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Radiation Damage in Nuclear Structural Materials – Past, Present and Future Challenges

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Abstract. Positron annihilation spectroscopy (PAS) had been used at the Slovak University of Technology for almost 20 years for various studies of radiation effects in nuclear grade structural materials [1-4]. In the early years, the studies were often focused radiation-induced precipitation of impurities in reactor pressure vessel (RPV) steels for the second generation of nuclear fission reactors, mostly VVER type [5]. These studies were motivated by the search for an optimal heat treatment for the recovery of the material's mechanical properties. Next generation of reactor pressure vessel (RPV) steels with improved chemical composition turned the aim of the research to increased displacement damage and the experimental simulation high neutron fluencies by utilizing ion implantations [6]. Thermonuclear fusion and spallation target technologies introduced even more severe radiation conditions with high production rates of gaseous transmutation products such as hydrogen and helium. Recent research activities of the Institute of Nuclear and Physical Engineering (INPE) are focused on distinct effects of flux, temperature and oxide dispersions on the behavior of nuclear materials exposed in harsh radiation conditions [7,8].

This contribution reviews experimental PAS studies of various grades of nuclear structural steels investigated by authors during last years. The new results are compared to those from the earlier studies in order to distinguish between various processes involved in the formation of the microstructure of irradiated materials. Future challenges in the understanding and characterization of advanced structural materials for nuclear applications are also outlined.

INTRODUCTION - STATE OF THE ART

One of the most fundamental tasks of nuclear-reactor safety research is assessing the integrity of the RPV. The embrittlement of RPV steel is a very complicated process depending on many factors (thermal and radiation treatment, chemical compositions, preparing conditions, ageing, etc.). 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 data and do not provide the desired accuracy. Therefore, many additional test methods [9] have been developed to unravel the complex microscopic mechanisms responsible for RPV steel embrittlement.

RPV embrittlement poses one of limiting factors in the lifetime of vessels of today's nuclear power plant (NPP) This problem is very serious in Eastern (Russian) types of nuclear reactors (VVER). It is due to the narrower gap between the outside surface of the core barrel and the inside surface of the RPV than in Western RPV's. The neutron flux and consequently neutron fluency on the RPV wall is generally much higher for VVER-440 type reactors than for other equivalent types. This influence of neutron flux (even with neutrons of energy over 0.1 MeV) on RPV embrittlement is much more pronounced than other contributions, e.g., from coolant temperature or from the operational pressure in the primary circuit.

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Clear differences between western and eastern types of the commercially used RPV-steel were observed and discussed using PAS LT, MS and TEM [5]. According to the results from the PAS mean lifetime analyses, the observable differences in the values and behaviours of successive annealing curves are caused not only by different chemical compositions, but also by different preparing technologies of the steels. This fact is remarkable mainly in the 15Kh2MFA steel, from which the base material and simulated HAZ were studied simultaneously.

Changes in steel microstructure due to starting phase transition from b.c.c to f.c.c. were observed on all westerntypes of RPV-steels using PAS at ~ 725-750 °C. In case of Russian RPV-steels, this phase transition was not observed up to 800 °C. The same phenomenon appeared also in case of MS, when the presence of the paramagnetic austenite was clearly detected at 750 °C in Western batches of steels and at 800-850 °C in Russians steels [5].

Interactions between the fast neutron and the lattice atom can transfer energies ranging from a few eV to tens of keV. The primary knock-on atoms (PKA) lose energy though interacting with both the bound electrons and atoms of the solid. If the energy transferred to a lattice atom is greater that some threshold value Ed (typically >40eV), then the atom will be displaced from its lattice site, creating a Frenkel defect, i.e. a vacancy and an interstitial. If the PKA energy is much greater than a few keV the PKA is able to displace many atoms, rapidly entering a regime where the collisions occur every lattice spacing (displacement cascade). It is important to understand, that in the evolution of a cascade not only a heavily damaged region containing a large number of displacement atoms is established, but that during the subsequent evolution considerable point defect motion, recombination and clustering may occur. Both vacancy and interstitial point defects are expected to be mobile in the temperature range of most operating pressure vessels. However, they are also expected to interact with solute atoms.

From the extensive positron studies performed on 15Kh2MFA steel [10, 12,13,14] and from metallurgical and transmission electron microsc1opy (TEM) studies [11] supported also by the Mössbauer spectrometry results [5,14] it can be accepted that irradiation-induced carbides play an important role by strengthening and radiation embrittlement. TEM results of irradiated 15Kh2MFA steels confirmed that the radiation damage structure is heterogeneous and contains dislocation loops and very fine VC or V4C3 precipitates. Dislocation loops predominate in bainite structures while VC is present mostly in the ferritic grains of the weld metal. From several Small Angle Neutron Scattering (SANS) measurements performed on 15Kh2MFA steels [15], the reason of the irradiation hardening seems to be the fine-disperse defects within the range 1-4 nm. Further, in steels containing residual levels of elements such a copper, which are in super-saturated solution, radiation-enhanced diffusion will occur at these temperatures, which leads to the formation of small clusters which can again harden the matrix [11].

Generally, the thermal treatment together with neutron irradiation lead to microstructure consisting of small clusters (<5nm in diameter) which create obstacles to the free movement of dislocations thereby producing an increase in the yield stress, hardness and the ductile-brittle transition temperature of the material.

In order to decide whether or not a precipitate can trap positrons, the positron affinity A+ for the host material as well and the precipitate were calculated [16]. It was shown that the A+ values are relatively high and the positron lifetimes are very short for the perfect MC carbides. This fact confirms that the perfect MC (M□Cr, V, Ti, Mn, Fe, Zr, Nb) carbides are very dense materials and when embedded in an Fe matrix cannot trap positrons. In general, the radiation damage from the PAS point of view is interpreted preliminarily as a combination of radiation induced point defects, dislocations and small vacancy clusters [17,18] mainly in the region of precipitate-matrix interface.

According to our measurements performed at different lifetime techniques (Gent, Bratislava, Munich) and many trials to fit the spectra in several components, the structure of RPV steels is so complex that only one or two "steelcomponent analysis" is useable for serious considerations. It is important to be very precise during sample preparation and by excluding contributions from oxides, source, back-diffusion, etc.

A long positron lifetime component τ_2=260 ps, reported in [19] from an extensive study of Fe-1,25%C alloy and attributed to the positron trapping in cementite (Fe_3C) and at the cementite-ferrite interface, was not observed in our spectra (probably due to 10 times lower concentration of C in studied RPV-steels).

PAS can identify not only differences between Western and Russian types of steel and also changes in preparing technologies in the same type of steel. It was confirmed that Russian steels are more sensitive to elevated temperature (annealing) and their heat affected zone is the most sensitive place for thermal and neutron embrittlement in the reactor. The minimal values of mean lifetime (about 500 °C) in the case of Russian steels confirm that the temperature, used for annealing of the 1st and 2nd units of NPP Bohunice (Slovakia) and Loviisa (Finland), 475 °C for 144 hours, was chosen correctly. A rapid increase in the vacancy-type defect formation in the temperature region 525-600°C was observed in Russian types of steels. The positron lifetime in Western steels decreases slowly up to about 750°C where phase transition from b.c.c. to f.c.c. structure occurs [5]. Many PAS works were focused on registration of optimal annealing procedures. One of the most appreciated author´s contributions in the “positron community” from the industrial point of view was PAS confirmation of optimal annealing temperature of 15Kh2MFA RPV steel. PAS results show that annealing over 500 °C causes creation of the next large amount of small defects connected very probably to carbide precipitation [5].

CURRENT AND FUTURE CHALLENGES

Based on obtained experiences and PAS techniques potential in microstructural evaluation of reactor steels following challenges could be identified for applications of PAS techniques: i) in new steels development, ii) in evaluation of defects mobility due to different treatments, iii) in improvements of FWHM and more precise characterisation of defects, iv) in clever combination with other techniques. We focused our activities on application of PAS techniques on ions (mostly H^{++}, He^{+}, Fe^{+}) implantation as experimental simulation of radiation damage and followed studies of such created defects. Although theoretical studies were done in the past, experimental verification of predicted behaviour of defects due to ageing or thermal treatment is still ongoing. In our studies we focused on hydrogen ions implantations as proton (and from the mass point of view) also neutron treatment and Helium ions implantations for alpha treatment experimental simulations. Based on proton implantation and results comparison with neutron irradiation [20] we could conclude: i) The average positron lifetime was a bit higher in the implanted specimens than in the original irradiated one with identical amount of impacted particles (~2.0×10^{19} cm^{-2}). That might indicate that hydrogen (protons) implantation causes slightly more damage and we must take it into account in further experiments. Essentially, the effect of defects agglomeration into slightly bigger clusters and decrease of their intensities shows that hydrogen implantation can be plausible used to simulate neutron irradiation damage under certain circumstances. ii) We state that no large voids or vacancy clusters were formed due to irradiation or implantation in investigated German RPV steels which could cause dangerous embrittlement of RPV and limit the operation of NPPs.

Our PAS results from last decade on different specimens after Helium implantations were published in [21-24]. In recent work [25] we focused on Fe 11.62 wt% Cr model alloy (see [26] for more details). This alloy was implanted by 250 keV He^{2+} ions to three different fluencies (3×10^{17}, 9×10^{17} and 1.5×10^{18} cm^{-2}) at T<100°C. In order to estimate displacement damage, produced by cascade collisions, a Monte Carlo simulation code - Stopping and Range of Ions in Materials (SRIM) was used. The profile of the implanted He ions together with the dpa profile can be seen in the Fig.1. Based on the suggestions of Stoller et al. [27], following variables were applied for the SRIM calculations: Displacement energy = 40 eV; Lattice binding energy = 0 eV; type of TRIM calculation – “Quick damage calculation”. Dpa and He concentrations were calculated for four different target depths and three applied fluencies. Summary of the investigated conditions is listed in the Tab.1.

TABLE 1. Irradiation conditions, as investigated by 6, 7, 8, 9 and 10 keV positrons respectively

He ion fluence	depth [nm] / dpa / He [appm]			
3.0×10^{17} cm^{-2}	100/2.1/921	130/2.3/1370	160/2.4/1864	230/3.0/3348
9.0×10^{17} cm^{-2}	100/6.4/2763	130/6.8/4111	160/7.3/5591	230/9.1/10045
1.5×10^{17} cm^{-2}	100/10.6/4604	130/11.3/6851	160/12.2/9319	190/13.3/11179

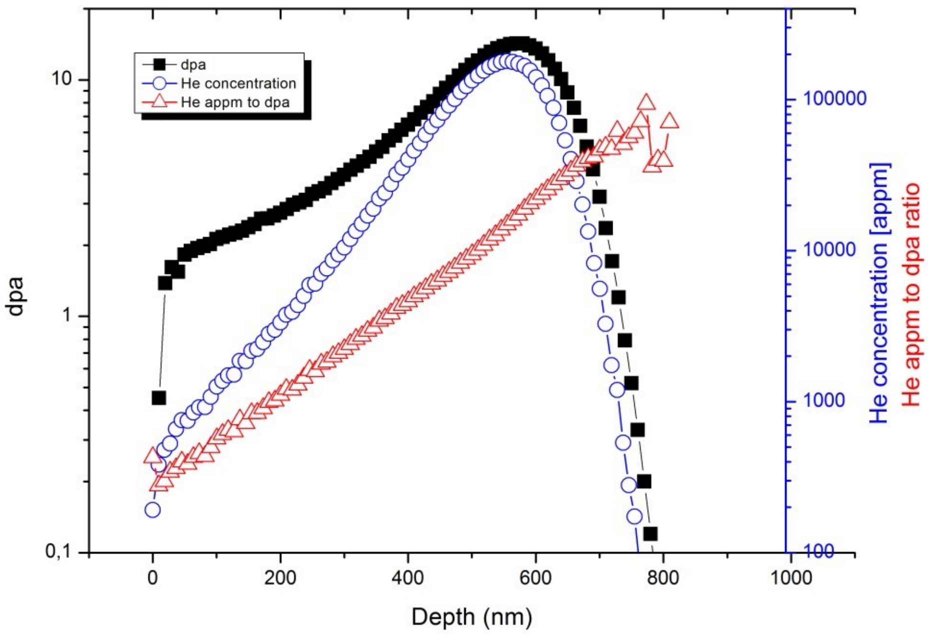

FIGURE 1. Displacement damage peak and He profile as simulated by SRIM code.

The irradiation conditions produced by implantation of helium ions provide extremely harsh environment comparing to the one in the spallation target. While the displacement damage (dose) in the studied helium-implanted samples is comparable to a two-years campaign in the spallation target, dpa rate and helium production rate are increased by orders of magnitude in the He implantation experiments. The concentration of helium is comparable for low-dpa samples, but in the high-dpa implantation sample, it goes well above the spallation conditions.

The evolution of τ_{av} with He appm indicate the role of accumulated helium on the formation of complex vacancy-type defect clusters. The Fig.2 also suggest that accelerated displacement damage rate of ion implantations does not produce substantionaly different defects and at fluence <10^{18} cm^{-2} the values of τ_{av} are very similar to irradiation in spallation environment.

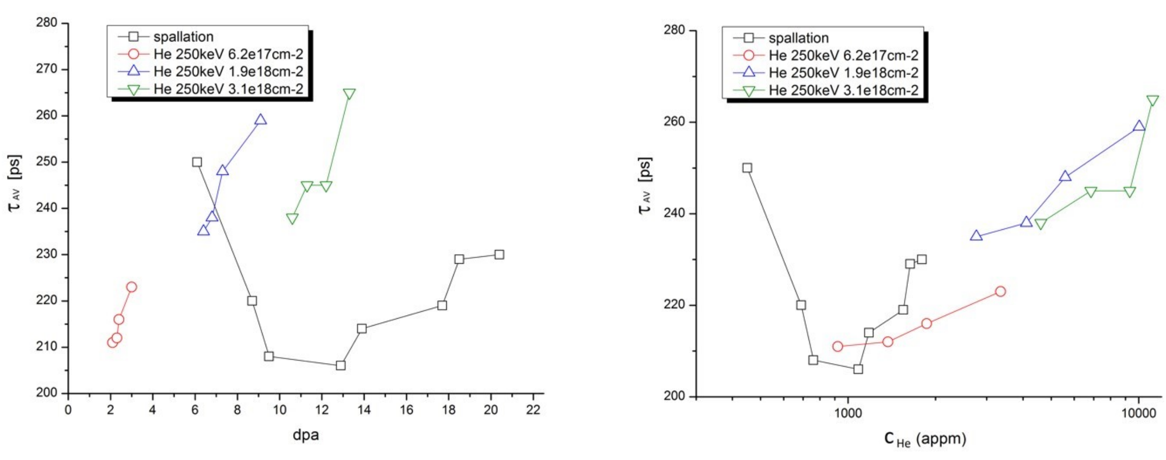

FIGURE 2. Average positron lifetime as a function of displacement damage (left) and as a function of helium concentration (right).

Decomposition of the acquired lifetime spectra into reduced bulk component τ_1 and two defect components τ_2 and τ_3 enable a good insight into the evolution of helium bubbles. The bimodal defect distribution is a realistic approximation of the vacancy-type defects and can be clearly seen in the distinct lifetime τ_2 and τ_3. The first defect component τ_2 can be attributed to small vacancy clusters, which are expected to be filled by helium to the equilibrium He-to-vacancy level or to be slightly over pressurized [28]. The second defect component τ_3 characterizes large stable vacancy agglomerates such as bubbles or voids.

While the evolution of the small defect component τ_2 is rather continuous across both experiments, large defect component τ_3 indicates different mechanism involved in the growing of the relevant defects. It is important to stress the fact, that positron lifetime is not only a function of size of a cluster, but also a function of the helium content. Lifetimes in bubbles are generally shorter than lifetimes in empty voids and the "reduction" reflect the gas density inside the bubble. Moreover, in big voids (> 50 vacancies / 1nm), positron lifetime has a value of 450-500 ps, independent on the void size [29]. Such defects can be expected in all samples with 700 or more appm of He [30].
Shorter lifetime τ_3 in the implanted samples therefore does reflect neither smaller bubbles nor does it reflect lower density of helium.

Very promising for the next PAS studies could be also combined use of computer simulations and proper PAS experiments with the aim to describe creation and mobility of vacancy type defects. Our activities based on ab initio approach or molecular dynamic approach in this area were reported in [31, 32]. In the future, the use of PAS techniques can be foreseen by the development of new types of steels with well-defined parameters (materials for fusion reactors, etc.) or by evaluation of effectiveness of post-radiation heat treatment. Application of a high precise positron microscope with higher resolution in the new steels development would be surely one of the progressive ways [33].

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