COMPARISON OF LIFETIME AND DOPPLER BROADENING PAS TECHNIQUES OF IRRADIATED AND ANNEALED JRQ STEEL

Stanislav Pecko**, Martin Petriska, Vladimír Slugeň

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

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

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1. Introduction

A reactor pressure vessel (RPV) is something like a heart of nuclear power plants. Heated water is transported from the nuclear reactor through a primary circuit, which is like bloodstream full of veins or arteries. The warm heart made from high-quality steel is the place, where the real spirit of the nuclear plant overlies. A vital force supplying the heart consists from a released energy from the controlled nuclear fission of uranium nuclei. But unfortunately, this intense power is accompanied with a strong stream of neutral particles, which are impacting into walls of the heart tirelessly. Other issues, such as overwhelming pressure and temperature, just enlarge demands, which are placed on the heart. The RPV cannot be operating with large quality deterioration susceptible to any failure as well as a living organisms cannot live without the healthy heart. That is the reason why the continual and systematic research of a resistance to damage of materials is still needful. Technically, it is possible to replace the RPV as well as the surgeons can change the ill heart for a healthy one. But from economic point of view it is not the reasonably achievable solution and never was carried out. A Doppler broadening of positron annihilation spectra and measurement of positron lifetimes are suitable positron annihilation spectroscopy (PAS) methods to determine the impact of neutron irradiation to qualitative changes of RPV steels from the defect distribution and chemical changes point of view.

2. Studied specimens

Studied steel of RPV steel belongs to Japan Reference Quality (JRQ) steel corresponding to ASTM A533B cl. 1 steel specification [1]. JRQ steel was manufactured in Japan according to the order of the International Atomic Energy Agency (IAEA) and was used in various studies focused mainly on neutron embrittlement studies with various levels of irradiation and annealing conditions since 1980s. Copper content of JRQ is 0.15 wt%, where the value is higher than the actual recommended level 0.008% [1, 2]. The chemical composition of the studied steel is listed in Tab. 1, the annealing conditions are listed in Tab. 2 and the neutron fluence is listed in Tab. 3.

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

Tab. 2: *Annealing conditions of JRQ-I3 irradiated specimen*

Specimen	JRQ-A1	JRQ-A2	JRQ-A3	JRQ-A4	JRQ-A5	JRQ-A6
Annealing Temperature [°C]	350	375	400	425	450	475

Tab. 3: *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)
JRQ-I1	10	0.15
JRQ-I2	77	3.0
JRQ-I3	139	5.4

3. Experimental techniques

Positron annihilation lifetime spectroscopy (PALS) is a well-established non-destructive spectroscopic method for evaluation of defect-size (size of clusters) in materials and its qualitative density by positron annihilation intensity. Resolution of PALS is relatively very high with the ability to recognize one defect per 10^7 atoms [3, 4]. 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.) [2].

Coincidence Doppler broadening spectroscopy (CDBS) is another positron-based well-established complementary spectroscopic method [5]. It is based on a measuring of annihilation energy with Doppler energy shifts by an amount of ΔE (511 keV ± ΔE). A huge amount of annihilation events is measured to give the complete Doppler spectrum, where the energy line is broadened due to the individual Doppler shifts along the annihilation direction. What CDBS gives is the information on the electron momentum distribution in the specimen. An evaluation of CDBS spectrum leads to two parameters, which describes the measured Doppler spectrum [6]. S-parameter corresponds to positron annihilations with valence electrons and W-parameter corresponds to positron annihilations with core electrons [7]. We can say that the S-parameter is sensitive to open volume defects and W is sensitive to the chemical surrounding at the annihilation site.

Measurement of neutron irradiated specimens 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. Our PALS set-up at the Institute of Nuclear and Physical Engineering (INPE) is built in an air conditioned casing with stable temperature. FWHM (Full width at half maximum) value for our PALS set-up is stable about 175 ps. Also CDBS measurements were done at INPE in the table-mounted set-up in air conditioned room.

4. Experimental results and discussion

All specimens were measured by PALS and the results were evaluated according to standard trapping model [9-12]. Size of vacancies is estimated from calculated data for pure iron [13]. PALS has been evaluated by *LT-9* and CDBS by *CBDS Tools* software. As evaluation tool for PALS was chosen the average positron lifetime (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). Likewise we used as evaluation tool an S/S_{ref} parameter for CDBS measurements. It is normalized value to the respective un-irradiated value. Related results to this article are listed in Tab. 4.

Tab. 4: *Results of CDBS and PALS (only τ_{avg})*

Specimen	As Received	I1	I2	I3	I3-A1	I3-A2	I3-A3	I3-A4	I3-A5	I3-A6
S parameter	0.3518	0.3623	0.3652	0.3642	0.3663	0.3630	0.3618	0.3580	0.3612	0.3619
W parameter	0.0314	0.0298	0.0288	0.0292	0.0287	0.0304	0.0320	0.0329	0.0319	0.0302
τ_{avg} [ps]	163.2489	165.5	168.3	168.5	167.8	167.7	164.5	162.9	161.1	160.9
$\Delta\tau_{avg}$ [ps]	1.4885	1.728	1.508	2.612	0.889	0.277	0.129	0.223	0.008	0.39

The results have been plotted to two different graphs (directly comparable are just parameters related to defects - τ_{avg} and S parameter), where first (Fig. 1) shows the irradiation dependence of τ_{avg} and S/S_{ref} parameters. As we can see in the Fig. 1, the behaviour of both, the τ_{avg} and S/S_{ref}, as a function of the fluence is very similar. There is an observable increase in the both of defect dependent parameters. Slight difference can be noticed after the second level of irradiation, where τ_{avg} is lightly still increasing and vice versa S/S_{ref} parameter is lightly decreasing.

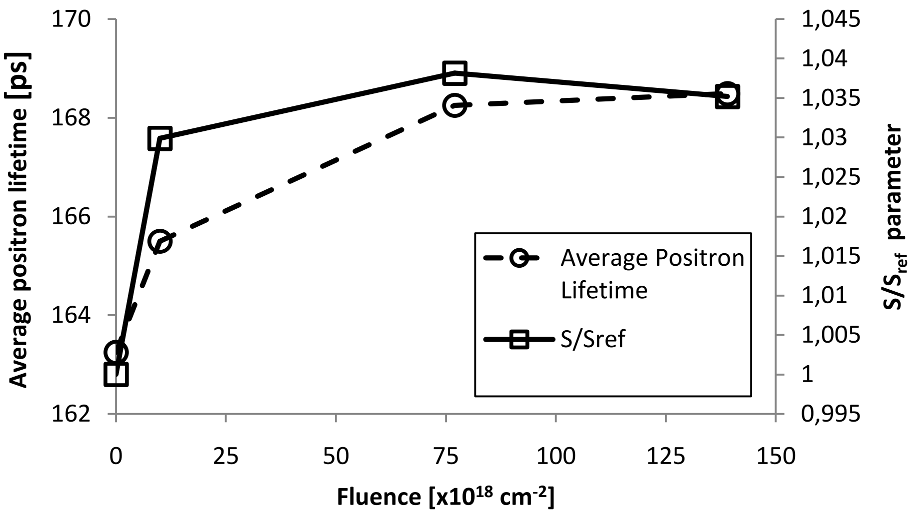

Fig. 1: *Irradiation dependence of average positron lifetime and S/S_{ref} parameter of positron annihilation spectroscopy techniques*

In Fig. 2 can be seen the behaviour of τ_{avg} and S/S_{ref} as a function of the annealing temperature of irradiated JRQ steel after third level of the irradiation ($139x10^{18}$ cm^{-2}). τ_{avg} parameter has the decreasing tendency with all of the annealing temperature range. Otherwise, S/S_{ref} parameter decreases just up to 425 °C, after that increases over again with very similar rate as in the decrease. Although during first four annealing steps is observable a similar behaviour, the last two steps shows an anticoincidence. From this point of view of PAS techniques, an ideal temperature of annealing seems to be 425 °C by CDBS and 475°C by PALS results (for annealing of RPV steels is currently recommended temperature around 475°C [14, 15]). The process of embrittlement of RPV steels is not just caused due to evolution of vacancy type defects. This process is much more complicated and broader. In JRQ steel with not insignificant amount of Cu in the chemical composition, other effects have been influencing the embrittlement - copper segregation and clustering on the one side and evolution of dislocation loops on the other side [14].

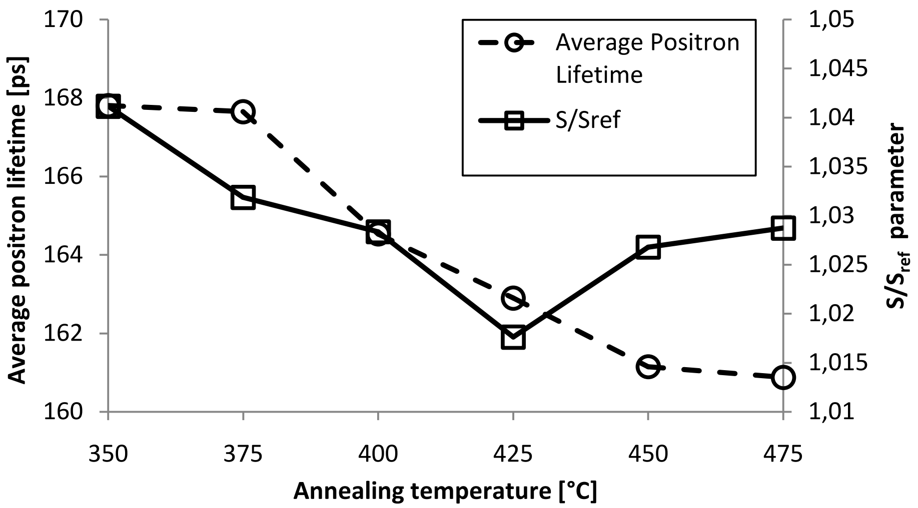

Fig. 2: *Annealing temperature dependence of average positron lifetime and S/S_{ref} parameter of positron annihilation spectroscopy techniques*

This difference can be caused by various reasons. Both PAS techniques are sensitive to vacancy type defects, but in the case of the annealing, second part of the plot shows clear discrepancy. The real reason must be find out in ongoing research which be held in a near future. Also from a decomposition of positron lifetime spectra has not been found any clue, which could explain this divergence. Specific positron lifetimes in defect and bulk components with pertaining intensities have been published in [16].

5. Conclusion

The PALS and CDBS have been done on RPV steel marked as JRQ. Both techniques are suitable non-destructive complementary methods, but they are not usually used at once and compared to each other. During the neutron irradiation the agreement of both PAS techniques have been very good and convincing. But the mismatch has been found in the annealing experiment in the last two annealing temperatures. The reason of that is still not clear and must be checked.

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