PALS combined with Charpy-V tests at WWER reactor pressure vessel steels

V Slugeň[1], A Kryukov[2], M Petriska[1], J Veterníková[1], S Sojak[1], V Sabelová[1], R Hinca[1]
[1]Institute for Nuclear and Physical Engineering, Slovak University of Technology, Ilkovičova 3, 81219 Bratislava, Slovakia
[2]European Commission, Joint Research Centre - Institute for Energy, PO Box 2, 1755 ZG Petten, The Netherlands

Abstract. This paper presents results from our long-term studies of irradiated, commercially used WWER reactor pressure vessel steels. Results from Charpy-V tests and positron annihilation spectroscopy techniques are compared and discussed in details, having in mind actual state of art and other microstructural studies in this area. The optimal region for annealing of irradiation induced defects was analyzed. It was shown that WWER steel with low impurity contents has good radiation stability and operation these reactor pressure vessels could be extended beyond a design lifetime.

1. Introduction

The basic reason of RPV mechanical properties degradation is the neutron irradiation, resulting in hardening and embrittlement of the steel which the RPV is made of. The prediction of radiation embrittlement is performed usually in accordance with relevant codes and standards that are based on large amount of information from surveillance and test irradiation programmes. Considerable data exists regarding the effect of neutron irradiation on pressure vessel steels; from both mechanical properties and microstructure features, [1].

In the last years, several reliable and predictive modelling approaches have been developed (thanks also to EC-framework projects PERFECT, REWE and PERFORM60) [2], according to which the assumed RPV-steel damage is generated by three major contributions and their synergisms:

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

The neutron embrittlement of RPV steel was a serious problem in first generation of WWER reactors: WWER-440/230. The major part of the WWER-440/230 pressure vessels were made of steel with high phosphorous and copper content in the welds. The radiation embrittlement caused by the influence of these impurities shorted the safe operational lifetime below the design value.

Undoubtedly, changes in the structure could be basically registered using several methods. Material properties of the RPV steels and influence of thermal and neutron treatment on these properties are routinely investigated by macroscopic methods such as Charpy V-notch and tensile tests. 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 [3] 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 [4-6]. In this paper we focused mostly on comparison of Charpy-V results and PAS studies performed on irradiated WWER RPV steels.

2. Expermental

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 conventional lifetime set-up as well as 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 there (neutron fluence in the range from 7,8 10^{23} m^{-2} up to 2,5 10^{24} m^{-2}). The chemical composition and the irradiation conditions of the studied RPV-steel specimens are shown in Table 1 and the irradiation conditions in Table 2.

Table 1 – The chemical composition of the studied RPV-steel specimens.

Code	**Type of steel**	**Contents of alloying elements in RPV specimens (wt.%)**											
		C	Si	Mn	Mo	Ni	Cr	Cu	P	S	V	Co	Total
ZM - Base metal WWER-440	15Kh2MFA	0.14	0.31	0.37	0.58	0.20	2.64	0.08	0.011	0.017	0.27	0.019	4.651
ZK - Weld metal WWER-440	Sv10KhMFT	0.048	0.37	1.11	0.39	0.12	1.00	0.104	0.01	0.013	0.13	0.020	3.347

Table 2 – Irradiation conditions of specimens at the 3rd unit of nuclear power plant Bohunice (Slovakia).

	Material	Code of sample	Time of irrad. [eff. days]	Neutron Fluency [m^{-2}]	Damage [dpa]	Thickness of sample [μm]
1	Base material – non-irradiated	ZM	0	0	0	60
2	Base material – 1 year irradiated	ZM1Y	280,8	7,81E23	0,08	50
3	Base material – 2 year irradiated	ZM2Y	578,5	1,64E24	0,17	40
4	Base material – 3 year irradiated	ZM3Y	894,3	2,54E24	0,26	30
5	Weld – non-irradiated	ZK	0	0	0	55
6	Weld – 1 year irradiated	ZK1Y	280,8	7,81E23	0,08	45
7	Weld – 2 years irradiated	ZK2Y	578,5	1,64E24	0,17	25

8	Weld – 3 years irradiated	ZK3Y	894,3	2,54E24	0,26	47

PLEPS technique at University of Bundeswehr (Neubiberg, Germany) [7] was used in 2000-2004 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 μm) specimens, therefore reducing the disturbing ^{60}Co 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, used in our laboratory in Bratislava, 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. Results were reported in [8].

Actually, we would like to compare this output to Charpy V tests performed on the same type of steels. Our current results show that the Charpy shift increases with the level of irradiation dose (Fig.1). 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.

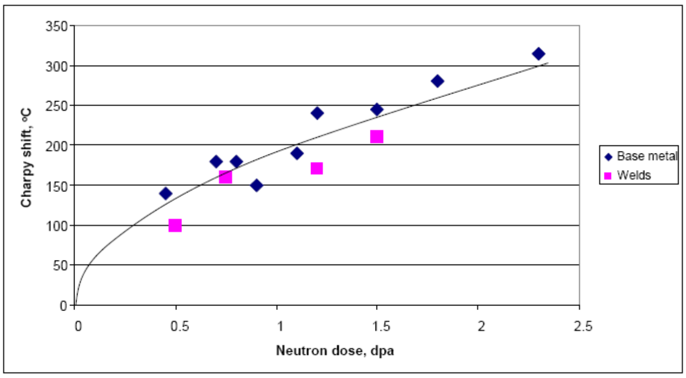

Fig.1 – Charpy shift versus neutron dose for WWER-440 steels.

An excellent correlation between Charpy test and positron annihilation results can be observed in case of post-irradiation annealing experiments, comparing Fig. 2 to Fig.3.

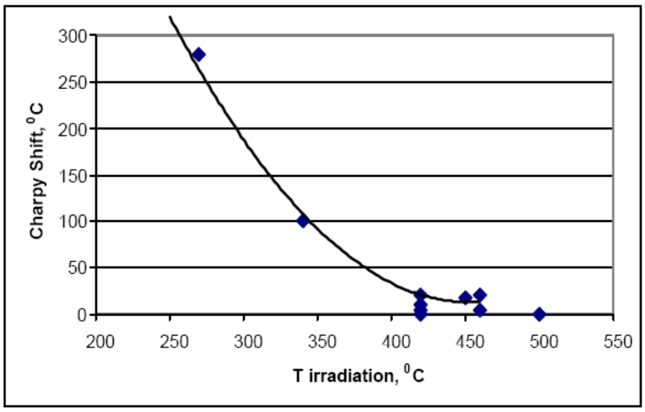

Fig.2 – Charpy shift of irradiated specimens in dependence to irradiation temperature.

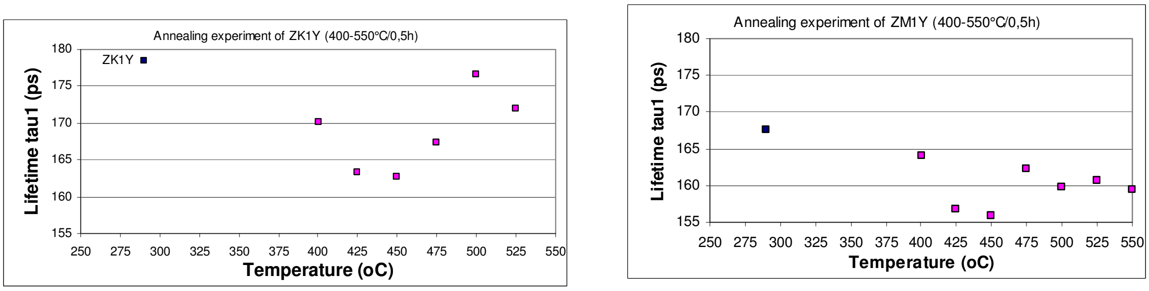

Fig. 3 - Positron lifetime in defects (Tau1) of irradiated and annealed WWER-440 base (ZM1Y) and weld metals (ZK1Y) to level of neutron fluence 1.25x10^{24} m^{-2} (~0,13 dpa). The error bars were less than 3ps.

Positron annihilation lifetime results are presented in Fig.3. Intensities of this "defects component" assigned as tau1 changed during isochronal annealing minimal (± 2%). Increase of the positron lifetime values after temperature of about 475 °C could be caused by the carbides precipitation or recombination (mostly VC, Cr_7C_3, $Cr_{23}C_6$ are created after irradiation at about 270 °C) and this increase was observed at all types of WWER steels also in the past. 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.%).

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-irradiationn annealing experiments shown that the optimal region for removing of irradiation caused defects is 425-475 ℃. In contrast to RPV steels commercially used in Western Europe, where the positron lifetime permanently decrease up to 700 ºC, 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 ºC, 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. This challenge has several open questions having in mind the fact that protons have positive charge and their behaviour in metals can be different. Nevertheless, advantages connected with much comfortable handling, transport and measurement with “not active” specimens seems to be very promising and valuable for those who would like to apply additional non-destructive techniques in this study.

4. References

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