Pulsed low-energy positron beams: A useful tool to investigate defect structures in deformed metals and alloys

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Abstract. To understand in more detail the behaviour of deformed metallic materials, the knowledge of defects and defect distributions at an atomistic level is important. To investigate the plastic zone in front of a crack tip, the positron annihilation lifetime spectroscopy with a pulsed monoenergetic positron beam of variable energy allows the detection of vacancies, dislocations, vacancy clusters and micro voids in the crack surface near region. Moreover, for a given defect type it is possible to determine its concentration. The positron lifetime measurements in samples of different materials (aluminium, copper) showed different defect profiles for crack surfaces produced by monotonic and cyclic deformation. In addition, this technique was able to characterize the kind of damage (monotonic or cyclic) by analysing the different positron lifetimes.

1. Introduction

The lifetime prediction of cyclically deformed constructions used for airplanes and cars becomes increasingly important in order to minimize the amount of very expensive and time consuming mechanical tests. Typically, linear damage accumulation models were used for the fatigue lifetime prediction, but in most cases the damage evolution is non-linear, and, consequently, microstructural aspects of damage have to be taken into account.

In general, two stages can be distinguished in the fatigue lifetime: The early stage of deformation, accompanied by the formation of the crack, and the stage characterized by the cyclic growth of the fatigue crack. Considering the crack as a kind of damage phenomenon it is necessary to understand the formation as well as the propagation of the crack. The latter can be measured by conventional techniques like compliance measurements or by the DC potential drop method.

In case of monotonic or cyclic crack propagation of ductile materials it has been found that the process zone (monotonic or cyclic plastic zone) in front of the crack tip determines the behaviour of the crack expansion. Consequently, the knowledge of defects and defect distributions at an atomistic level within this zone is highly desirable.

Positron annihilation lifetime spectroscopy with pulsed monoenergetic positron beams of variable energy allows for depth profiling of vacancies, dislocations, vacancy clusters and micro voids. Moreover, for a given defect it is possible to determine its concentration. Therefore, this technique offers a unique tool for investigations in deformed materials.
In this paper we will give a short overview of defect characterizations with pulsed low energy positron beams in cracked specimens of pure copper and a technical aluminum alloy. Most of the results have been published and discussed in more detail elsewhere [1,3-5]. Our aim is mainly to draw the attention on a method of potentially great interest to a wider scientific community of materials scientists.

2. Positron lifetime spectroscopy with pulsed positron beams

The positron is the anti particle of the electron. In condensed matter it will annihilate with an electron into two or more gamma quanta with a lifetime characteristic for the local electron density at the annihilation site. A positron with energies in the keV-range, implanted in a metal, thermalizes rapidly within a few picoseconds. This time is short compared to the time of hundreds of picoseconds typically spent before annihilation. In a defect free crystalline material, the positron survives for a time which is characteristic for the material. In the presence of open volume defects, due to the missing repulsive ionic potential, transition to a localized state, trapping, may occur. The electron density at the site of the defect is locally reduced and, consequently, the positron lifetime is prolonged with respect to the lifetime in the defect free bulk. Trapping occurs also at regions with different positron affinity than the crystalline matrix, e.g. at precipitates. Again, the lifetime is characteristic of the material and the type of defect. The high trapping rate into open volume defects and regions of high positron affinity is the reason for the high sensitivity of positrons as defect probes: Defect concentrations in the ppm-range may still be detected, from the trapping rate of the positron in a given defect the defect concentration may be inferred.

In positron lifetime spectroscopy the time spent by the positron between its implantation in the solid and its annihilation is measured. In a pure, defect free material, this yields an exponential decay spectrum which may be characterized by a single lifetime. In the presence of defects, additional exponential components with characteristic defect lifetimes may be detected. The relative intensities convey information on the defect concentration. Another useful but less specific parameter to characterize the lifetime spectrum is the intensity-weighted sum of all lifetimes, the mean lifetime. A mean lifetime longer than the lifetime in the defect free material points to the presence of defects.

To depth resolve defect structures one has to employ monoenergetic positron beams of variable energy. The implantation profile of such a beam may be approximated by the derivative of a Gaussian centred about a mean implantation depth. The mean implantation depth \( \bar{z} \) scales with the density \( \rho \) of the material and the beam energy \( E \) as \( \bar{z} \propto E^{1.6} / \rho \). Positrons implanted with a given energy sample a region extending from the surface down to two times the mean implantation depth.

To perform positron lifetime measurements with a monoenergetic beam, the beam has to be pulsed. A pulsed low energy beam system of variable energy for depth profiling of laterally homogeneous defect distributions, PLEPS [6], was developed at our institute and operates nowadays at the high intensity positron source NEPOMUC at the Munich research reactor FRM-II [7]. Also, a Scanning Positron Microscope (SPM) [8] has been developed, which offers additional lateral resolution down to 1 micrometer. After demonstrating its usefulness with a weak conventional positron source in the laboratory, which entailed however excessively long measurement times, it is currently installed at NEPOMUC and will be operative soon. Further technical details of both beam systems can be found in the references [6, 8].
3. Experimental results

3.1. Sample preparation

To gain more information about the crack evolution, fatigue crack propagation tests have been undertaken with single edged notched (SEN) specimens. For crack initiation, a starter notch of 1 mm depth was cut into the small side of the specimens. The specimens were cyclically deformed in a fatigue testing machine under symmetric push pull conditions. The actual crack length was measured by a potential drop method with a detection limit of 1 \( \mu \)m /cycle. Both signals, the stress amplitude as well as the crack length, were used to control the stress intensity factor, \( \Delta K \), and, as a consequence, it was possible to keep it constant. This procedure allows keeping the crack growth rate constant, too. The latter was important to obtain a constant defect configuration in front of the crack tip. Now the defect configuration within the crack tip near region was measured with PLEPS and SPM as it can be seen in the Figure 1.

![Figure 1. Employing PLEPS and the SPM on cracked specimen](image)

3.2. Results in pure copper

At first, the surface of the SEN specimen of a high purity copper alloy as seen by the SPM at 16 keV implantation energy is shown in Figure 2 [1]. The fatigue crack propagates from the lower right hand side to the upper left hand side of the picture. Different greyscales in the map correspond to different mean positron lifetimes. Close to the crack flanks the longest mean positron lifetimes have been measured. The band of enhanced lifetime is 100 \( \mu \)m wide, nearly symmetrical, and can be correlated with the cyclic plastic zone. The typical positron lifetime in plastically deformed copper is 172 ps [2]. Therefore, the maximum value of 250 ps in the mean positron lifetime indicates that defects with a larger free volume than mono vacancies and dislocations must exist close to the crack flanks.

![Figure 2. Mean lifetime map of a fatigue crack tip in pure copper](image)

To get additional information, a line scan was performed perpendicular to the crack as can be seen in Figure 3 [1]. The spectra could be decomposed into two positron lifetimes. The shorter one of about 190 ps was slightly larger than the 172 ps. This indicates the presence of small vacancy clusters close to the crack flanks. The longer lifetime of about 400 ps can be attributed to larger vacancy clusters. Taking the intensity of the second lifetime into account on

![Figure 3. Line scan over the crack tip in pure copper](image)
can see that positron trapping in vacancy clusters becomes most pronounced close to the crack flanks. Since the crack developed into two branches, two pronounced maxima are visible in the plots.

Now, the specimen was cracked and the fatigue crack surface was measured by the depth sensitive PLEPS. Figure 4 shows the mean lifetime vs. the mean implantation depth [3]. The solid line follows the prediction of a diffusion trapping model for positrons in pure copper without any deformation. The squares represent the lifetime measurement of a specimen of pure copper, carefully polished after the annealing at 800 °C in high vacuum. A good correlation between the model and the measurement had been found and the specimen was used as a reference. Lifetime measurements on the cracked surface of a monotonously fractured specimen showed that positron trapping at higher implantation energies occurs mainly in dislocations. In the case of fatigue fracture, a mean lifetime of 270 ps was found at higher implantation energies. Decomposition of the lifetime spectra into two components pointed to the presence of dislocations and of open volumes of 10 to 20 vacancies. This indicates that within the cyclic plastic zone the defect structure must differ from the defect structure induced by monotonic deformation [3].

3.3. Results in the AlMgSi-alloy AA6013

Starting from these results, a technical AlMgSi-alloy (AA6013) has been investigated by using SPM and PLEPS. At first the SEN-specimen was cyclically deformed at $K = 12$ MPa$m^{-1}$ in ambient air. After localizing the crack tip region of the fatigue crack the latter was investigated by the SPM as can be seen in Figure 5 [3]. The fatigue crack tip was surrounded by a plastic zone with a high dislocation density. The mean lifetime value of 240 ps points to a high density of dislocations as it is expected for plastically deformed material. At a radial distance of more than 60 μm from the crack tip, where the deformation becomes less pronounced, the positron lifetime was measured to be about 220 ps, corresponding to total trapping in precipitates, typical for
the untreated material. It becomes thus possible to estimate the dislocation density $c_{\text{disl}}$ from the SPM-diagram to [6]:

$$c_{\text{disl}}(x, y) = 4 \cdot 10^{11} \frac{\tau(x, y) - 220 \text{ ps}}{240 \text{ ps} - \tau(x, y)} \left[ \text{cm}^{-2} \right].$$

In addition, the dislocation density close to the crack tip was found to be as high as in heavily deformed metallic materials where a dislocation density of several $10^{12}$ cm$^{-2}$ can be expected. With increasing distance from the crack tip $c_{\text{disl}}$ decreases, as it can be seen in Figure 6. At a distance of 120 $\mu$m from the crack tip the density was found to be more than two orders of magnitude smaller than in the crack tip near region.

In addition, measurements with PLEPS on the cracked surface of fatigued samples were performed. Besides a lifetime corresponding to dislocations, a second lifetime, associated to small vacancy clusters of up to 20 vacancies was observed. Those vacancy clusters vanished within a distance of 2 $\mu$m from the crack surface [3]. As in the case of copper, on the cracked surface of monotonously fractured specimens, total trapping of positrons in dislocations, but no vacancy clusters were observed.

4. Summary and conclusions

The experiments have shown that positron lifetime spectroscopy with pulsed low energy beams of variable energy is able to elucidate complex defect structures at an atomistic level and to distinguish between different kinds of defects close to the crack tip for pure copper and aluminium alloys. The measurements clearly show that the defect configuration is quite different in the crack tip near area for specimens fractured under cyclic or monotonic loading conditions. The mean lifetime in the specimens fractured under cyclic loading conditions is enhanced with respect to the mean lifetime in those fractured under monotonic loading conditions. The presence of a second lifetime was only found in fatigued specimens and indicate a complex defect structure consisting of dislocation networks and small vacancy clusters. Those defects are located within the cyclic plastic zone in front of the crack tip. The presence of vacancy clusters of less than 1 nm in diameter confirms earlier results from Page et al. [9]. They found an increasing volume fraction of voids by SANS, in OFHC copper cyclically deformed at 405°C. Obviously, vacancy clusters are characteristic for fatigued copper, even at room temperature. It is well known that ordered dislocation structures develop during cyclic deformation under stress as well as under strain control. The latter had been studied with single crystals and polycrystals of Cu and Al-based alloys, cyclically deformed under strain as well as under stress control [10-12]. In case of low cycle fatigue dislocation networks as well as persistent slip bands (PSB) have been found. The kind of dislocation structure depends on the degree of the strain amplitude. For single edge-notched specimens fatigued under stress control, the formation of a persistent slip band has been found in front of the notch, where the highest stress fields will occur. In the early stage of crack propagation the fatigue crack follows the slip band and, in single crystals, oriented for single slip a crystallographic stage I crack has been observed by TEM [12]. It should be mentioned that these kind of dislocation structure has been only observed under cyclic deformation. In addition, the existence of the PSB and corresponding micro cracks has been detected by Katagiri et al. in the late 70s by using TEM [13].
Now, it can be summarized that in the light of fracture mechanics the dislocation arrangement has to be taken into account in order to understand the crack extension under cyclic deformation. It is assumed that the kind of the plastic zone in front of the crack tip determines the crack growth behaviour. The existence of the dislocation network was confirmed for a copper bulk material by ECCI measurements undertaken with SEM in front of the fatigue crack tip [14]. In case of monotonic deformation, however, this kind of dislocation arrangement cannot be found. The extension of the cyclic plastic zone in front of a fatigue crack can be determined by positron lifetime measurements with the SPM, taking the dislocation density into account as it is seen in Figure 6. The difference of the monotonic and cyclic plastic zones must be reflected in the monotonic and cyclic crack growth rate, as special fatigue tests in aluminium alloys, in ferritic and in austenitic steel indicate [15]. Currently, cracked surfaces of ferritic and austenitic steels are investigated with PLEPS at NE-POMUC.

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