Defect studies of stainless steel via positron annihilation energy spectroscopy

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Abstract. High Energy proton (up to 800 MeV) and spallation neutron irradiated samples of stainless steel 316L and Mod 9Cr1Mo were studied using positron annihilation energy spectroscopy. Doses delivered to 316L were up to 10 displacements per atom (dpa) and doses to 9Cr1Mo were up to 2.5dpa. We studied the change of $T$-parameter, which is calculated as the ratio of the number of counts in the wings of the Doppler-broadened 511 keV peak to the number of counts in the center of the peak. $T$-parameter is related to the density of defects in the sample of interest. Higher defect densities induce, generally, smaller $T$-parameter, although this is complicated by additional effects that include the size, nature and other properties of defects that may lead to saturation of $T$-parameter. For the large doses studied, positron annihilation energy spectroscopy showed that the $T$-parameter dropped sharply from 0 to 3 dpa, and continued dropping up to 10 dpa. In 9Cr1Mo, similarly, $T$-parameter dropped sharply from 0 dpa to 1dpa, but from 1 dpa to 2.5 dpa it remained constant, indicating that the density of defects or $T$-parameter saturated with dose above 1 dpa in 9Cr1Mo. These results, where the change in $T$-parameter from zero dose to 1 or more dpa, is much larger than the effect that we see from one irradiated specimen to another, led us in both cases to investigate lower doses. We measured energy spectra in 316L and 9Cr1Mo that were irradiated under the similar conditions as the above samples, but with doses less than 0.1dpa. These results fill in the gap between 0 and 1 dpa and suggest that most of the change in $T$-parameter occurs below 0.05 dpa.

1. Introduction
Positron annihilation spectroscopy is currently well known as a powerful tool of microstructure investigations of condensed matter. The interaction of positrons with matter is used to study configuration and properties of materials at the atomic level. This technique, which is a non-destructive spectroscopy technique to study voids and defects in solids, was developed in the early 50s and has advanced quickly since then.

The technique is based on the fact that a positron that enters a material will die away by annihilation. In the annihilation process of a positron and an electron, 511 keV photons are released that can be detected. The 511 keV spectral line is Doppler broadened due to existing finite momentum of the annihilation positron-electron pair. Doppler broadening is proportional to the momentum of the annihilation pair.
The properties of inhomogeneities (or defects) within a material are different from that of the rest of a material. These defects in turn influence various material properties. Positron annihilation energy and lifetime spectroscopy (PAES and PALS), can be used to verify both the density and size of defects in the material. Doppler broadening of the positron-electron annihilation line changes due to presence of defects in a material. If positrons are injected into a solid body, the shape of the annihilation peak strongly depends on whether they end up in a vicinity of large electron density or in a void where electrons are absent, and positrons annihilate effectively with lower momentum valence electrons of neighbouring atoms. In the latter case the broadening of the annihilation peak is less than when annihilating in the bulk material. Accurate analysis of the Doppler broadened spectral line is required to extract information about the material.

The technique requires a source of positrons. A radioactive isotope of sodium is often used. However for acquiring information from deep inside of material, a positron source inside of it is required. Activation is one method used to achieve this. A technique of activation is described below.

2. Methods
The source of positrons was radioactivity internal to the samples. Part of the samples of interest contained internal $^{22}$Na positron source as a result of daughter products of high energy proton and spallation neutron irradiation. The rest of the samples were activated by bremsstrahlung radiation from a 20MeV electron linear accelerator [1]. The photo-nuclear reaction $^{54}$Fe ($\gamma$, n) $^{53}$Fe was used for obtaining radioactive positron source $^{53}$Fe with 8.51 min half-life.

Our experimental technique is based on the measurement of the Doppler broadening of the 511 keV annihilation gamma line. A high purity germanium detector with energy resolution of 1.2MeV at 662MeV was used for measuring energy spectra. The detector was mounted to a spectroscopy amplifier and to an analogue to digital converter in order to obtain an energy spectrum from each specimen studied. The scheme of the experimental setup used is shown in figure 1. Energy resolution was controlled by keeping $^{133}$Ba and $^{137}$Cs in front of the detector during measurements.

![Experimental Setup Diagram](image)

**Figure 1.** Schematic sketch of positron annihilation energy spectroscopy experimental setup.

STW analysis of Doppler broadened 511 keV lines was completed by computer code written at the Idaho State University. $S$, $W$ and $T$-parameter values were extracted from the peak analysis. $S$ and $W$ have been defined as relative counts in the centre and wing of the peak with respect to total counts in the peak. $T$ was defined as ratio of $W$ and $S$. One should then expect to get lower $T$-parameter for highly damaged samples, due to less broadening of the annihilation peak.

3. Results
Samples studied were irradiated at Los Alamos National Lab with high energy protons (up to 800 MeV) and spallation neutrons. Materials used were stainless steel 316L and Mod 9Cr1Mo [2, 3]. A table below shows composition of these steel samples.
Table 1. Chemical composition of 316L and 9Cr1Mo.

| Material   | Al  | C    | Cr   | Cu  | Fe  | Mn |
|------------|-----|------|------|-----|-----|----|
| 316L       |     | 0.019| 17.3 | 0.26| Bal | 1.75|
| Lot E835   | Ni  | P    | S    | Si  | Ti  |    |
| Mod 9Cr-1Mo| 0.002| 0.089| 9.24 | 0.08| Bal | 0.47|
| Lot 10148  | 0.16 | 0.021| 0.006| 0.28| 0.002|   |

Doses delivered to 316L were up to 10 displacements per atom (dpa) and doses to 9Cr1Mo were up to about 2.5dpa [4]. The high dose samples resulted in 42 – 430 mR/hr activities at contact or around 1 mR/hr at one meter after allowing to decay ~ 8 years after irradiation. The low dose samples were irradiated under the same conditions, with irradiation temperature of 74C and 127C and activities obtained after irradiation were 6 mR/hr at contact or 0.5 mR/hr at one meter.

Results from analysis of Doppler broadened energy spectra are given in table 2.

Table 2. $T$-parameter values of irradiation damaged 316L and 9Cr1Mo.

| 9Cr1Mo Dose (dpa) | $T$-parameter | Error in $T$-parameter |
|-------------------|---------------|------------------------|
| 0                 | 0.52341       | 0.00336                |
| 0.054             | 0.3889        | 0.0028                 |
| 0.086             | 0.37443       | 0.0034                 |
| 1.02              | 0.30987       | 0.0012                 |
| 1.58              | 0.30844       | 9.48E-4                |
| 2.43              | 0.30831       | 8.025E-4               |

| 316L Dose (dpa) | $T$-parameter | Error in $T$-parameter |
|-----------------|---------------|------------------------|
| 0               | 0.49683       | 0.0032                 |
| 0.054           | 0.41973       | 0.00762                |
| 0.084           | 0.40503       | 0.0095                 |
| 3.56            | 0.35322       | 9.8E-4                 |
| 10.3            | 0.33135       | 0.00145                |

The dependences of $T$-parameter on dose for each type of material are shown on figures 2 and 3.
4. Conclusions
We investigated the change of $T$-parameter, which is calculated as the ratio of the number of counts in the wings of the Doppler-broadened 511 keV peak to the number of counts in the center of the peak. $T$-parameter is related to the density of defects in the sample of interest and generally higher defect densities induce smaller $T$-parameter.

As a result we saw that for 316L, the $T$-parameter drops quickly after low dose and monotonically to higher doses up to 10 dpa. In 9Cr1Mo also drops quickly for low doses but saturates for doses above 1 dpa. In addition, for both types of steel, we see that the biggest changes are for doses less than 0.05 dpa. The reason that the 9Cr1Mo shows a saturation in defect density after low dose compared to what is observed in the 316L is that the 9Cr1Mo has a much finer microstructure which would serve to act as sinks for defects leading to the observed saturation in defect density at a lower dose.

In order to understand the early saturation in 9Cr1Mo $T$-parameter (~1dpa) PALS studies will be performed, to compose defect size evolution as a function of dose.

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References
[1] Selim F A, Wells D P, Harmon J F, Scates W, Kwofie J, Spaulding R, Duttagupta S P, Jones J L, White T and Roney T 2002 Nucl. Instr. Meth. B 192 197
[2] Sencer B H, Garner F A, Gelles D S, Bond G M and Maloy S A 2002 J. Nucl. Mater. 307-311 266
[3] Sencer B H, Bond G M, Hamilton M L, Garner F A, Maloy S A and Sommer W F 2001 J. Nucl. Mater. 296 112
[4] Wells D P, Hunt A W, Tchelidze L, Kumar J, Smith K, Thompson S, Selim F, Williams J, Harmon J F, Maloy S A et al 2006 Nucl. Instr. Meth. A 562 688