Effects of proton irradiation on Chinese domestic RPV steels by variable energy positron annihilation spectroscopy

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Abstract. The effects of proton irradiation on Chinese domestic reactor pressure vessel (RPV) A508-3 steels have been investigated by variable energy positron annihilation spectroscopy. The samples were irradiated using 110 keV protons at doses ranging from 0.2 to 2.0 × 10 17 cm -2 at room temperature. Defect profiles were analyzed by measuring the S parameter as a function of incident positron energy from 0.25 to 26keV, corresponding to mean depths of up to ~940nm. It is interesting to note that the S-parameter increases rapidly with increasing dose, which implies that matrix damage has a direct relation to dose-dependent effects in microstructural evolution.

1. Introduction
Positron annihilation spectroscopy (PAS) has been widely used as a probe for defect studies at the atomic dimension in solids, such as vacancies, vacancy clusters, dislocations, and nanometer-scale voids. The use of variable energy positron beams allows one to probe the depth profile of solids, and is particularly suitable for analysis of surface and near-surface defects [1]. PAS investigations of irradiation-induced defects in reactor pressure vessel (RPV) steels have been reported [2, 3]. Irradiation-induced embrittlement of RPV steels is one of the important degradation phenomena in the long term operation of light water reactors. Embrittlement is considered to be mainly caused by irradiation-induced changes in the microstructure such as the formation of solute nanoclusters, matrix defects (due to irradiation-induced point defect clusters), and dislocation loops [4-6]. However, despite the extensive research, the details of the microstructural changes are not well understood. In the present study, proton irradiation induced defects in Chinese domestic A508-3 steels were investigated by slow positron beam Doppler broadening spectroscopy.

2. Experiment
The domestic A508-3 steels were fabricated by China Firstheavy Industries. The forgings of these alloys were heated to 890°C and kept for 7h, followed by quenching into water, then reheated to 650°C and kept for 10h, and then cooled in the air. The chemical composition of these steels is shown in Table 1. The samples of 15×15×1 mm 3 were cut from as-received materials for PAS measurements. The sample surfaces were carefully mechanically ground on successively finer abrasive papers, and
then polished using diamond polishing suspensions in order to remove the effects of surface damage induced by artifacts.

| Element | C    | Si    | Mn    | S    | P    | Cr   | Ni    | Cu    | Mo    | V    | Total |
|---------|------|-------|-------|------|------|------|-------|-------|-------|------|-------|
| A508-3  | 0.167| 0.193 | 1.35  | 0.002| 0.005| 0.086| 0.738 | 0.027 | 0.481 | 0.007| 3.056 |

A set of samples were irradiated by protons with the aim to simulate the radiation damage and embrittlement of real RPV steels under neutrons irradiation in nuclear reactors. The irradiation was performed at a terminal chamber of the ECR source 320kV high-voltage platform in Institute of Modern Physics (IMP) in Lanzhou, China. The samples were irradiated using 110 keV protons at doses ranging from \(2.0 \times 10^{15}\) cm\(^{-2}\) at room temperature.

The Doppler broadening parameters \(S\) and \(W\), linked to the annihilation of positrons by low- and high-momentum electrons, respectively, were measured for slow positrons incident with energies \(E\) from 0.25 to 26 keV by a high-purity Ge detector (\(\sim 1.64\) keV resolution at 511 keV). Open-volume defects such as vacancies and voids are populated by lower-momentum electrons, and thus have relatively large \(S\) and small \(W\) parameters.

3. Results and Discussion

Fig.1 shows the \(S\) and \(\Delta S/S\) parameters \(\Delta S/S = (S_{irradiated} - S_{unirradiated})/S_{unirradiated}\) as a function of incident positron energy (mean implantation depth) in as-received (unirradiated) and irradiated samples. The mean depth of the annihilating positron from the surface is calculated using the following established equation:

\[
Z(E) = \left(4 \times 10^3 / \rho \right) E^{1.6}
\]

Where \(Z(E)\) is expressed in units of nanometers, \(\rho\) is the density in units of kg/m\(^3\) (In the calculation we used the density of pure iron with a value of \(7.86 \times 10^3\) kg/m\(^3\)), and \(E\) is the incident positron energy in keV. The top axis in figure 1 gives the calculated mean depth from the surface of the annihilated positron, according to equation (1). The \(S\) parameter for the irradiated sample became much larger than that for the unirradiated ones, which clearly indicates that positrons detect the presence of vacancy-type defects generated during proton irradiation.

Figure 1 \(S\) parameter and \(\Delta S/S\) as a function of incident positron energy (mean implantation depth) unirradiated and irradiated steels.
A simple trapping model can be employed. The fraction of positrons annihilating in the bulk ($f_b$), in the surface ($f_s$) and of those trapped and annihilated at defects ($f_d$) can be expressed as follows:

$$S = f_sS_s + f_dS_d + f_bS_b$$  \hspace{1cm} (2)

where $S_s$, $S_d$ and $S_b$ are the $S$ parameters corresponding to the surface, the defect and the bulk, respectively, and $f_s$, $f_d$ and $f_b$ are the fractions of positron annihilated in the same three states ($f_s + f_d + f_b = 1$). As can be seen in figure 1(a), $S$ for the unirradiated sample decreases from its surface value to its bulk value as positron energy increases, as $f_s$ decreases from 1 to 0. However, for irradiated samples, there exist ranges of positron energy (such as between 3 and 14 keV) over which the essentially constant $S$ value can be directly attributed to damaged region induced by proton irradiation. At higher positron energy, $S$ increases with energy from $S_d$ to $S_b$, as $f_d$ decreases from 1 to 0 and $f_b$ increases from 0 to 1. Implanted and thermalized positrons annihilate either in bulk or at the trapping sites.

When the samples were applied to different dose, the $\Delta S/S$ parameter increases with the dose, and the increases in the $\Delta S/S$ parameter as a function of mean implantation depth appear a peak value at $-12$ keV, corresponding to the depth of about 270 nm (see figure 1(b)). Fig.2 shows the proton range distribution and the target displacement profile after proton irradiation with 110 keV protons by SRIM 2006 calculations. The cascades region corresponds to the region where incident ions mainly interact with atoms of the solid via nuclear collisions and finally stop into the lattice. The tracks region is localized between the surface and the nuclear cascades region, and corresponding to the zone where ions slow down mainly by electronic energy loss processes. The other part is non-implanted region. The projected range of hydrogen was at the depth of about 550 nm. Compared to figure 1(b) and figure 2(b), both have a similar peak. The main vacancy source is located in the cascades region, however, the $\Delta S/S$ parameters are higher for the tracks region than for the cascades region, i.e. their peaks move to lower positron energy, only half of the projected range of hydrogen. This can be explained by the formation of vacancy-H complex. Ishizaki et al. [7] reported that vacancy–H complexes and vacancy–He complexes were introduced in Fe irradiated with H or He ions. A large number of hydrogen atoms were strongly trapped at vacancies in the cascades region and the vacancy–H complexes formed [8]. As mentioned above, H ions were implanted to depths of about 550 nm; surface-to-peak depths were about 270 nm. Therefore, a large number of H atoms were strongly trapped at vacancies in the cascades region. The H atoms recombined with some vacancy clusters to form vacancy cluster–H complexes. When positrons are trapped at vacancy-type defects coupled with H, the value of $S$ is lower than that of vacancy-type defects. Therefore, the $S$ parameter of cascades region is lower than that of tracks region due to the suppression of vacancy clustering by vacancy trapping of H near the projected range of H. The number of H atoms in the cascades region is less than that in cascades region. Thus, $S$ parameter does not significantly decrease in the tracks region.

![Figure 2](results.png)

**Figure 2** Results obtained from SRIM 2006 calculations for proton irradiation with 110 keV protons showing: (a) the proton range distribution during irradiation and (b) the target displacement profile after irradiation.
Information about the number of defect types could be revealed by plotting the $S$-parameter as a function of the $W$-parameter ($S$-$W$ plots). Fig.3 shows that $S$- and $W$- values for all samples fall on a common locus, which only consists of one straight-line segment. This suggests that only one type of defect (open-volume defect) is present in all measurement both in unirradiated and irradiated steels. Therefore, it is interpreted that an increase in $S$-parameter in Fig.1 mainly comes from the formation of defects and voids near the surface during proton irradiation. Compared to unirradiated sample, the relative change of $\Delta S/S$ parameter in the most heavily irradiated sample (see Fig.1) exceeds 9%, which means that there exist large-size vacancy clusters or vacancy cluster-H complexes induced due to proton irradiation. Further investigations of annealing experiment at elevated temperature are in progress and will be reported in the future.

![Figure 3](image)

**Figure 3** $S$ parameter as a function of the $W$ parameter for unirradiated and irradiated steels. ($S$ is from figure 2 data)

4. Conclusions

Slow positron beam Doppler broadening measurements show that $S$ parameter increases evidently with dose after proton irradiation owing to open-volume defects induced by proton irradiation. When positrons are trapped at vacancy-type defects coupled with hydrogen, the value of $S$ is lower than that of vacancy-type defects. Therefore, the $S$ parameter of cascades region is lower than that of tracks region due to the suppression of vacancy clustering by vacancy trapping of hydrogen near the projected range of hydrogen.

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