APPLICATION OF PAS AND CHARPY-V TESTS AT WWER REACTOR PRESSURE VESSEL STEELS

V. Slugeň¹, A. Kryukov², S. Sojak¹, M. Petriska¹, J. Veterniková¹, V. Sabelová¹, R. Hinca¹, M. Stacho¹

¹Institute for Nuclear and Physical Engineering, Slovak University of Technology, Ilkovičova 3, 81219 Bratislava, Slovakia ²European Commission, Joint Research Centre - Institute for Energy, PO Box 2, 1755 ZG Petten, The Netherlands E-mail: vladimir.slugen@stuba.sk

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1. Introduction

The global trend in world nuclear power industry is the lifetime prolongation of the second generation of nuclear power plants (NPP) which were originally designed up to 40 years of reactor operation. Actually, the possibility of reactor lifetime extensions to 60 years or more are evaluated. The reactor pressure vessel (RPV) is the crucial component in consideration about the possible NPP lifetime prolongation.

In the last years, several reliable and predictive modelling approaches have been developed (thanks also to EC-framework projects PERFECT, REWE and PERFORM60), according to which the assumed RPV-steel damage is generated by three major contributions and their synergisms [1]:

- i) copper rich radiation induced nano-precipitates,
- ii) phosphorus segregation at different internal surfaces and
- iii) the basic damage of the material matrix.

Nickel acting as amplifier of one or more of the three basic contributors also influences the radiation damage. Several studies [2] have suggested that carbide formation and precipitation could be important damaging mechanisms.

A number of semi-empirical laws, based on macroscopic data, have been established, but, unfortunately, these laws are not completely consistent with all measured data and do not provide the desired accuracy. Therefore, many additional test methods, excellent summarised in have been developed to unravel the complex microscopic mechanisms responsible for RPV steel embrittlement. The possible contribution of non-destructive techniques as Mössbauer spectroscopy (MS), positron annihilation spectroscopy (PAS) and transmission electron microscopy (TEM) was analysed in [4]. For SANS applications papers [3] are very valuable. The influence of different chemical elements on irradiation included hardening embrittlement was analysed in and the Atom probe tomography at RPV steels was reported in [4]. In this paper we focused mostly on comparison of Charpy-V results and PAS studies performed on irradiated WWER RPV steels.

2. Experimental results

In the framework of the "Extended Surveillance Specimen Program", started in 1995 at the nuclear power plant (NPP) Bohunice (Slovakia), several specimens, which were prepared originally for Mössbauer spectroscopy measurements, but because of the proper size (10x10x0,05 mm) and the polished surface also suitable for positron annihilation studies using the pulsed low energy positron system (PLEPS) measurement, were selected and measured before their placement into the special irradiation chambers, near the core of the

operated nuclear reactor, and after 1, 2 and 3 years resistance at this place (neutron fluence in the range from 7,8 10^{23} m⁻² up to 2,5 10^{24} m⁻²).

PLEPS technique at University of Bundeswehr (Neubiberg, Germany) [5] was used for the investigation of neutron-irradiated RPV-steels. This system enables the study of the micro structural changes in the region from 20 to 550 nm (depth profiling) with small and very thin (<50 μ m) specimens, therefore reducing the disturbing ⁶⁰Co radiation contribution to the lifetime spectra to a minimum. Such a disturbance is the limiting factor for the investigation of highly-irradiated RPV specimens with conventional positron lifetime systems. In comparison to a triple coincidence setup of positron-lifetime spectroscopy, reported in [6], PLEPS reduces the time for the measurements by about a factor 500, resulting in qualitatively comparable spectra and enables in addition the estimation of the defect concentrations. A deep analysis of these results in two lifetime components as well as the theory to the calculation of trapping rate and defects concentration was published in [7]. Actually, we would like to compare this output to Charpy V tests performed on the same type of steels.

According to the results from our PLEPS measurements performed on different irradiated RPV-steels and theory based on the diffusion trapping model, the total trapping rate κ in ns⁻¹ as well as the total defect concentration c_d (the same values but in ppm) can be calculated from the shape of the PLEPS. There was observed that the defects concentration increases slightly for both base and weld materials as a function of the irradiation dose (see Fig. 1).

The weld material (Sv10KhMFT) seems to be less sensitive to the changes caused by neutron-irradiation than the base material (15Kh2MFA). The reason could be more Cr and V contents in the weld. Nevertheless, the differences in the positron trapping rate κ are not too large. It seems reasonable to relate the observed trapping rates with the ones which have been derived for trapping into precipitated carbides from electron microscopic images [8]. Accordingly, in the type of steel 15Kh2MFA the trapping rate into chromium carbides (Cr₇C₃, Cr₂₃C₆) is predicted as κ_{Cr} =1,8x10⁸ s⁻¹ and into vanadium carbides as κ_{VC} =2,2x10¹⁰ s⁻¹. Thus precipitated vanadium carbide could indeed account for the observed trapping rates. Based on results reported in [9], the Charpy shift increases with the level of irradiation dose (Fig.2). Also in this case values for welds are slightly lower than at base metal. It is necessary to note that the irradiation damage over 1 dpa is behind expected damage during expected reactor operating lifetime.



An excellent correlation between Charpy test and positron annihilation results can be observed in case of post-irradiation annealing experiments, comparing Fig. 3 to Fig. 4.

Fig.1: The irradiation induced defects concentration c_d at WWER-440 RPV-steel specimens from base (ZM) and weld (ZK) metals after 1, 2 and 3 years residence in reactor NPP Bohunice (Slovakia) at temperature 280°C.



Fig.2: Charpy shift versus neutron dose for WWER-440 steels



Fig. 3: Charpy shift of irradiated specimens in dependence to irradiation temperature.



Fig. 4: The 3D presentation of positron lifetime results of irradiated (neutron fluence 1.25x10²⁴ m-2) and annealed WWER-440 weld metal.

Intensities of this "defects component" tau1 changed during isochronal annealing minimal (\pm 2%). Increase of the positron lifetime values after temperature of about 475 °C could be caused by the carbides precipitation or recombination (mostly VC, Cr₇C₃, Cr₂₃C₆ are created after irradiation at about 270 °C) and this increase was observed at all types of WWER steels also in the past [10]. This precipitation should not be significant in case of Western type of steel, where zero Vanadium and less chromium content is present (up to 0,7 wt.% in contrast to WWER base metal – up to 3 wt.%). Our deep positron annihilation study and the annealing of defects from 1 year irradiated WWER-440 weld specimen (ZK1Y) in form of positron lifetime study is presented in Fig.4.

Neglecting the surface region of about 200 nm, based on the PLEPS results, the optimal temperature for annealing of this type of steel is between 425 and 475 °C. The annealing temperature for both units at NPP Bohunice (Slovakia, annealing performed in 1993), as well as NPP Loviisa (Finland, annealing performed in 1996) was stated basically on Charpy shift experiences at 475 °C.

3. Conclusion

Positron Annihilation Spectroscopy 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 NPP. Actual EURATOM framework projects (LONGLIFE, NULIFE) are focused on studies towards ensuring of longer NPP operation including PAS analyses.

The second generation of WWER RPV steels of WWER-440 (V-213) is comparable with RPV-steels used in Western Europe and their quality enables prolongation of NPP operating lifetime over projected 40 years. The embrittlement of CrMoV steel is very low due to the low phosphorus and copper content.

Clear correlation between PAS results and Charpy shifts measurements performed on irradiated WWER commercially used RPV-steels was presented and discussed in details. Neutron irradiation at the reactor operating temperatures causes dominantly point defects, which are well detectable for positron annihilation lifetime techniques. The post-irradiation annealing experiments shown that optimal region for removing of irradiation caused defects is 425-475 °C. In contrast to RPV steels commercially used in Western Europe, where the positron lifetime permanently decrease up to 700 °C, different chemical composition (V and Cr content) is responsible for this effect. Based on PAS results, due to creation of additional

defects in RPV steels over 475 °C, we recommend not overrunning this temperature by annealing.

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. Therefore, we would like to use our results collected during last 20 years from measurement of different RPV-steels in "as received", irradiated and post-irradiation annealed and compare them to results where the real neutron irradiation will be replaced by proton implantation.

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