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Non-destructive Evaluation of WWER-class RPV Steels

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ABSTRACT

Non-destructive methods as Positron annihilation spectroscopy (PAS) and Mössbauer spectroscopy (MS) were applied in the evaluation of the microstructure parameters and degradation processes of nuclear reactor pressure vessel (RPV) steel surveillance specimens. Study was oriented to the material investigation of Russian WWER-1000 steels (15Kh2MNFAA and 12Kh2N2MAA) with higher Ni content (1,26 wt.% in base metal and 1,7 wt.% in weld). For comparison, the WWER-440 weld metal (Sv10KhMFT) without Ni was measured, too. Specimens were studied in as received stage, after irradiation in LVR-15 experimental reactor to the neutron fluence $F_{(E>0.5\ MeV)} = 4x10^{23}\ m^{-2}s^{-1}$ and after annealing in vacuum at 475 ºC/2h. Changes due to different chemical composition and due to irradiation were registered using MS. Post-irradiation thermal treatment and annealing of defects was well detected by different PAS techniques. It was observed that the sensitivity of PAS and MS parameters to defined irradiation treatment decreases with Ni-content increase. Results confirm the hypothesis that Ni affects size (decrease) and distribution (more homogeneous) of the Cu- and P-rich clusters and M_xC_x carbides.

1 INTRODUCTION

As a result of radiation damage of the reactor pressure vessel (RPV), the threshold of the vessel steel DBTT shifts to high temperatures and can approach operating temperatures of the reactor. This phenomenon, the so-called radiation embrittlement, is the most important, with regard to practice, consequence of neutron irradiation on vessel steels behavior [1].

The neutron embrittlement of RPV steels is also a pronounced problem in Russian types of nuclear reactors of VVER class. It seems to be generally accepted that even in the Western types of RPV steels containing more than 0.1 wt. % of Cu, the Cu- and P-rich precipitates play a dominant role in thermal and neutron embrittlement. In case of VVER-type RPV steels, several studies [2,3] have suggested that carbide formation is an important additional microstructural mechanism. The contribution of Ni content to irradiation hardening and embrittlement was not fully realised until recently. The Ni effect was noticed in the case of RPV steel plates, forging, as well as weldments. It was shown that this effect is important for steels having Ni contents greater than 0.4 wt%. Materials with high Cu and Ni contents will have high irradiation sensitivity while those with low Cu and Ni content will be practically insensitive to irradiation [9]. Most of the published data show that Ni effect is very dependent on Cu content, and Ni content does not appear to play any role when the Cu content is low (<0.1 wt%). This suggested the existence of a synergetic mechanism resulting from interactions between Cu and Ni [4].

2 EXPERIMENTS AND RESULTS

The study was focused on the commercial RPV steels used in VVER concepts. According to experimental possibilities and practical experiences, the positron annihilation spectroscopy (PAS), Mössbauer spectroscopy (MS) were applied in investigation of RPV steels at the Department of Nuclear Physics and Technology, Slovak University of Technology, Bratislava. These methods are based on non-destructive indirect investigation of materials. Above mentioned techniques were applied in the frame of RPV steels study with the aim to achieve comprehensive information about the irradiation embrittlement phenomena.

For the experimental PAS and MS measurement the original VVER base and weld metals were selected in frame of this study, in particular Sv-10KhMFT (VVER-440/213, weld metal), 15Kh2MFA (VVER-440/213, base metal),15Kh2NMFAA (VVER-1000, base metal), Sv-12Kh2N2MAA (VVER-1000, weld metal). The chemical composition of specimens is given in Table 1. The specimens of VVER-1000 RPV steels were irradiated in the light water experimental reactor LVR-15 of Nuclear Research Institute (NRI) Řež, Prague to the specified level. On other hand, the specimens of VVER-440 RPV steels were irradiated in frame of the surveillance program of NPP Bohunice. All samples were cut and surface was precisely polished for experimental needs.

Table 1 **- Chemical composition of selected materials (in wt.%)**

RPV steel	Steel code	C	Si	Mn	S	P	Cr	Ni	Mo	V	Cu
VVER-440 weld metal	[a]Sv-10KhMFT	0.037	0.59	1.I	0.017	0.012	I.37	-	0.50	0.20	0.06
VVER-440 base metal	[a]15Kh2MFA	0.14	0.31	0.37	0.017	0.014	II.64	0.2	0.58	0.27	0.090
VVER-1000 base metal	[b]15Kh2NMFAA	0.17	0.30	0.46	0.01	0.008	2.II	I.26	0.50	0.1	< 0.08
VVER-1000 weld metal	[c]Sv-12Kh2N2MAA	0.11	0.14	0.73	0.008	0.006	1.IX	1.VII	0.55	0.01	< 0.08

(a) F(E>0.5 MeV) = 1.64x10(24) n/m2 , at Tirr = 290 °C,

(b) F(E>0.5 MeV) = 1.47x10(24) n/m2 , at Tirr = 290 °C,

(c) F(E>0.5 MeV) = 1.25x10(24) n/m2 , at Tirr = 290 °C.

3 PAS AND MS RESULTS

The PAS lifetime (LT) measurements were reported in details on the last Annual meeting of KTG in Düsseldorf. In this paper we focus our attention on results from Mössbauer spectroscopy (MS). PAS results from two component analyses are shown in Fig.1 only for comparison to MS results.

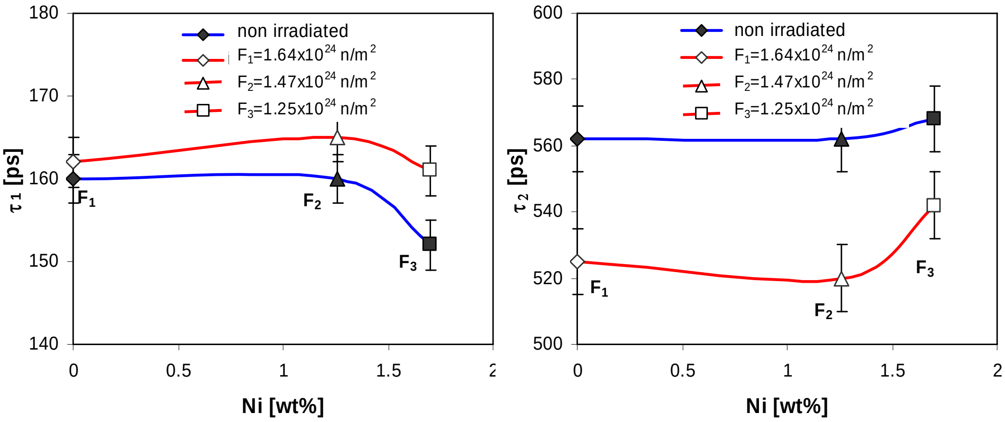

Figure 1 – Two components lifetime analysis of irradiated and non-irradiated state versus of nickel content.

Irradiation causes decrease of τ2 value (large defects cluster, voids) in all studied materials. The τ1 values (assigned to mono, di-vacancies and /or dislocation lines) stayed almost unchanged for Sv-10KhMFT (VVER 440 weld metal without nickel). In the case of Sv-12Kh2N2MAA steel (significantly) and 15Kh2NMFAA sample (partially), the intensity of this dominant component decreased. Highest changes were observed for τ1 parameter and its intensity on Sv-12Kh2N2MAA sample (VVER 1000 weld metal with 1.7% nickel).

MS results are summarised in Figure 2. All RPV steel samples investigated show the Mössbauer spectra typical for this type of steel with low alloy-element concentrations, the main features being the presence of 4 magnetically split sub-spectra with isomer and quadrupole shifts close to 0 mm/s and, in certain cases a weak superimposed doublet component (its contribution is on the level of 1% only; therefore we have neglected its existence). For the first sextet, hyperfine field Hhf1 was fixed at the level of 33.0 T (Fe-atoms in pure iron matrix not surrounded by foreign atoms in their close-neighbour shells). The second and third sextets with hyperfine fields Hhf of about 33.6 T and 30.6 T, respectively, can be assigned to the influence of alloying elements which are in first neighbourhood to Fe-atoms. Fourth sextet (Hhf of about 30.1 T and 28.5 T) are associated with iron atoms surrounded in their second or next neighbour shells by alloying elements. These hyperfine fields stayed almost stable for all irradiated or annealed specimens. Therefore, we focused our attention to the changes in Arel parameter (relative area in % describing the contribution of this particular spectrum to whole MS spectrum). Results relevant to the comparison between irradiated and non-irradiated RPV-steel specimens are reported in details in [5].

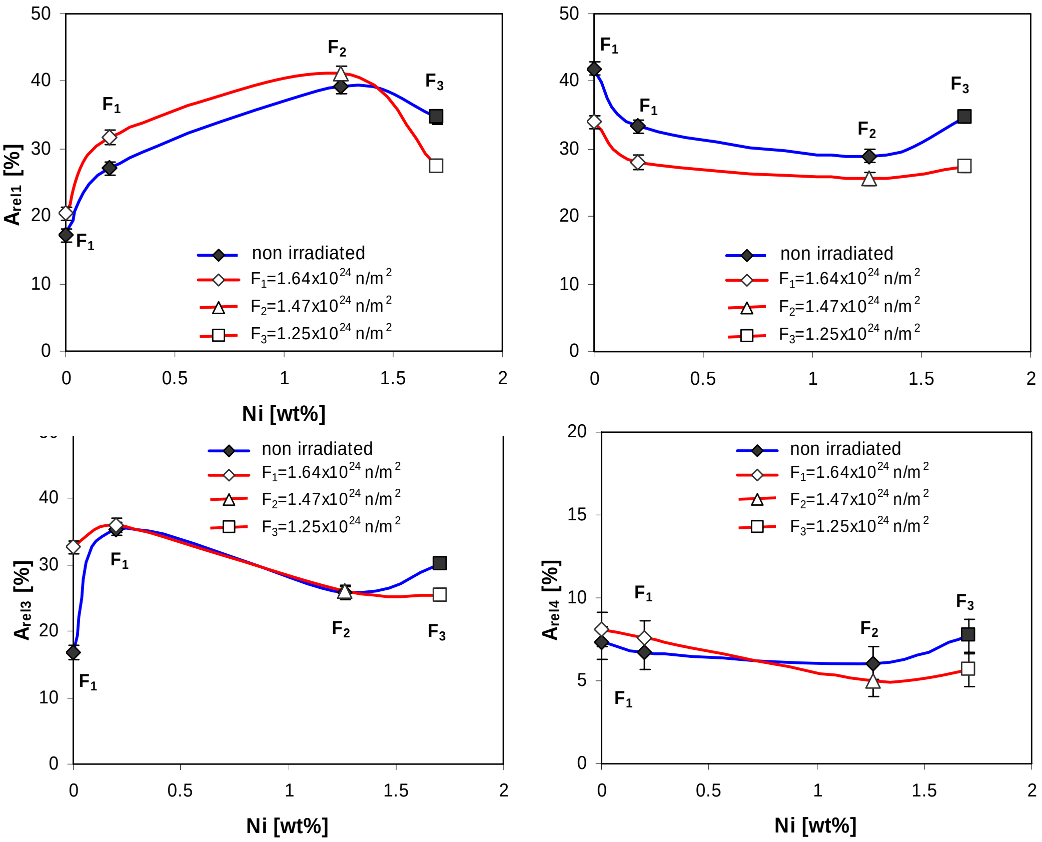

Figure 2 – Correlation of MS results (Arel parameters) versus nickel content for irradiated and non-irradiated state.

4 CORRELATION WITH TEM INVESTIGATION

The very essential information about the defect clusters could be provided by transmission electron microscopy (TEM). Number density, size and structure of features with diameter above the visibility limit of 2 nm are detectable in TEM. The secondary hardening and precipitation kinetics can also be observed and analysed using this technique. Therefore, the application of TEM is in many respects very useful in steel investigations. Example of VVER RPV steels structure is shown by Figure 3.

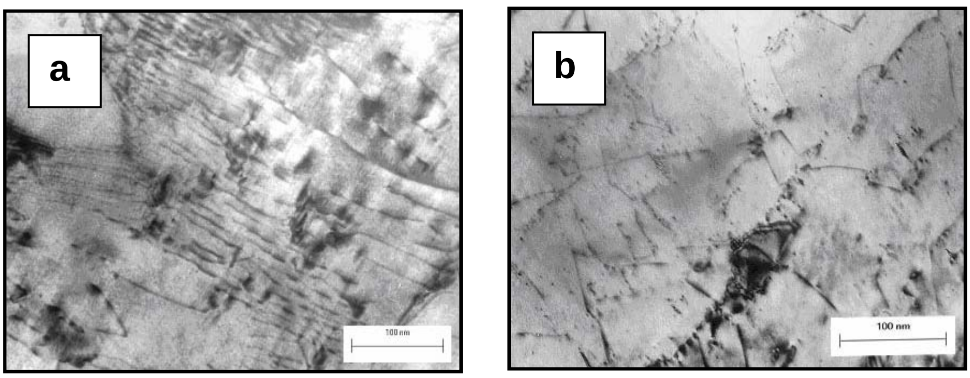

Figure 3 – (a) Structure of dislocation network and (b) Radiation-induced defects of 15Kh2MFA [5].

Two main groups of carbide phase were identified in 15Kh2MFA microstructure. M3C and M7C3 type carbide, segregation at grain boundaries and sub grains are finer than in 15Kh2MFA, but its quantity is lower. MC carbide distribution is identical with the distribution in networks proeutectoid ferrite, as well as in acicular ferrite. Dislocation substructure is evidently recovered in 15Kh2MFA after irradiation, the number of two-dimensional networks has increased, networks are more entire as in non-irradiation state.

5 CONCLUSION

The contribution of Ni content to irradiation hardening and embrittlement was studied using different PAS and MS techniques on RPV-steel specimens from VVER Round robin and surveillance programs. We focused our attention on the commercially used steels with the Cu content >0.08wt% having in mind the existence of a synergetic mechanism resulting from interactions between Cu and Ni. Differences in PAS parameters of VVER steels in not-irradiated condition, considering the different Ni content in these steels, were small. MS parameters reflect better the different chemical composition of specimens than PAS.

From analysis of PAS as well as MS parameters we can say that nickel affects both the size (decreases) and distribution (more homogeneous) of the Cu-rich clusters (or precipitates). It also takes part in the composition of these clusters, as well as of the P-rich ones. Because these clusters are supposed to act as obstacles to the dislocation motion resulting in hardening and embrittlement, Ni can contribute to irradiation hardening and embrittlement through influencing and participating in these clusters. Nevertheless, there exist a lot of open questions and there is a need for further investigation.

REFERENCES

[1] B.A.Gurovich, E.A.Kuleshova, O.V. Lavrenchuk, Jour. Of Nucl. Materials 228 (1196) 330-337

[2] L.M. Davies, Int. J. Press. Vess, & Piping 76 (1999) 163

[3] M. Grosse, V. Denner, J. Böhmert, M.-H. Mathon, J. Nucl. Mater. 277 (2000) 280

[4] V.Slugen, A.Zeman, J.Lipka, L.Debarberis, NDT&E Int. 37 (2004) 651-661

[5] V. Slugen, D. Segers, P.M.A. De Bakker, E. DeGrave, V. Magula, T. Van Hoecke and B. Van Waeyenberge, J. Nucl. Mater. 274 (1999) 273