EVOLUTION OF RADIATION INDUCED DAMAGE IN HIGH COPPER WESTERN REACTOR PRESSURE VESSEL STEELS

Stanislav Pecko[a*], Vladimír Slugeň[a]

[a]Institute of Nuclear and Physical Engineering, Slovak University of Technology, 81219 Bratislava, Slovakia

[*] *E-mail:stanislav.pecko@stuba.sk*

Received 07 May 2015; accepted 12 May 2015

1. Introduction

The reactor pressure vessel (RPV) is the crucial component of nuclear power plants (NPP) and it must be able to withstand the direct effect of neutron irradiation from fission, high pressure, high temperature and cyclic stress during the whole operation of NPP. It is not generally feasible to replace the RPV which therefore determines the overall lifetime of the nuclear facility. The strong emphasis is placed on the quality of the steels used in RPV focused onto radiation resistance, the impurities present in the material, proper metallurgy processes and resistance to change of the physical properties of the steels.

Western RPV steels were studied by Positron Annihilation Lifetime Spectroscopy (PALS) with emphasis to defect size and its concentration in the crystal lattice. PALS is unique non-destructive spectroscopic method with great sensitivity to vacancy type of defects. German and Japan RPV steels were irradiated in nuclear reactors more than 25 years ago which made these steels suitable to measurements without special handling and protection. German steel P370WM was implanted by hydrogen ions to obtain similar damage to the neutron irradiation. In this article the main focus is aimed to evaluation of neutron damage caused to as-received specimens with taking into account the chemical composition of each steels.

Nowadays the NPPs lifetime prolongation becomes a very actual issue. It is necessary to know in detail the status of devices, their condition and attrition to estimate their trend for the future. One of the most important issues is to determine the degree of the degradation process of the RPV material. The neutron embrittlement process is a crucial problem with structural and safety issues in the operation of NPP.

2. Studied specimens

Two different families of RPV steels were measured by PALS – two Japan model steels corresponding to ASTM A533B cl. 1 steel specification [1] and two German commercial steels from CARINA/CARISMA program [2, 3].

Japan steels, marked as JRQ and JPA, were manufactured in Japan according to the order of the International Atomic Energy Agency (IAEA) and were used in various studies focused mainly on neutron embrittlement studies with various levels of irradiation and annealing conditions since 1980s. The main distinction of these steels is in the copper content, which is in both cases higher than the actual recommended level 0.008% [1, 4]. The JRQ steel is considered as low-Cu (0.15 wt%) and JPA as high-Cu (0.29 wt%) material.

German steel, marked as P370WM, was delivered from AREVA NP GmbH Erlangen and belong to commercial RPV steels used since 70-ties. There were just two irradiated materials and each of them has two different cuts from the same bulk. The P370WM is weld material with the high content of Copper (0.22 wt%). This limit of the Cu content should affect the final radiation damage because of the negative impact of these impurities on radiation and mechanical properties. The chemical composition of the studied steels is listed in Tab. 1 and the neutron fluence is listed in Tab. 2.

Tab. 1: *Composition of the materials in wt% (balance Fe)*

Steel	C	Mn	Si	Cr	Ni	Mo	V	S	P	Cu
JRQ	**0.20**	**1.42**	**0.23**	**0.13**	**0.80**	**0.52**	**0.008**	**0.005**	**0.02**	**0.15**
JPA	**0.18**	**1.33**	**0.26**	**0.16**	**0.73**	**0.55**	**0.006**	**0.004**	**0.02**	**0.29**
P370WM	**0.08**	**1.14**	**0.15**	**0.74**	**1.11**	**0.60**	**-**	**0.013**	**0.015**	**0.22**

Tab. 2: *Irradiation conditions for Japan and German steels*

Material code	Neutron fluence / 10^{18} cm^{-2} (E > 0.5 MeV)	Flux density / 10^{12} cm^{-2} s^{-1} (E > 0.5 MeV)	Activity [kBq] (on 24.01.2013)
JRQ-I1	**10**	**0.15**	**-**
JRQ-I2	**77**	**3.0**	**-**
JRQ-I3	**139**	**5.4**	**-**
JPA-I1	**10**	**0.15**	**-**
JPA-I2 a-e	**80**	**3.1**	**-**
JPA-I3	**143**	**5.5**	**-**
P370WM-D77	**22.1**	**-**	**12.85**
P370WM-D161	**22.3**	**-**	**97.31**

Ion implantation is an effective method for the study of the basic effects of irradiation on the material. We chose an ion implantation with hydrogen nuclei. The choice seems to be ideal from the same mass of protons and neutrons point of view [5]. Energy of implantation was considered at 100 keV and it was carried out at our linear accelerator 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 in the creation of 16.8 vacancies per ion. Implantation was performed in three levels of irradiation where the third level is equal, from number of particles point of view, to neutron irradiation. The implantation parameters are in Tab. 3.

Tab. 3: *Overview of implanted parameters in specimen P370WM*

Hydrogen implantation	Implanted dose [C/cm^2]	Number of implanted ions [cm^{-2}]	Dose in implanted region [dpa]
1. level	**0.10**	**6.24×10^{17}**	**1.980**
2. level	**0.82**	**5.12×10^{18}**	**16.235**
3. level	**3.20**	**2.00×10^{19}**	**63.354**

3. Experimental technique – PALS

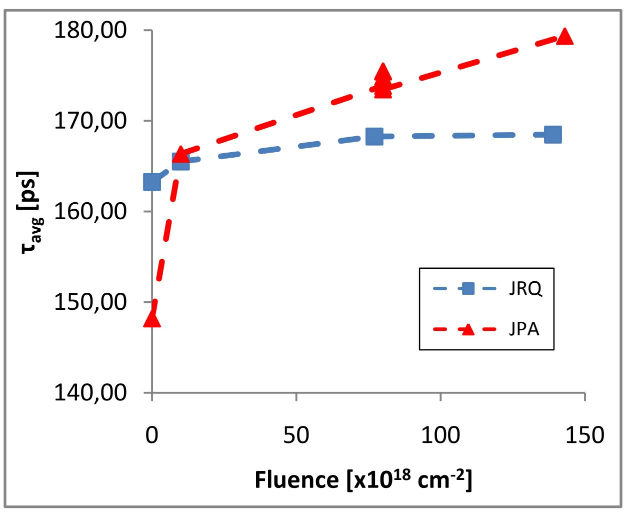
Positron annihilation lifetime spectroscopy is a well-established non-destructive spectroscopic method for evaluation of defect-size (size of clusters) in materials and its density by positron annihilation intensity. Resolution of PALS is relatively very high with the ability to recognize one defect per 10^7 atoms [6, 7]. These defects cannot be seen by an electron microscope, however PALS can, and in this sense it provides a unique type of information for microstructural studies of selected materials before and after external treatment (irradiation, annealing, etc.) [4].

Measurement of neutron irradiated steels by PALS has to be performed by the triple-coincidence method due to the influence of the ^{60}Co [8, 9], which was induced in the steels during irradiation. Nucleus of the ^{60}Co emits two γ-photons with energy of 1173 keV and 1332 keV respectively, and can be detected as a false start signal. Our PALS set-up at the Institute of Nuclear and Physical Engineering is built in an air conditioned casing with stable temperature. FWHM (Full width at half maximum) value for our set-up is stable about 175 ps.

4. Experimental results and discussion

All specimens were measured by PALS and the results were evaluated according to standard trapping model [10-13]. Size of vacancies is estimated from calculated data for pure iron [14]. In as-received state of all steels the main defect type corresponds mainly to divacancies.

The main focus of this article is evaluation of the effect of irradiation to the creation of induced defects in the crystal lattice and the influence of irradiation fluence to the defects growth. As evaluation tool was chosen the average lifetime of positrons (τ_{avg}) which can be interpreted as a qualitative PALS parameter. Average positron lifetime is calculated from measured data and includes only annihilation in material, components τ_1 (annihilation in bulk) and τ_2 (annihilation in defects).



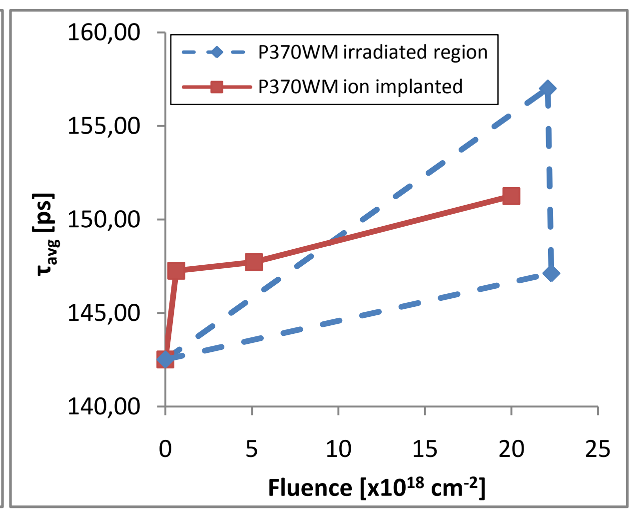


Fig. 1: *Average positron lifetime depending on the neutron fluence for Japan steels*

Fig. 2: *Average positron lifetime depending on the neutron fluence for German irradiated and implanted P370WM steel*

In the Fig. 1 is shown how the average lifetime is increasing with fluence. As you can see, τ_{avg} of JPA steel has grown rapidly after first level of irradiation, conversely τ_{avg} of JRQ steel has increased just slightly. The reason of this contrast is most probably due to different copper content of the steels. Debarberis shows in Fig. 3 [15] that the essential part of damage in the steel, from ductile-brittle transition temperature point of view, is caused in the beginning of irradiation, mostly due to precipitation of Cu in the crystal lattice. Copper atoms precipitate to the grain boundaries due to irradiation, where the movement of dislocations is hindered by solute obstacles which are responsible for embrittlement of the material. This approach is in the coincidence with our results of measured specimens with higher Cu content.

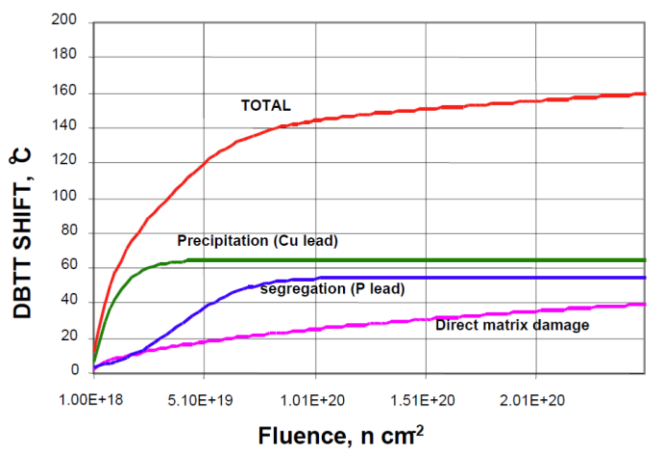


Fig. 3: *Scheme of irradiation embrittlement mechanisms calculated by a semi-mechanistic analytical model [17]*

The τ_{avg} parameter has also increased by the impact of the hydrogen implantation to P370WM specimen. In the Fig. 2 is clearly shown that the increase of τ_{avg} is very similar to behaviour of JPA steel, where both JPA and P370WM are steels with high-Cu content. Implantation experiment has proved that the same amount of impacted particles, hydrogen ions and neutrons, caused very similar damage from PALS point of view. The τ_{avg} of implantation has achieved the region of neutron irradiation about the fluence of 20×10^{18} cm^{-2}.

The hydrogen ion implantation experiment and results, which confirm its use to simulation of neutron irradiation, were more closely described in [16].

5. Conclusion

Copper has especially very strong influence on the sensitivity of steel to neutron irradiation. As the copper solubility limit at the irradiation temperature is very low (≈0.007% in pure iron), copper atoms have a propensity to form precipitates or clusters in RPV steels in operation [1]. The PALS is one of the non-destructive spectroscopic methods which can contribute to the complex evaluation of the RPV-steels microstructure and can in this way contribute to the nuclear safety of NPPs or their feasibility to the long-term operation.

This paper provides experimental demonstration of the evaluation of radiation induced defects in high copper RPV steels. The assumption of high increase of damage creation during first level of irradiation [15] was observed in our

PALS results from average positron lifetime point of view. This effect was detected in our high-Cu steels – JPA and P370WM, on the other hand, the JRQ steel with relatively lower Cu content was less sensitive to defect increase during irradiation. We confirmed that Cu content has crucial effect to defect creation and therefore is responsible for radiation embrittlement.

6. Acknowledgement

This article was created with the support of the Ministry of Education, Science, Research and Sport of the Slovak Republic within the Research and Development Operational Programme for the project "University Science Park of STU Bratislava", ITMS 26240220084, co-funded by the European Regional Development Fund. This article was also granted by VEGA 1/0204/13

7. References

[1] IAEA Nuclear Energy Series No. NP-T-3.11, *Integrity of Reactor Pressure Vessel in Nuclear power Plants: Assessment of Irradiation Embrittlement Effects in Reactor Pressure Vessel Steels* (2009)

[2] Hein H et al, *ASTM Int.* **6** (2009) Paper ID JAI101962

[3] Hein H et al, PVP2009-77035, *ASME Pressure Vessels and Piping Division Conference*, July 26-30 (2009), Prague, Czech Republic

[4] Slugeň V, *Safety of VVER-440 Reactors – Barriers Against Fission Products Release*, Springer, 2011, ISBN 978-1-84996-419-7

[5] Mota F, Ortiz C J, Vila R, *Primary displacement damage calculation induced by neutron and ion using binary collision approximation techniques*, Laboratorio Nacional de Fusión – CIEMAT, Madrid, First Technical Meeting on Primary Radiation Damage, IAEA Vienna, October 1-4 (2012)

[6] Grafutin V I, Prokopev E P, *Physics - Uspekhi* **45** (1) 59 - 74 (2002)

[7] Eldrup M, Singh B N, *J. Nucl. Mater.* **251** (1997) 132-138

[8] Čížek J, Bečvář F, Procházka I, *Nucl. Instrum. Meth.* A **450** (2000) 325-337

[9] Saito H, Nagashima Y, Kurihara T, Hyodo T, *Nucl. Instrum. Meth.* A **487** (2002) 612-617

[10] Saito H, Nagashima Y, Kurihara T, Hyodo T, *Nucl. Instrum. Meth.* A **487** (2002) 612-617

[11] Hautojärv P, Pöllönen L, Vehanen V, Yli-Kauppila J, *J. Nucl. Mater.* **114** (1983) 250

[12] Vehanen A, Hautojärvi P, Johansson J, Yli-Kauppila J, Moser P, *Phys. Rev.* **B25** (1982) 762.

[13] Brauer G, Sob M, Kocik J, *Report ZfK*-**647** (1990)

[14] Cizek J, Prochazka I, Kocik J, Keilova E, *Phys. stat. sol.* (a) **178** (2000) 651

[15] Debarberis et al, *Int. J. Pres. Ves. Pip.* **82** (2005)

[16] Pecko S, Sojak S, Slugeň V, *Appl. Surf. Sci.* **312** (2014) 172-175