Positron annihilation Doppler broadening study of the helium-implanted (ODS) EUROFER

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Positron annihilation Doppler broadening spectroscopy (DBS) measurements were conducted to investigate the evolution of radiation-induced open-volume defects in Eurofer97 steel and its ODS variant. Helium-implanted samples have been investigated by slow positron beam technique. With this approach, we were able to obtain a significant amount of positron lifetime and Doppler broadening data and evaluate them with respect to the dpa and helium concentration effects. The results of the experiment confirm a more pronounced creation of growing helium-vacancy agglomerations for the Eurofer steel, comparing to its ODS variant. The bubble-to-void transformation was indicated by the change in the trend of the S-parameter vs. dpa plot of the Eurofer sample. In the case of the ODS Eurofer, the coalescence of helium bubbles, accompanied by saturation of the S-parameter, was not so evident. The present study provides the first experimental DBS data, showing a retarded formation of cavities, caused by the presence of oxide dispersoids in the ODS steel.

KEYWORDS: Eurofer, ODS Eurofer, positron annihilation; helium bubbles; gas-filled cavities

1. Introduction

Production of hydrogen and helium in metals during irradiation in the nuclear facilities changes the recovery processes within the crystal lattice and results in much more pronounced radiation effects at the same fluences [1]. Recent work of Dai et al. [2] reviews microstructural changes of ferritic/martensitic (f/m) steels after irradiation in spallation target environments with high production rates of hydrogen and helium. Numerous TEM studies reviewed therein describe the formation of small 1-3 nm helium bubbles in the specimens irradiated at temperatures above 180°C to doses above 10 dpa and 500 appm He. These bubbles grow with irradiation temperature from small helium-vacancy agglomerates and, together with defect clusters and dislocation loops, affect the physical and mechanical properties of the irradiated material [3]. Based on extensive positron lifetime data on several f/m steels irradiated in STIP program of the Swiss spallation neutron source (SINQ), three stages of the evolution of helium-vacancy agglomerations were determined. Stage 1, from lowest irradiation temperature ~80°C to about 150°C, where the mean positron lifetime (MLT) decreases. This decrease of positron lifetime is due to the migration of helium into existing defects, which dominates
over the production of radiation-induced defects; Stage 2, from 150 °C to 300 °C, where
the MLT rapidly increases due to growth of vacancy agglomerates, driven by the
absorption of new radiation-induced vacancies; and Stage 3 from 300°C and beyond
where the MLT increases, but slower than that in Stage 2. This was attributed to the
coalesscence of helium bubbles, which decrease the number density of open-volume
defects and thus reduces the positron trapping at these defects.

While the size of the bubbles seems to generally increase with temperature, the
number density of the bubbles varies, depending on the actual temperature range and/or
the irradiation dose respectively. Many recent studies reported a saturation or even
decrease of the number density of growing helium bubbles at high irradiation
temperatures (high dpa) [4,5]. This points to a coalescence of large open-volume defects,
a mechanism which has been suggested for the third stage of the evolution of positron
lifetime reported in [2]. According to Surh et al. [6], at high temperatures, the coalescence
of small clusters greatly reduces the total number of helium bubbles and voids, which is in
an agreement with the positron annihilation spectroscopy (PAS) studies, where a decrease
of positron trapping at large vacancy clusters was observed [2].

This work utilizes a depth-profiled slow-positron beam measurement on the
characterization of different regions of the implantation profile in He-implanted samples
of the Eurofer [7] steel and its ODS variant [8]. With this approach, we were able to
obtain a significant amount of Doppler broadening data and from a single He-implanted
sample and to evaluate them with respect to the dpa and helium concentration.

2. Experimental

Two reduced-activation ferritic/martensitic (RAFM) steels were investigated, namely 9% CrWVTa alloy - Eurofer 97 and its ODS variant. The nominal chemical
composition of Eurofer, as well as basic material characteristics, can be found in [7].
The ODS variant was made by the mechanical alloying of the base steel sputtered in an
inert atmosphere with 0.3 wt.% yttrium oxide powder [8].

Samples prepared by electrical discharge cutting and mechanical polishing
underwent two different irradiation experiments. Two samples of both investigated
materials were implanted by 500 keV He$^{2+}$ ions with a fluence $10^{22}$ m$^{-2}$ at the former
linear accelerator of the Institute of Nuclear and Physical Engineering, Slovak
University of Technology. Irradiation temperature was kept below 70°C [9]. The
number of displacements per atom (dpa), calculated according to NRT model [10] and
averaged over 1.6 µm region was 6.5 with a peak value of 29.5. The dpa profile
including helium ion distribution and He-to-dpa ratio as obtained from SRIM code is
shown in the Fig.1.
3. Results and discussion

Doppler broadening spectroscopy (DBS) on He-implanted samples was performed at the Aalto University, Finland and preliminarily reported in [9]. DBS spectra were recorded by one HPGe detector with Gaussian resolution function of 1.24 keV. The energy windows for calculation of the S parameter was ±0.83 keV from the 511 keV annihilation peak. The energy of the monoenergetic positron beam was ranging between 0.5 – 36 keV, probing the region 1.7 nm to 1579 nm (mean implantation depth [11]) below the surface. This corresponds to a displacement damage range of 0.5 – 30 dpa, helium concentration range of 200 - 5×10^5 appm and to He-to-dpa ratio range of 200 - 3×10^4. The relative S parameter plotted as a function of depth (Fig.2a) and He concentration (Fig.2b) was obtained as a ratio to the un-implanted (as-received) material. One can see a distinct peak in the depth profile of the S parameter (Fig.2a). While the position of this peak is in a good agreement with the simulation (Fig.1), the width of the peak is much broader than the predicted peak. The broadening of the damage region or eventually the shift towards the surface in comparison to the SRIM simulation occurs, due to the presence of additional scattering sites for the incoming ions, such as nanoclusters, precipitates, and grain boundaries [12]. These are not considered in the theoretical calculations.
Fig. 2. Relative S parameter obtained from the slow positron beam experiments on materials implanted by 500 keV He\(^{2+}\) ions with a fluence \(10^{22} \text{ m}^{-2}\), as a function of depth (a) and as a function of helium concentration (b).

The depth profile of the S parameter provides a good insight into the radiation-induced production of vacancy-type defects. A somehow better understanding can be, however, obtained when the data are plotted with respect to the helium concentration (Fig. 2b). Such plot reveals distinct behaviour of the Eurofer and its ODS variant. As discussed earlier, a slower increase or saturation of positron lifetime and/or S parameter, following the previous rapid increase, indicate the growth of vacancy agglomerations by coalescence. Coalescence reactions continually, preferentially consume the smaller, more mobile clusters. This stage of the evolution of 3D open-volume defects is often connected to so-called bubble-to-void transition [13]. It is characterized by a presence of stable voids and steady volumetric swelling, which does not depend anymore on the helium supply [6]. The increase of size and the reduction of the number density of voids results in a saturation/decrease of positron trapping. Such behaviour can be clearly seen in the S parameter profile of the Eurofer. In the case of the ODS Eurofer, the saturation is hardly visible in the plot.

In addition to the distinct behaviour of the studied materials discussed above, one can see a different evolution of the S parameter also in the low-dpa (low He concentration) data points obtained from the measurements on the investigated materials. While the first stage of the evolution of helium-vacancy agglomeration, with positron trapping decreasing with dpa, can be clearly seen in the Eurofer sample, such decrease cannot be seen in the ODS Eurofer data. This finding is rather surprising since the presence of oxide dispersoids in the later material was expected to contain more defects to be potentially filled with helium, comparing to conventional f/m steel. It would suggest a more pronounced decrease of positron trapping at defects in the initial stage of the implantation profile. However, the opposite behaviour was observed, when the S parameter did not decrease in the ODS Eurofer at all. Further study is required to explain this mechanism.
4. Conclusions

Positron annihilation Doppler broadening measurements were conducted to investigate the effect of yttria nanofeatures on the formation of large vacancy agglomeration in He-implanted 9% CrWVTa alloy Eurofer 97. Relatively large number of data points obtained from single implanted sample using variable energy positron beam, enable to study effects of dpa and helium concentration. The results confirm a more pronounced creation of growing open/volume defects for the Eurofer steel, comparing to its ODS variant. The bubble-to-void transformation was indirectly but clearly confirmed for the Eurofer. In the case of the ODS Eurofer, the coalescence of helium bubbles, accompanied by saturation of the S parameter, was not so apparent. This present study provides unique experimental positron annihilation data, showing the onset of the void formation in the oxide-dispersion strengthened steels.

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