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Experimental studies of irradiated and hydrogen implantation damaged reactor steels

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Radiation degradation of nuclear materials can be experimentally simulated via ion implantation. In our case, German reactor pressure vessel (RPV) steels were studied by positron annihilation lifetime spectroscopy (PALS). This unique non-destructive method can be effectively applied for the evaluation of microstructural changes and for the analysis of degradation of reactor steels due to neutron irradiation and proton implantation. Studied specimens of German reactor pressure vessel steels are originally from CARINA/CARISMA program. Eight specimens were measured in as-received state and two specimens were irradiated by neutrons in German experimental reactor VAK (Versuchsatomkraftwerk Kahl) in the 1980s. One of the specimens which was in as-received and neutron irradiated condition was also used for simulation of neutron damage by hydrogen nuclei implantation. Defects with the size of about 1–2 vacancies with relatively small contribution (with intensity on the level of 20–40 %) were observed in "as-received" steels. A significant increase in the size of the induced defects due to neutron damage was observed in the irradiated specimens resulting in 2–3 vacancies. The size and intensity of defects reached a similar level as in the specimens irradiated in the nuclear reactor due to the implantation of hydrogen ions with energies of 100 keV (up to the depth <500 nm).

1. Introduction

A Reactor Pressure Vessel (RPV) is a crucial structural component in a nuclear power plant (NPP) which has to resist the effect of neutron damage, high pressure and temperature and cyclic stress during NPP operation. Replacing the RPV is generally not feasible and therefore determines the overall life of the NPP. The safety and life extension of NPPs is a very actual issue nowadays. It is not only necessary to know the condition and degradation level of RPVs in detail but also to predict their behavior trend towards the future. In this paper, we focus on the evaluation of German RPV steels from positron annihilation point of view.

2. Specimens

All specimens (two pieces from each material assembled in sandwich set-up) were delivered from AREVA NP GmbH Erlangen and belong to the family of commercial RPV steels used since the 70's. These steels have been comprehensively studied in research programs CARISMA [1] and CARINA [2]. The irradiation facility VAK (Versuchsatomkraftwerk Kahl), which is an experimental boiling water reactor, was chosen mainly due to the fact that it produces very similar spectra of neutrons as in German commercial pressurized water reactors (PWR). The irradiation temperature ranged between 280 and 290 °C. The chemical composition of the studied steels is listed in Table 1.

Two samples from different parts of the bulk material were cut from the irradiated materials. In Table 2 neutron fluences and activities of the measured specimens are listed. The irradiated materials differ in chemical composition mainly in Copper and Nickel content. Content of Cu and Ni should affect the final radiation damage inflicted by the negative impact of these impurities on radiation and mechanical properties.

3. Experimental technique – PALS

Positron annihilation lifetime spectroscopy is a well-established non-destructive spectroscopic method for evaluation of defect-size (size of clusters) in materials and their density described by positron annihilation intensity. Sensitivity of PALS is relatively high

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Table 1
Chemical composition of CARINA/CARISMA studied steels in wt. %.

Specimen	German PWR generation	C [%]	Si [%]	Mn [%]	P [%]	S [%]	Cr [%]	Mo [%]	Ni [%]	Cu [%]
P370 WM	1	0.08	0.15	1.14	0.015	0.013	0.74	0.60	1.11	0.22
P16 WM	3	0.05	0.15	1.14	0.012	0.007	0.07	0.46	1.69	0.08

Table 2
Neutron fluence and measured activity on 24.01.2013.

Specimen	P370WM-D77	P370WM-D161	P16WM-S103	P16WM-GS67
Fluence [cm^{-2}]	2.21×10^{19}	2.23×10^{19}	1.16×10^{19}	4.81×10^{19}
Activity [kBq]	12.85	97.31	40.09	16.6

with the ability to recognize one defect per 10^7 atoms [3]. For example, an electron microscope is not able to sense defects that PALS can. Therefore, PALS provides unique information for microstructural studies of selected materials before and after external treatment (irradiation, annealing, etc.).

Principle of PALS (Fig. 1) is based on positron-electron (e^+—e^-) annihilation where the positron is produced by β^+ decay of the sodium source during which aγ-photon with energy of 1274 keV is emitted providing a start signal for PALS technique. The positron afterwards thermalizes (~1 ps) and diffuses into the specimen (~100 nm). Subsequently the positron is trapped in a vacancy-type defect due to an attractive potential caused by the lack of positively charged nuclei (repulsive forces). After some time, depending on the defect size and electron density, the positron recombines with an electron and annihilates producing two γ-photons with energies 511 keV, which provide the stop signal for PALS equipment [4].

Our PALS equipment has a time resolution of about 190 ps for 2 detector set-up and about 170 ps for 3 detector set-up. Non-radioactive specimens (as-received and implanted) were measured using the two detector set-up and irradiated specimens were measured with three detectors due to induced radioactivity from ^{60}Co caused by transmutation process. Our source of positrons ^{22}Na is deposited on a Kapton foil. Kapton was used, mainly because of the single lifetime which positrons can obtain in this material simplifying the fitting process. For evaluation and interpretation of measurements we used LifeTime 9 which gives us the following parameters: τ_1-reduced positron lifetime in bulk, τ_2-positron lifetime in defects and τ_3 is the annihilation outside of the sample (air).

4. Hydrogen ion implantation

Ion implantation is an effective method for the study of basic effects of irradiation on the material, such as volume increase, creep and segregation of chemical elements in the matter. We chose hydrogen nuclei for ion implantation. We chose hydrogen because it has the same mass of protons and neutrons [5].

The energy of implanted hydrogen atoms was set to 100 keV and was carried out utilizing our linear accelerator localized at the Institute of Nuclear and Physical Engineering in Bratislava. The simulation from SRIM code for hydrogen implantation in the RPV steel shows that the maximum depth of damage is about 0.64 µm and the maximum damage is about 0.44 µm creating about 16.8 vacancies per ion (Fig. 1). Implantation was performed in three levels where the third level is equal to the damage, from number of particles point of view, which would be created by neutron irradiation (Table 3). It is necessary to note that dose calculations in our experiments are just local values in the layout of implantation range (depth range up to 640 nm). That is the main difference between hydrogen implantation and neutron irradiation in this case because the damage caused by ions is only in the surface layer and neutron damage is spread throughout the volume of the specimens.

5. PALS results

PALS results concerning German specimens in as-received state were published in Ref. [6]. In this work, results of measured specimens in as-received state show that small defects present in the specimens were induced during the fabrication process. They were identified as a composition of dislocation and mono-, di- and 3-vacancies according to obtained data [7]. In the present article we focused on neutron irradiated specimens and ion implantation treated specimen P370 WM and their mutual comparison in emphasis on defect size and defect intensity.

5.1. *PALS results from neutron irradiated German RPV steels*

The well-known hardening effect caused by irradiation which depends on neutron fluence is obviously more pronounced for the specimen materials with higher Cu (P370 WM) and Ni (P16 WM)

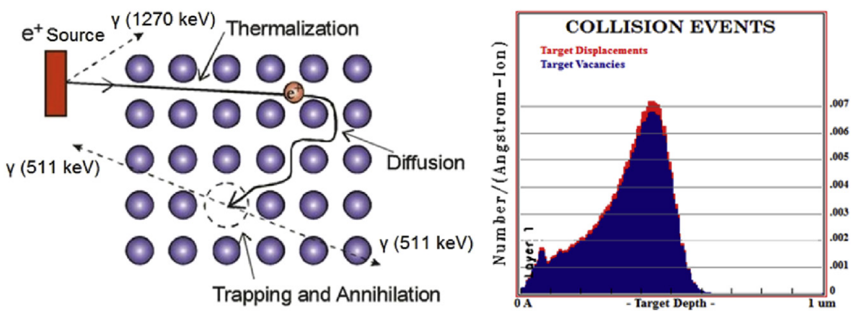

Fig. 1. Scheme of PAS and SRIM code simulation of implantation.