ADVANCED CHARACTERIZATION OF IRRADIATED WWER REACTOR PRESSURE VESSEL STEELS

V. Slugeň[1], S. Sojak[1], W. Egger[2]

[1]*Institute for Nuclear and Physical Engineering, Slovak University of Technology, Ilkovičova 3, 81219 Bratislava, Slovakia*

[2]*Universität der Bundeswehr, W. von Heisenberg Weg 39, Neubiberg, Germany*

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1. Introduction

The nuclear reactor pressure vessel (RPV) is a key component if considering the safety of the nuclear power plant (NPP) operation and possible lifetime prolongation. It is necessary to ensure the integrity of the reactor pressure vessel during normal and abnormal operating conditions. The ability of RPV to resist brittle fracture is of particular importance, especially under the loss of coolant accident (LOCA) conditions. The degradation of RPV steel is a complicated process depending on many factors (thermal and radiation treatment, chemical composition, manufacturing conditions, ageing, microstructure of the RPV material, operational history, etc.).

In this paper we focus on the comparison of German and Russian RPV-steels from the positron annihilation spectroscopy (PAS) point of view, having in mind knowledge obtained from other techniques from the past [1].

2. Experimental

The chemical composition of the studied non-irradiated German steels is listed in Table 1. These steels have been comprehensively studied in the research programs CARISMA and CARINA [2]. All specimens belong to commercial reactor pressure vessel steels and vary mostly in the content of Cu, P and Ni.

Neutron irradiated specimens (Tab. 2) of western CARINA/CARISMA reactor steels were delivered to our Institute in September 2012. Due to the radioactivity of mentioned neutron irradiated specimen, the special safety requirements for transport of specimens from AREVA NP GmbH Erlangen (Germany) to the Institute of Nuclear and Physical Engineering, FEI STU Bratislava (Slovakia) were needed in form of licensed transport using marked barrel to avoid any influence of activated specimens on outer environment. Activity of radioisotope ^{60}Co in irradiated specimens to the date 24.01.2013 is shown in Table 3. Presence of other radioisotopes was not detected.

Positron annihilation spectroscopy (PAS) is used in RPV characterization since 80-ties. Its techniques were proved as very valuable to get insight on the population of vacancy-type defects that form under irradiation [3].

Tab. 1 *Chemical composition of non-irradiated CARINA/CARISMA steels.*

Material	Project	German PWR Generation	Project Code	Cu [wt.%]	P [wt.%]	Ni [wt.%]
20MnMoNi5-5 JSW	CARISMA	4 (Konvoi)	P141 BM	0.05	0.01	0.79
22NiMoCr3-7 Klöckner	CARISMA	1-2	P7 BM	0.12	0.02	0.97
22NiMoCr3-7 JSW	CARISMA	3-4	P147 BM	0.05	0.01	0.84
S3NiMo1/OP 41 TT UP, GHH	CARISMA	4 (Konvoi)	P141 WM	0.03	0.02	1.01
NiCrMo1 UP(modified)/ LW320, LW330	CARISMA/C ARINA	1	P370 WM	0.22	0.02	1.11
22NiMoCr3-7 JSW	CARINA	3	P150 BM	0.05	0.008	0.83
22NiMoCr3-7 Klöckner	CARINA	1-2	P151 BM	0.09	0.007	0.97
Molytherme Electrode Sulzer	CARINA	1	P152 WM	0.03	0.015	0.08

BM – base metal
WM – weld metal

Tab. 2 *Chemical composition of RPV steel specimens selected for PAS studies.*

Mat.	Manufacturing Type	Place of analysis	C [%]	Si [%]	Mn [%]	P [%]	S [%]	Cr [%]	Mo [%]	Ni [%]	Cu [%]	V [%]	Ta [%]	Co [%]	Al [%]	Sn [%]	As [%]	Sb [%]
P16 WM	Submerged arc welding	Mean values	0.05	0.15	1.14	0.012	0.007	0.07	0.46	1.69	0.08	0.004	0.002	0.024	0.022	-	-	-
P370 WM	Submerged arc welding	Mean values	0.08	0.15	1.14	0.015	0.013	0.74	0.60	1.11	0.22	0.010	-	-	0.013	0.013	0.014	-

Tab. 3 *Activities of ^{60}Co in neutron irradiated specimens to the date of 24.01.2013.*

Specimen	P370WM-D77	P370WM-D161	P16WM-S103	P16WM-GS67
Activity [kBq]	12.85	97.31	40.09	161.46

The CARISMA materials were irradiated in a German test reactor, the VAK (Versuchsatomkraftwerk Kahl), in the 1980s in the frame of a dedicated irradiation program in order to make provisions for future changes of RPV safety requirements and possible Long Term Operation (LTO) measures. The irradiation temperature was mainly in a range between

280 and 290 °C [2].

All 3 specimens P370 WM (two irradiated D-77 and D-161, as well as one non irradiated CD159) are from the same bulk but cut at different positions. The same is valid also for both P16 WM specimens (S103 and GS67). Unfortunately, there was not specimen from non irradiated P16 WM material.

Mean positron lifetime (MLT) includes positron annihilation in air and therefore some variables can be introduces into the data evaluation. Therefore, average lifetime was calculated from measured data, including only annihilation in components $\boldsymbol{\tau_1}$ and $\boldsymbol{\tau_2}$.

It was proved that performed irradiation treatment caused increase of positron lifetimes in studied specimens. In the case of both P370SG specimens, the increase of $\boldsymbol{\tau_{avg}}$ parameter was from 142 ps to 147ps and 157 ps, respectively. In the case of P16WM steels, the lifetime $\boldsymbol{\tau_2}$ is at the level of about 195 ps. It shows the presence of the small vacancy clusters with the size of about 1-2 vacancies. In the case of the steel P370WM, the lifetimes in defects are higher and with values of about 210 ps. It could indicate vacancy clusters of 2-3 vacancies [4].

For the first component $\boldsymbol{\tau_1}$ are the positron lifetimes from about 100 – 115 ps and this could be caused by the superposition of the reduced bulk component and another component which is expressed by a bit higher values of positron lifetimes containing some partial dislocations (shallow traps).

Considering the data from the plot of intensities assigned to the first lifetime component (I_1) and defect (I_2) structures can be seen that the intensities of defects in the case of high Ni (1,69 wt.%), low Cr 0,07 wt.% and low Cu (0.08 wt.%) P16 WM steels are at level of about 75%. This means that the smaller defects (dislocation lines and mono- or divacancies) in relatively high amount are more homogeneously distributed in the microstructure of P16 WM. In the case of steels with relatively low Ni (1,11 wt.%) but high Cr (0.74 wt.%) and Cu (0.22 wt.%) P370 WM , the intensities of defects are much lower but the lifetimes in defects are higher. So the defects are bigger (2-3 vacancies) and not so homogeneously distributed. This can be due to Cu precipitation after irradiation. According to the lifetimes values from our measurements performed on CARINA/CARISMA RPV-steels we calculated the total trapping rate κ in ns^{-1} as well as the total defect concentration c_d (the same values but in ppm). A quantitative analysis of the trapping rates and the vacancy concentration can be calculated using the two-state trapping model. The positron trapping rate for vacancy κ_v can be calculated [5] as

$$\kappa_v = \mu_v C_v = (\tau_{avg} - \tau_b)/\tau_b(\tau_v - \tau_{avg}) \quad (1)$$

$$\tau_b = (I_1/\tau_1 + I_2/\tau_2)^{-1} \quad (2)$$

where κ_v is the trapping coefficient for defects (vacancy), C_v is the vacancy concentration, τ_b and τ_v are the positron annihilation lifetime in bulk and in vacancy, respectively. The concentration of vacancies is calculated from Eq. (1) by giving the value of the specific trapping coefficient for a single vacancy in pure Fe, $\mu_v = 1.1 \times 10^{15}\ s^{-1}$. For more defects N, as clusters of vacancies, can be the specific trapping coefficient calculated as $\mu_N = N^* \mu_v$ [5]. Results are summarised in Table 4.

Tab. 4 *Calculated concentrations of vacancies in irradiated specimens.*

Specimen	P370 WM-CD159 Not irradiated	P370WM-D77 Irradiated (fluence 2.21E19 cm^{-2} E> 1MeV)	P370WM-D161 Irradiated (fluence 2.23E19 cm^{-2} E> 1MeV)	P16WM-S103 Irradiated (fluence 1.16E19 cm^{-2} E> 1MeV)	P16WM-GS67 Irradiated (fluence 4.81E19 cm^{-2} E> 1MeV)
Vacancy concentration [ppm]	1.35	3.09	2.38	12.3	11.2

We registered only a slight increase of the concentration of vacancies due to irradiation - from 1.35 to 3.09 and 2.38, respectively in the case of P370 WM. This fact is visible. Nevertheless, much higher concentration of vacancies we observed in the case of P16WM (in comparison to P370WM). Unfortunately, this concentration after irradiation can be not compared with not irradiated material, because the not irradiated P16WM was not available for measurement. There cannot be stated that the small differences in neutron fluencies caused the higher concentrations of defects in studied materials.

3. Conclusion

Based on our PAS results we stated that no large voids or vacancy cluster were formed due to irradiation in Russian as well as in German RPV steels. The performed neutron treatment of RPV steels caused at both studied materials an increased trend of defects concentration with increased neutron doses. Although these doses correspond to the neutron treatment up to 30 years of reactor operation (specimens were placed into irradiation chambers very closed to reactor core and the acceleration factor of irradiation was about 11). In the future, PAS techniques can be applied effectively also for evaluation of microstructural changes caused by extreme external loads simulating irradiation by proton implantation and for the evaluation of the effectiveness of post-irradiation thermal treatments.

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4. References

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