APPLICATION OF POSITRON ANNIHILATION SPECTROSCOPY FOR INVESTIGATION OF REACTOR STEELS

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1. Introduction

Radiation, heat, and mechanical resistance are crucial parameters of structural materials of Nuclear Power Plants (NPP) which limit their planned lifetime. Much higher radiation damage is expected in new generations of nuclear power plants, such as Generation IV and fusion reactors. Therefore, the investigation of new structural materials is, among others, focused on the study of reduced activation ferritic/martensitic (RAFM) steels with good characteristics as lower activation, good resistance to volume swelling, good radiation, and heat resistance (up to 550 °C).

Our work is focused on the study of radiation damage simulated by ion implantations and thermal treatment evaluation of RAFM steels in the form of binary Fe-Cr model alloys. In order to study the microstructure recovery after ion irradiation, we applied an approach for restoration of initial physical and mechanical characteristics of structural materials in the form of thermal annealing, with the goal to decrease the size and amount of accumulated defects. The experimental analysis of material damage at microstructural level was performed by the pulsed low energy positron system (PLEPS) [1] at the high intensity positron source NEPOMUC [2] at the Munich research reactor FRM-II.

2. Materials treatment

Detailed chemical composition of studied Fe-Cr alloy can be seen in the table 1. Fabrication processes and treatments of the alloy can be found in [3]. “As-received” material was cut into desired dimensions, ground and polished to mirror-like surface before exposure to helium implantation.

Tab.1: *Chemical composition of studied Fe-Cr alloy (wt%) [3].*

Alloy	Cr^*	O^*	N^*	C^*	Mn	P	Ni	Cu	V
Fe-11.62%Cr	11.62	0.031	0.024	0.028	0.03	0.05	0.09	0.01	0.002

* *measured after heat treatment during fabrication*

Accelerated helium ions were used to obtain cascade collisions in the microstructure of studied material without neutron activation. Helium implantation was performed in two steps with ions energy of 250keV and 100keV, respectively. Implantations at the linear accelerator of the Slovak University of Technology in Bratislava [4] were performed at dose of 0.3 C/cm^2 (1.87x10^{18} cm^{-2}) corresponding to ~ 55 dpa. The maximum temperature during implantation did not exceed 100 °C.

After PLEPS measurement at different ion implantation doses specimens were thermally treated with the aim to understand the influence of annealing on structure formation and defect behaviour. Thermal treatment was performed at „Universitaet der Bundeswehr“ in Neubiberg (Munich, Germany). Specimens were annealed in argon atmosphere (10 kPa) at temperatures of 400, 475, 525 and 600 ºC for 2 hours, then gradually cooled down (2 °C/min) and repeatedly measured after each temperature by PLEPS technique.

3. Results

Depth profiling of vacancy type defects was performed by PLEPS using positron energies between 2 and 18 keV corresponding to the mean penetration depths 15 – 525 nm. The evaluation of Fe-11.62%Cr measured spectra was performed by PosWin code [5] and the spectra were decomposed into the three components assigned as, τ_1- positron annihilation in bulk, τ_2- positron annihilation in defects (vacancies, vacancy clusters) and τ_3- positron annihilation in large defects (voids).

Mean positron lifetimes (MLT) were calculated from the three partial components for all annealing temperatures. Specimens implanted at a level of 0.3 C/cm^2 showed significant decrease of mean positron lifetime at temperature of 600 ºC (fig. 1).

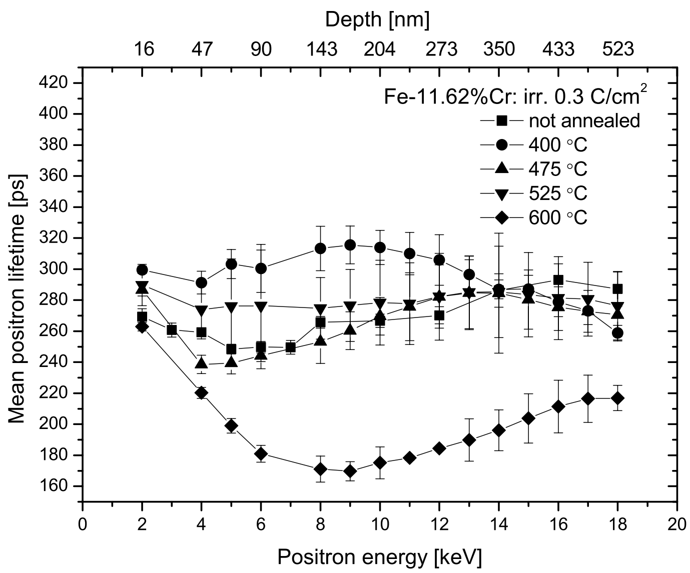

Fig.1: *Mean positron lifetimes of annealed Fe-11.62%Cr alloys.*

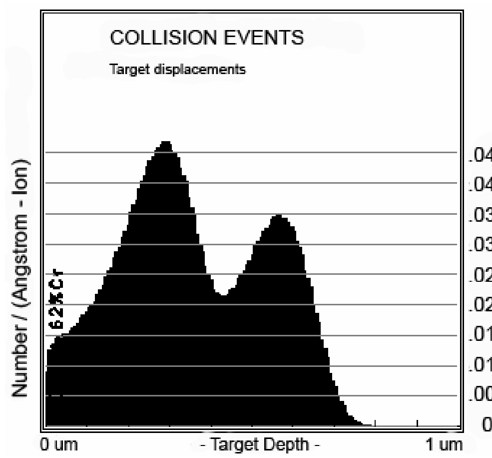

Fig.2: *SRIM simulation of damage by 100keV (left peak) and 250keV (right peak) He ions.*

Analysis based on the MLT at this temperature, at the depth of main damage caused by He ions (fig. 2), could be interpreted as extensive decrease of defect size to monovacancies or small vacancy clusters [6, 7]. Components reflecting positron lifetimes in bulk and defects showed a decrease. Small clusters and the bulk with dislocations were recognized. Therefore, taking into account the literature [8], recovery of the microstructure at this temperature can be considered. The growth of MLT from the depth of ~170 nm to the surface could be among others caused by the oxide layer, which was also found by energy dispersive X-ray analysis.

The interpretation of data, obtained by the PLEPS technique at lower annealing temperatures (400, 475, 525 ºC), was more difficult due to the behavior of the mean positron lifetimes (fig. 1), as no expected differences in trend lines and maximum damage peaks from 100 keV and 250 keV He ions were observed. Therefore, these results were supported by the scanning electron microscopy (SEM) performed at the Institute of Materials Science (Faculty of Materials Science and Technology, STU). A diagonal cut under a 12º angle was performed prior to SEM measurements (fig. 3) of an implanted Fe-11.62%Cr specimen. The figure shows major damage in two regions. Considering the results found in SRIM (Stopping and Range of Ions in Matter) simulation, these two regions could be assigned to the maximum damage caused by He ion implantation. The first peak (fig. 2) in a depth of ~300 nm is very significant and implies to damage from 100 keV and partially also from 250 keV helium ions. This can be also recognized from SEM figures, in which the area closer to the surface has more significant damage (100 keV He) than the one further from the surface influenced mainly by 250 keV He ions. Therefore, we can assume that MLTs measured at temperatures of 400, 475, 525 ºC were influenced by these two heavily damaged regions. Regeneration of the microstructure after annealing at temperature of 600 ºC, followed by significant mean positron lifetimes decrease, was also confirmed by preliminary SEM measurement of this structure, where the damaged regions mostly disappeared.

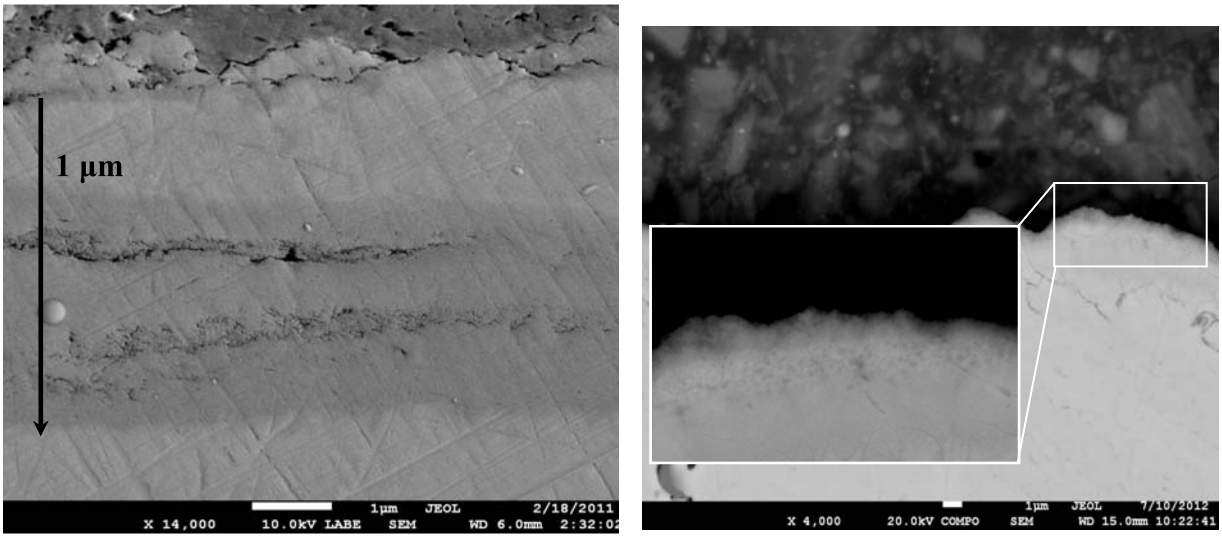

Fig. 3: *Alloy Fe-11.62%Cr implanted at 0.3 C/cm^2 level and annealed at 475 ºC (left) and 600 °C (right). Annealing at temperatures between 0.3T_m-0.4T_m (T_m is melting temperature-in case of RAFM steels it is about 1400-1500 °C depending on the chromium content) can cause void and dislocation structure formation (the dislocation loops are unstable and grow into a dislocation network) and diffusion is sufficient for the formation of precipitates. This corresponds well to results obtained here for an annealing temperature of 475 ºC, as ion damaged area was enlarged and voids reached width of tens of nanometers. Heating of the RAFM steels above 0.4T_m led to continuous annealing out of displacement damage, resulting in little change in strength (at these temperatures strength sometimes decreases because irradiation-enhanced diffusion accelerates the normal thermal aging process) [8].*

4. Conclusion

Investigation of irradiated Fe-11.62%Cr alloys after thermal annealing was performed in this work. Evaluation of the measured data for annealed Fe-11.62%Cr alloys by the PLEPS technique was made considering SEM results for a specimen annealed at 475 °C, which showed major damage in two helium affected regions. Mean lifetimes describing damage of annealed (400-525 °C) specimens were at a level of ~290 ps and in comparison to not annealed specimen, annealing effect was not observed. Only at 600 °C temperature, the mean positron lifetime showed a major decrease. Therefore, considering the literature [8] regarding the temperature influence on reduced activation ferritic/martensitic steels above $0.4T_m$, we could say that such large voids in affected area (<1 μm) were continuously annealed out or at least their size decreased to monovacancies and small vacancy clusters not exceeding size of 5 vacancies. These outcomes were supported also by the preliminary SEM results where the microstructure regeneration is noticeable.

Ion implantation damage produced in specimens was at a very high dpa level and together with thermal annealing and oxide layer on the surface, created complicated system for the final evaluation of measured data. Therefore, even if the positron annihilation spectroscopy is very useful technique for studies of vacancy type defects, we experienced some difficulties with characterization of large defects represented by MLT. Therefore, additional destructive or non-destructive techniques were applied in order to give us a complementary view on the examined materials.

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