HELIUM IMPLANTED FeCr ALLOYS STUDIED BY POSITRON ANNIHILATION LIFETIME TECHNIQUE

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The influence of chromium on the radiation damage resistance of iron based alloys has been studied using conventional positron lifetime technique and a pulsed low energy positron beam. To simulate high neutron flux, the helium implantation has been used. Different levels of helium doses ($6.24 \cdot 10^{17}$–$3.12 \cdot 10^{18}$ cm^{-2}) corresponding to a local damage of up to 90 dpa were accumulated in a thin <1 μm region. Four different binary FeCr alloys (2.6; 4.6; 8.4; 11.6 wt.% of Cr) have been used in this study. The obtained results show that chromium has a significant effect on the size and distribution of the created defects. The character of these defects has been determined as large voids (>1 nm) and small vacancy clusters together with the initial dislocations and small point defects.

1. INTRODUCTION

Structural materials used in today's nuclear power plants are considered as insufficient for the new generation of nuclear power facilities, e.g. generation IV fission reactors and fusion reactors. High temperatures and high irradiation loads on the reactor materials on the one hand, and the required low activation of these materials on the other hand, demands the substitution of the low-alloy ferritic steels and austenitic steels currently in use. The Reduced Activation Ferritic/Martensitic (RAFM) steels with high chromium content (up to 12 wt.%) seem to be the most promising materials, which meet these technical demands. These ferritic/martensitic steels have been originally developed for temperatures of about 650 °C and higher steam pressures in conventional power plants. In the 1970 s these materials have been considered for nuclear fast reactor programs and later as structural materials for fusion reactors. This research has introduced also the concept of low-activation materials [1].

In the early development the chromium was added only for corrosion resistance. However, the nuclear applications of the chromium ferritic/martensitic steels showed the importance of chromium as an alloying element for the improvement of resistance to radiation induced microstructural changes. In particular, the decrease of the void swelling due to chromium addition has been discussed in the last decade [2, 3]. Currently, this phenomenon is studied using experimental techniques as well as computer simulations.

This paper discusses our recent positron annihilation lifetime experiments focused on the influence of chromium on the microstructural changes in iron based alloys under radiation treatment. Positron annihilation lifetime spectroscopy is a suitable techniques for the observation of small atomic defects as they are created e.g. by cascade collisions [4, 5]. To simulate high neutron fluencies, helium implanted specimens have been studied by conventional positron annihilation lifetime spectroscopy (PALS) and positron annihilation lifetime spectroscopy with the slow pulsed positron beam PLEPS (Pulsed Low Energy Positron System) [6] at the high intensity positron source NEPOMUC [7]. Applications of the PLEPS techniques on reactor pressure vessel steels and advanced nuclear materials are described mostly in [8-10].

2. EXPERIMENTAL

2.1. Materials and sample preparation

To study the influence of chromium concentration on the radiation resisistance, four Fe-Cr binary alloys with different Cr content have been selected. The detailed chemical composition of the alloys can be seen in the Table 1. After casting, the obtained ingots were cold worked under protective atmosphere to fabricate plates of 9 mm in thickness. Further, the alloys were treated for 3 hours at 1050 °C in high vacuum for austenisation and stabilization. The duration of the heat treatment was chosen so as to get rid of any possible precipitation or phase transformation that might have been happening during hot rolling and also to allow a maximum degassing of the alloys. This treatment was than followed by air cooling to room temperature. The tempering procedure is of critical importance for this type of steel, and because the intention was to compare the chemical effect and not the microstructure alone, it was found that tempering at 730 °C for about 4 hours followed by air cooling, was the best to ensure a full martensitisation especially the high Cr-content model alloys. All the alloys were heat-treated to ensure well defined martensitisation. More detailed information about these materials and the fabrication processes can be found in [11].

Table 1

Chemical composition of the studied materials

Alloy ID	Cr*	O*	N*	C*	Mn	P	Si	Al	Ti	Ni	Cu	V
L251	2.36	0.035	0.012	0.008	0.009	0.013	0.002	0.003	0.004	0.044	0.005	0.001
L259	4.62	0.066	0.013	0.02	0.02	0.011	0.006	0.003	0.003	0.06	0.01	0.001
L252	8.39	0.067	0.015	0.021	0.03	0.012	7E-04	0.007	0.003	0.07	0.01	0.002
L253	11.62	0.031	0.024	0.028	0.03	0.05	0.006	0.003	0.004	0.09	0.01	0.002

*Measured after heat treatment.

The as-received materials have been cut to the desired dimensions, ground and then carefully polished to mirror-like surfaces before exposure to helium implantation. Subsequently, an non-implanted reference sample and the implanted samples were investigated with positrons.

2.2. Radiation treatment

To obtain cascade collisions in the microstructure of the studied materials without neutron activation, accelerated helium ions have been used. Helium implantation at two different energies and five different dose levels has been performed at the linear accelerator of the Slovak University of Technology in Bratislava [12]. To ensure good sensitivity of the employed positron lifetime techniques, the ion energies were set to 250 and 100 keV. To calculate the dpa (Displacement per Atom) parameter, the depth sensitivity of the experimental technique has to be considered. In the case of conventional PALS, the used positron source, ^{22}Na, has a continuous spectrum of positrons up to energy of 545 keV [13]. The corresponding exponential implantation profile [14] has a mean implantation depth of <10 μm, and more than 99.9 % of positrons annihilate within 100 μm from the surface. This value was used for calculation of the average dpa (dpa$_{\text{PALS}}$, Table 2) for all levels of helium treatment. In the case of monoenergetic positrons, the implantation profile can be approximated by the derivative of a Gaussian with an energy dependent mean implantation depth [15]. The highest available positron implantation energy of 20 keV in our experiments corresponds to a mean positron implantation depth of 0.7 μm. Thus, monoenergetic positrons with energies between 10–20 keV sample the region of expected highest radiation damage according to the calculations in Table 2 (dpa$_{\text{PLEPS}}$).

Table 2

Calculations of average DPA for different level of implantation in first 100 μm layer (DPA_{PALS}) and 800 nm (DPA_{PLEPS}) of studied Fe-Cr alloys

Dose [ions/cm^2] (C/cm^2)	$6.24 \cdot 10^{17}$ (0.1)	$1{,}25 \cdot 10^{18}$ (0.2)	$1{,}87 \cdot 10^{18}$ (0.3)	$2{,}5 \cdot 10^{18}$ (0.4)	$3{,}12 \cdot 10^{18}$ (0.5)
DPA_{PALS}	0.15	0.30	0.45	0.60	0,74
DPA_{PLEPS}	18.55	37.10	55.64	74.19	92.74

2.3. Positron annihilation lifetime spectroscopy (PALS)

Positrons are very sensitive probes for vacancy-type defects of atomic dimensions, e.g. vacancies, vacancy agglomerates, dislocations or inner surfaces. It is well established that positrons may be trapped at these defects and, because of the locally reduced electron density, the lifetime of the positron localized at the defect increases. This lifetime has characteristic values for each defect type and therefore it is possible to separate out various atomic defect configurations and their relative abundance with very high sensitivity (~1 ppm) and in a non-destructive way [16]. In conventional positron lifetime spectroscopy, radioactive β^+ isotopes with positron energy distributions up to several hundreds of keV are used. Therefore, depending on the density of the material, the positrons sample defects at depth of hundreds of micrometers below the surface. Consequently the spatial resolution is of the same order and all finer details on the (sub) micrometer scale are blurred. To gain more detailed information on the depth distributions of the defects with positron lifetime spectroscopy, pulsed monoenergetic positrons beams of variable energy are required. For our purposes, both techniques provide useful information.

The conventional positron lifetime measurements have been performed for all materials and all damage levels in the PALS laboratory at the Slovak University of Technology.

Depth profiling of defects up to one micrometer below the surface has been performed on the non-implanted Fe11.62%Cr sample and three Fe11.62%Cr samples with different damage levels using positron implantation energies between 1 and 18 keV. The depth profiles were measured with the Pulsed Low Energy Positron System (PLEPS) [6] at the high intensity positron source NEPOMUC [7] at the research reactor FRMII at TUM.

3. RESULTS AND DISCUSSION

3.1. Conventional PALS measurements

The measured positron lifetimes spectra were analyzed using the LT 9.0 program [17]. All the lifetime spectra could be decomposed into three lifetime components with variances close to one. The longest lifetime τ_3 (> 600 ps) with a very low intensity (< 1.5%) was similar in all measurements and shall not be discussed here.

The short lifetime of about 100 ps, describing positron annihilation in the undisturbed bulk material has been found in all materials at all implantation levels.

Fig. 1,a shows the second lifetime τ_2 as a function of chromium content and the helium implantation dose. This lifetime can be associated with the trapping of positrons in dislocations and small vacancy type defects. In the low chromium alloys (L251, L259) the defect lifetime increased with the implantation dose up to 235 ps. This value may be associated to small clusters of 4- 5 vacancies or slightly larger clusters containing helium. In the high chromium alloys (L252, L253) the defect lifetime increases from 180 ps in the untreated specimens to <200 ps in the damaged specimen.

The intensity I_2 of the second lifetime τ_2 is shown in Fig. 1,b and is almost independent from the implantation dose. However, it is increased for the high chromium alloys. This points to a higher density of uniformly distributed defects, which are smaller than in the low chromium alloys.

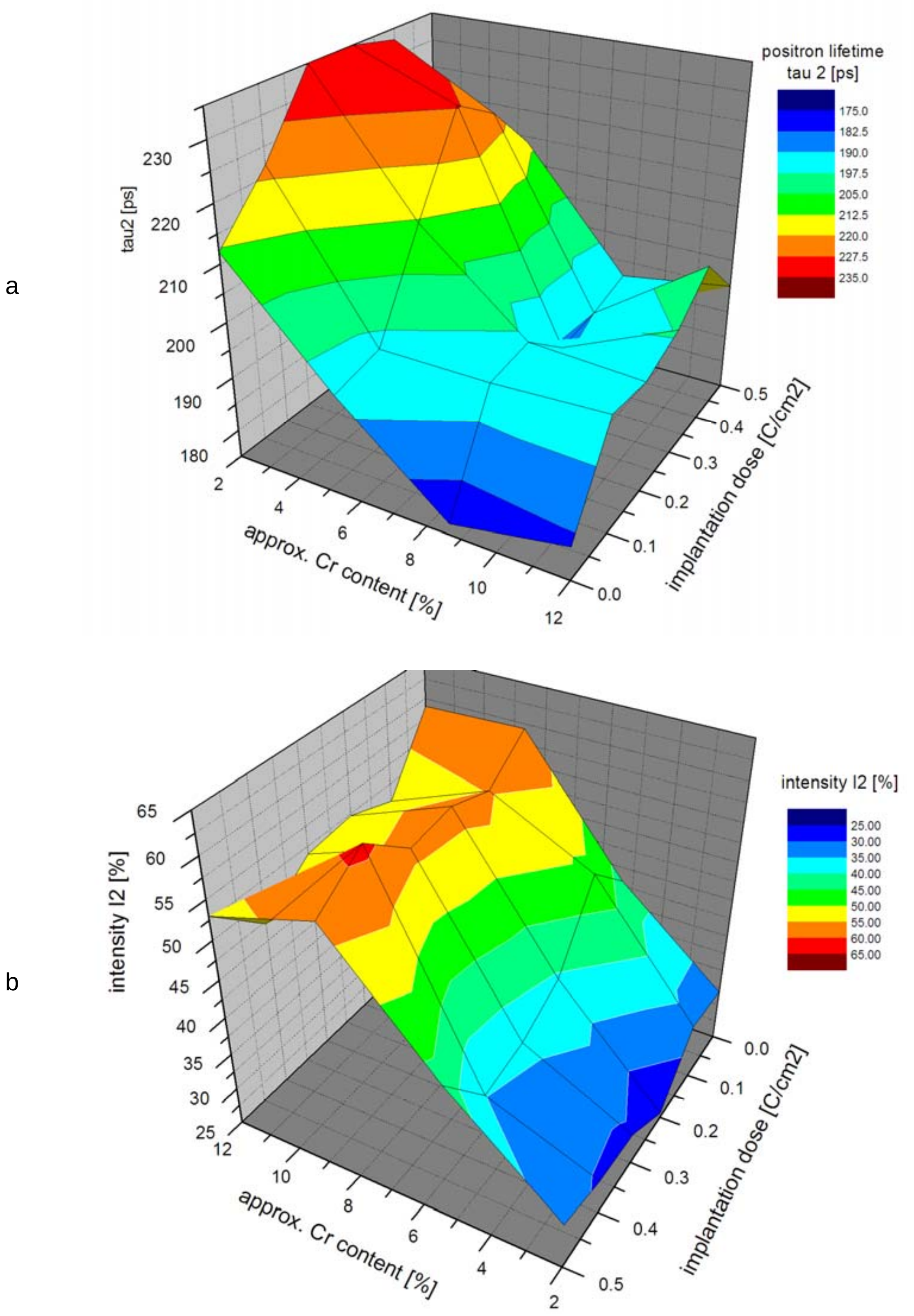

Fig.1. Positron annihilation in defects (component 2). Positron lifetime (a) and intensity of the annihilation (b)

These measurements clearly show that larger defects are created in the materials by the helium implantation. However, these defects are mostly created within one μm from the specimen surface. To study this damage zone in more detail with positron lifetime technique, measurements with PLEPS have been performed on the Fe11.62%Cr samples.

3.2. PLEPS measurements

The measured positron lifetimes spectra were analyzed using the LT 9.0 program [17] and a modified version of PosFIT [18]. The differences of these two analyses were negligible. All the lifetime spectra could be decomposed into three lifetime components with variances close to one.

Fig. 2a, 2b shows the positron mean lifetime as a function of helium implantation dose and mean positron implantation depth. The positron mean lifetime (MLT) is increasing with the implantation dose, thus indicating the creation of defects due to implantation.

The increase of the MLT close to the surface (<200 nm below the surface) is probably due to positrons annihilating in surface oxide layer. At higher depths the course of the MLT depth profile corresponds to the expected zone of maximum damage.

In the zone of maximum damage in the implanted specimens two different defect lifetimes have been observed. The shorter lifetime between 240 and 300 ps could be assigned to small vacancy clusters <6 vacancies or larger clusters filled with helium [19]. The longer lifetime between 400 and 500 ps corresponds to annihilation in large voids (>1 nm) [20]. The intensity of this longer component (I_3) increases dramatically with the helium implantation dose as can be seen from Fig. 2b. The course of the I_3 depth profile again corresponds to the expected zone of maximum damage.

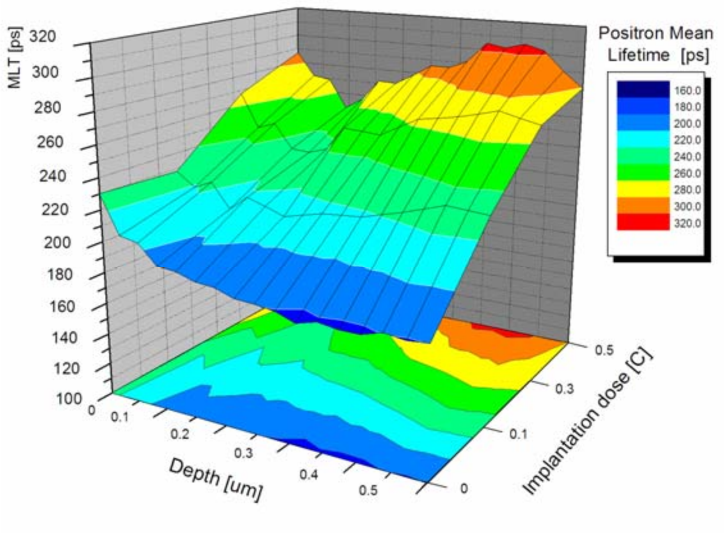

Fig. 2a. Positron mean lifetime in different treated Fe11.62%Cr alloy

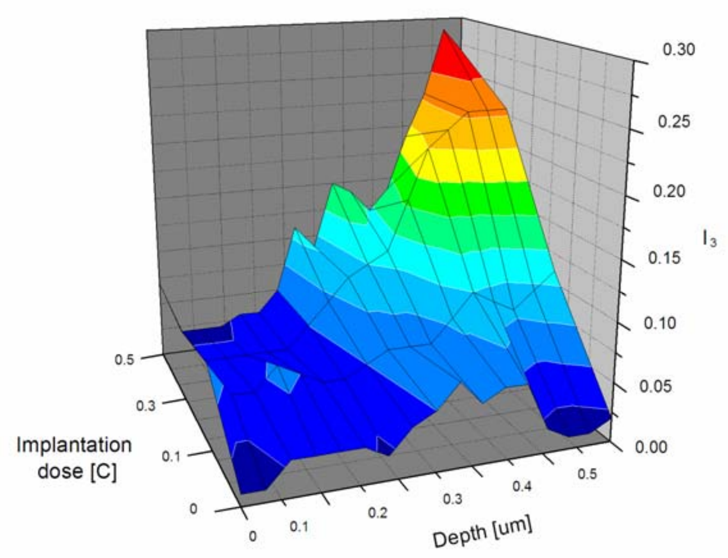

Fig. 2b. Intensity of annihilation in large defects (voids) measured in Fe11.62%Cr alloy

4. SUMMARY AND CONCLUSIONS

The present work demonstrates that conventional positron annihilation lifetime spectroscopy can provide valuable information about the microstructure of helium implanted Fe-Cr alloys. At the same time the connection between results from this technique and the pulsed slow positron beam lifetime measurements has been studied.

Positron lifetime experiments show that chromium plays an important role in the formation of the microstructure under radiation treatment. In particular, higher chromium content in FeCr alloys leads to a higher density of uniformly distributed small defects.

Depth profiles of defects, obtained with PLEPS, in the helium implanted region reflect the helium implantation profiles and show the creation of small vacancy clusters and large voids. These defects cannot be observed by any other technique in a non-destructive way.

Acknowledgement

This project was supported by VEGA 1/3188/06 and 7RP-Euratom/CU. The authors thank the NEPOMUC group of Ch. Hugenschmidt of FRMII at TUM for kindly cooperation and access to the PLEPS facility.

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СПЛАВЫ Fe-Cr, ИМПЛАНТИРОВАННЫЕ ГЕЛИЕМ И ИССЛЕДОВАННЫЕ МЕТОДОМ ИЗМЕРЕНИЯ ВРЕМЕНИ ЖИЗНИ ПОЗИТРОНОВ

Владимир Крсяк, Вернер Эггер, Мартин Петриска, Станислав Сояк

Исследовалось влияние хрома на стойкость к радиационному повреждению сплавов на основе железа с помощью общепринятого метода измерения времени жизни позитронов и импульсного пучка позитронов низкой энергии. Различные уровни доз гелия ($6{,}24\cdot10^{17}$-$3{,}12\cdot10^{18}$ см$^{-2}$), соответствующие локальному повреждению до 90 смещ./атом, накапливались в области толщиной менее 1 мкм. В настоящей работе использовались четыре бинарных сплава Fe-Cr (2,6; 4,6; 8,4; 11,6 вес. % Cr). Полученные результаты показывают, что хром оказывает значительное влияние на размер и распределение созданных дефектов. Характер этих дефектов определялся в виде больших пор (>1 нм) и малых вакансионных кластеров наряду с начальными дислокациями и малыми точечными дефектами.

СПЛАВИ Fe-Cr, ІМПЛАНТОВАНІ ГЕЛІЄМ ТА ДОСЛІДЖЕНІ МЕТОДОМ ВИМІРЮВАННЯ ЧАСУ ЖИТТЯ ПОЗИТРОНІВ

Володимир Крсяк, Вернер Еггер, Мартін Петріска, Станіслав Сояк

Досліджувався вплив хрому на стійкість до радіаційного пошкодження сплавів на основі заліза за допомогою загальновизнаного методу вимірювання часу життя позитронів та імпульсного пучка позитронів низької енергії. Різні рівні доз гелію ($6{,}24\cdot10^{17}$–$3{,}12\cdot10^{18}$ см$^{-2}$), що відповідають локальному пошкодженню до 90 зсувів/атом, накопичувались в області товщиною не менш 1 мкм. У даній роботі використовувались чотири бінарних сплави Fe-Cr (2,6; 4,6; 8,4; 11,6 ваг.% Cr). Отримані результати показують, що хром має значний вплив на розмір та розподіл створених дефектів. Характер цих дефектів визначався у вигляді великих пор (>1 нм) і малих вакансійних кластерів поряд з початковими дислокаціями і малими точковими дефектами.