microvoids are present which consist of three or four vacancies [6]. The annealing process induces the increase of this lifetime up to 271(10) ps and decrease of the intensity to 28%, which at 400 °C almost disappears. This indicates the onset of the recrystallization process and growth of new grains. In figure 2 we depicted only the mean value of the positron lifetime which is a robust parameter sensitive to the open volume defects versus the annealing temperature. The increase of the mean positron lifetime, observed at 200 °C can be explained by motion of dislocations with jogs which always induces creation of point defects like vacancies and interstitial atoms [7].

The measurement of the positron lifetime using the 22Na source enables us to observe only the subsurface layer of thickness of c.a. 26 μm, which was directly exposed to the treatment. The layers
below can exhibit other properties, especially when specific surface processes take place. Then, we skip to the DSIP technique which allows us to observe deeper layers in the treated sample. However, we have to measure the S-parameter instead of the mean positron lifetime, but at this stage, only such a measurement mode is available.

In figure 3 a) there is shown the number of counts per second in the positron annihilation peak as the function of slit position related to the sandwich of two samples and the positron source (see figure 1) between them. It says how many positrons from the source can reach the particular depth position in the sandwich. This dependency is symmetric because, the source emits positrons homogeneously into the full solid angle and it is located between two samples. The left part corresponds to the reference well-annealed virgin copper sample and right part corresponds to the sandblasted sample. Apparent minimum in the middle of figure 3 a) can be explained by nonhomogeneous distribution of the isotope in the source region. In the samples regions the positron implantation profile can be described by the exponentially decreasing function either in left or in the right part of figure 3 a). In both regions corresponding to copper samples the positron implantation profile, not sensitive to the defects and other microstructure features, is identical, as we expected. Nevertheless, careful observation of the positron annihilation peak and extraction of the S-parameters reveals new feature. Figure 3 b) shows the value of the S-parameter as a function of the slit position.

As we expected the value of the S-parameter is constant and low in the part where the virgin well-annealed sample was located. The S-parameter value increases up to a certain maximal value in the part corresponding to source region. Positrons annihilate also in the source and one should expect a certain value of this parameter, which depends on the source design and source envelope material. Then, in the part of the sandblasted sample, the S-parameter value is again lower than in the source region. Generally, the detected dependency in this region is different than in the region of the virgin sample. It is due to the presence of defects generated during surface treatment. Since the number of positrons, which reach deeper layers exponentially decreases, the DSIP method allows to scan only the region to the depth of c.a. 150 μm from the treated surface. The right part (not hatched area) represents the interesting region of the treated sample. It can be concluded that the S-parameter and hence the defect concentration decreases with the depth. We can apply the equation (3) taken from Ref. [4] for description of the S-parameter as a function of the sandwich position. (In the treated sample, the exponential decrease of the S-parameter is assumed (see Ref. [8]), but in virgin sample and source regions, the constant values of the S-parameter are assumed. Certainly, this dependency is convoluted with the spatial resolution of the apparatus.) These assumptions allow describing fairly well the detected dependency. The solid line in figure 3 b) represents the best fit. From this fit for the treated sample, we extracted the parameter $d_0$ (see (3) in Ref. [4]) which defines how fast the S-parameter profile and therefore the defect profile decays when the depth or sandwich position increases. Its value is equal to 157(40) μm and corresponds well with that obtained using another experimental technique based on sequential etching and measurement of the S-parameter (see Ref. [3]). Then the sample was annealed and the profile of the S-parameter was measured again.

It can be noticed that annealing at the temperature of 200°C does not change the S-parameter profile in comparison with the initial profile except the region near of the surface. However, for annealing at 300 °C the change in the gradient of the S-parameter is noticeable. More significant change is observed after annealing at 400 °C. That observations correlate with the results presented in figure 2. Nevertheless, it can be seen that the value of the S-parameter drops on the surface, then increases and reaches the maximum value at the depth of c.a. 60 μm from the treated surface and then decreases. Such a behavior indicates that the defects are annealed in the recrystallization process first close to the treated surface and than in deeper regions.

This is well understood if we consider that plastic deformation energy originates at the surface and then is distributed in the SZ zone. It is well known that the recrystallization process which is responsible for the reduction of the S-parameter is driven by that energy. We suppose that at the depth of c.a. 60 μm the energy stored in the plastic deformed area is relatively low and recrystallization process is not so efficient as in the region close to the surface.
The increase of the temperature to 600°C almost removes all defects produced previously in the sandblasting treatment, and the S-parameter profile becomes constant as it is in the left part corresponding to the reference sample. The increase of the S-parameter below 150 μm can be explained by two reasons. First, still the defects are not completely removed at this depth. Second the number of positrons which reach this depth is low and mainly background contributes to that value.

![Positron implantation profile and S-parameter profiles](image)

**Figure 3.** The positron implantation profile (a) and the profile of the S-parameter across the sandwich, which consists of the reference copper sample, the ⁶⁸Ge/⁶⁸Ga positron source and the sandblasted copper sample. The hatched area indicates the two first mentioned above parts of the sandwich. The positron implantation profile is shown (a) and the S-parameter profiles are presented for different annealing temperatures of the sandblasted sample (b). The solid line represents the best fit of the equation (3) in Ref. [4]. The axis on the top represents the depth from the treated surface of the sandblasted sample. (The error bars are marked only for the points of the dependency obtained for the sample after treatment and after annealing at 400 °C. In other cases the error bars are identical.)

### 4. Conclusions

Finally the following conclusions can be drawn:

The non-destructive technique, called DSIP applied to the studies of in the SZ in the sandblasted OFHC copper samples allows to observe microstructural changes induced by annealing at the depth of c.a. 150 μm from the treated surface. This was possible due to the application of the ⁶⁸Ge/⁶⁸Ga positron source.
The gradient of the $S$-parameter in the SZ caused by the gradient of defect concentration induced by sandblasting decreases after annealing at temperatures 300 °C and almost disappears at temperature higher than 400 °C. Annealing at 600 °C causes removing of all defects and recrystallization of the SZ.

The recrystallization process is faster close to the surface than in the deeper regions of the SZ. After annealing at 400 °C the $S$-parameter depth dependence exhibits different character, i.e. the $S$-parameter has a maximum value at the depth of c.a. 60 μm.

It seems that the SZ exhibits strong stability because only annealing at temperature higher than 300 °C induces some changes in the defect structure.

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