Behaviour of the 16%Cr ODS ferritic steel intended for nuclear fusion power industry after tests in the conditions of irradiation in the Dense Plasma Focus facility PF-1000U

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Abstract. Fe-16Cr ODS ferritic steel with oxide nanoparticles fabricated by mechanical alloying method have been subjected to radiation tests using PF-1000U facility with subsequent analytical examination of exposed samples.

1. Introduction

As it is known, the oxide dispersion strengthened (ODS) ferritic or ferritic-martensitic steels are manufactured by the method of mechanical alloying executed from powders of elements (or their alloys) by means of adding of the ultra-dispersive fine particles of the oxide Y₂O₃ with subsequent extrusion or high-temperature isostatic molding [1]. These types of steel represent a new class of materials that can be used at heightened temperatures due to dispersion of thermo-stable nanoparticles of oxides in a matrix. It is expected that a use of such ODS steels must increase their resistibility in relation to creeping as well as their anticorrosion ability against oxidizing at high temperatures, and, consequently, results in an increase of the working temperature of the first wall and blanket of the future nuclear fusion reactors (NFR) up to approximately 700°C and higher [2].

Advantages of the ODS steels are in a great extent determined by the sizes of the nanoparticles and by their stability. However, the size of the basic particle of the dispersive oxide Y₂O₃ is increased at the compaction and at the thermo-mechanical treatment of the ODS steels. Because of the fact for the increase of the stability of these oxides nanoparticles titanium and aluminum are inserted additionally to obtain smaller and more stable oxide nanoparticles [1-3].

Complicated mechanisms including fragmentation, dilution and decay of Y₂O₃ nanoparticles as well as the secondary precipitation of the nanoparticles of new complicated oxides have been proposed and examined in a number of works [4-7]. The investigations were fulfilled with a help of X-ray structure analysis, transmission electron microscopy, atomic-probe tomography and low-angle neutron scattering. Besides owing to use of APT [8], EDX and EELS [9] methods an existence of the cores of
oxides complexes, connected with the porous structures, was revealed. This fact shows that the proposed earlier mechanisms of dilution and secondary precipitation of the complexes have a right to existence.

In the work [10] an investigation of the nanoparticles in the steel K3-ODS has been provided with a use of the transmission electron microscopy of high resolution (HRTEM). Examination of crystal and interphase structures of the oxides nanoparticles was executed for better understanding of mechanisms of creation of these nanoparticles within the ODS steels. Oxides nanoparticles formed in the ODS steel 16Cr-4Al-2W-0.3Ti-0.3Y2O3(K3) are mainly the oxides of the type Y4Al2O9(YAM) with monoclinic structure. Big nanoparticles (>20 nm) have spherical shapes that are incoherent with the matrix whereas small particles (<10 nm) are coherent or semi-coherent with the matrix. At the same time three orientation conformities are fulfilled. The nanoparticle structure representing by itself a crystal core of the oxide connected with an amorphous shell is observed both in big and small nanoparticles. They appear in a process of the K3-ODS steel manufacturing.

This “core-shell” structure disappears after a prolonged annealing at 900°C during 168 h. It supports the idea that this structure of the oxide nanoparticles is quite far from the chemically equilibrium state. Three-stage mechanism of the creation of the ODS nanoparticles including fragmentation, amorphization and crystallization is proposed at the moment for understanding of the “core-shell” structures of nanoparticles observed in the manufactured ODS steels.

Kinetics of phase separation into enriched Fe (α) phase and enhanced Cr (α') phase occurred in the oxide dispersion strengthened steels during the annealing at the temperatures 300-500°C has been analyzed by means of the method of tomography of atomic probe (APT) in a combination with the thermoelectric measurements (TEM) [11]. A small-scale decrease of the lattice parameter appeared after the annealing and measured by the X-ray structure analysis was resulted from the difference in lattices parameters among the incipient α and α' regions. Elastic energy related to this discrepancy and dissimilar moduli of elasticity of α and α' phases are able to lead to an increase of internal stresses that prevent movement of dislocations and consequently result in strengthening of the material. Therupon a particular interest is the investigations of the influence of radiation action upon the structure and properties of the ODS steels. They allow to look in a new way on the problem of behavior of the base of a steel and of the disperse particles in it from the point of view of its maintenance in radiation fields of nuclear fusion reactors.

In this work an investigation of an influence of irradiation upon a structure of the surface of the 16Cr-4Al-2W-0.3Ti-0.3Y2O3 ODS ferritic steel produced by streams of deuterium plasma and fast ions (deuterons) is provided.

2. Irradiation conditions

Devices of the Dense Plasma Focus type (DPF) are very effective facilities modelling the conditions realized inside the NFR. They are widely used for testing materials that are counted as the perspective ones for implementation in elements of the first wall of the reactors with magnetic, but in particular wit inertial plasma confinement [12-14]. In the current experiments an irradiation of samples of the above-mentioned material was provided inside the working chamber of the facility named PF-1000U [15] (Institute of Plasma Physics and Laser Microfusion, Warsaw, Poland). It was operated with the capacitor bank energy $E_c \approx 170$ kJ (initial charging voltage $U_0 = 16$ kV) with the pure deuterium as a working gas under the pressure $p_0 = 470$ Pa. Its discharge current amplitude was 1.175 MA and the current rise-time was equal to 6 μs.

Specimens under tests were fixed in a special holder and they were placed in the chamber’s center (on the Z-axis of it) at a distance $l = 50$ cm from the anode surface (see figure 1). From this photo one may see that the anode in these experiments had an orifice in its center to prevent evaporation of copper (anode lid’s material) by a powerful fast electron beam generated in a DPF.

As it is known [16] on a certain stage of the DPF discharge the very powerful streams of plasma with the plasma density near the top of the pinch equal to $n \approx 10^{18}$ cm$^{-3}$ and the velocities of about (2-3)$\times 10^7$ cm/s as well as of fast ions (deuterons) having energy in the maximum of its distribution
function $E_i \sim 100$ keV and ion density of approximately $N_i \sim 10^{18}$ cm$^{-3}$ are generated. They are directed to the side opposite to the DPF anode.

**Figure 1.** Photograph of the sample’s positioning in relation to the electrodes inside the chamber of the PF-1000U facility.

Generally speaking the maximal values of the power flux densities of the streams of plasma and fast ions in the PF-1000U facility are culminated in a close vicinity of the upper part of the pinch, i.e., in a distance of about 10 cm from the anode face. Their values are $10^{10}$ and $10^{12-13}$ W/cm$^2$ correspondingly. Up to the distance of 5-10 cm from the top of the pinch (i.e., 15-20 cm from the anode lid) the beam of fast ions is practically concentrated within a narrow angle around the Z-axis due to current and charge compensation of its current by a back-current of electrons induced behind the front of the shock wave (SW) that are produced by a cumulative jet of plasma [16]. Then after the penetration of the beam of fast ions ($v \approx 3 \cdot 10^8$ cm/s) through the SW front ($v_{SW} \approx 3 \cdot 10^7$ cm/s) the above compensations are violated, and the fast ion beam are scattered. After this the densities of both streams (plasma and fast ions) decrease by an inverse square law. Besides, power flux densities of these streams on the target surface (and of the fast ion stream in particular) are proportional to the neutron yield of the device.

Taking into consideration these factors the values of the power flux densities of the above-mentioned radiation streams on the target’s surface had magnitudes presented in a Table 1. A Table 2 presents data on the mass changes of the samples in results of their irradiation in the chamber of the PF-1000U facility.

| Sample   | Number of irradiation pulses | Integral neutron yield (number of counts) | Average neutron yield per shot (neutrons/pulse) | Power flux density of a plasma stream (W/cm$^2$) | Power flux density of a fast ion stream (W/cm$^2$) |
|----------|------------------------------|------------------------------------------|-----------------------------------------------|-----------------------------------------------|-----------------------------------------------|
| ODS Jap 3 | 2                            | 10 827                                   | $1.4 \cdot 10^{10}$                            | $3 \cdot 10^8$                                 | $1 \cdot 10^8$                                |
| ODS Jap 2 | 9                            | 85 567                                   | $2.5 \cdot 10^{10}$                            | $5 \cdot 10^8$                                 | $2 \cdot 10^8$                                |

| Sample | Number of irradiation pulses | Sample mass before irradiation, g | Sample mass after irradiation, g | Change of sample mass after irradiation, g |
|--------|------------------------------|----------------------------------|----------------------------------|-------------------------------------------|
| ODS Jap 3 | 2                            | 0.3149                           | 0.3143                           | -0.0006                                   |
| ODS Jap 2 | 9                            | 0.3532                           | 0.3525                           | -0.0007                                   |
3. Material and methods of investigations

Analytical examinations were provided with flat polished samples of the original steel of the type 16Cr-4Al-2W-0.3Ti-0.3Y₂O₃ (steel K3) having sizes 15×15 mm.

Figure 2. Exterior views of the samples of the ODS steel of the type Fe-16Cr-4Al-2W-0.3Ti-0.3Y₂O₃: a— virgin sample; b— after 9 irradiation pulses; c— after 2 pulses of the streams of deuterium plasma and fast deuterons.

After irradiations according to the programs presented in the table 1 the samples were investigated by means of scanning electron microscopy using a device LEO1420 (Carl Zeiss) with a system of the energy dispersion micro-analysis INCA300 (Oxford Instruments) and X-ray diffraction analysis with the help of the diffractometer Rigaku (XRD) Ultima-IV (Japan). In these examinations the line of copper CuKα was applied.

The samples have been also investigated by a method of a semi-contact (tapping-mode) atomic-force microscopy (AFM). The samples were examined in different zones, the sizes of the images under inspection were 20×20 and 5×5 μm²; for a single region at each sample the images were formed for the sizes 1×1, 10×10 and 50×50 μm². After that the average parameter of roughness Rₐ and the difference in the peak’s heights Rₘₐₓ for each sample were calculated. An averaging procedure was provided with images of equal sizes (5×5 and 20×20 μm²) taken from dissimilar regions of a sample. Standard deviations were also computed.

4. Results of investigations

In figures 3-5 microstructures of the virgin and irradiated samples after 2 and 9 pulses of deuterium plasma and fast deuterons obtained by the method of scanning electron microscopy are presented.

Figure 3. SEM images of microstructure of a virgin sample of the 16Cr-4Al-2W-0.3Ti-0.3Y₂O₃ (K3) ODS steel.
Figure 4. SEM images of the microstructure of a sample of the 16Cr-4Al-2W-0.3Ti-0.3Y_2O_3 (K3) ODS steel after 2 irradiation pulses of powerful streams of deuterium plasma and fast deuterons.

Figure 5. SEM images of the microstructure of a sample of the 16Cr-4Al-2W-0.3Ti-0.3Y_2O_3 (K3) ODS steel after 9 irradiation pulses of powerful streams of deuterium plasma and fast deuterons.

Microstructure of the sample irradiated by 2 pulses does not show any noticeable distinctions from the virgin sample. However, machining marks on the polished surface of the sample disappear (figure 4). It denotes that the ion polishing of the sample at the irradiation takes place.

Figure 6. X-ray diffraction patterns of the samples of the 16Cr-4Al-2W-0.3Ti-0.3Y_2O_3 (K3) ODS steel: a — virgin sample; b — after 2 pulses; c — after 3 pulses of streams of deuterium plasma and fast deuterons.

Note that the peculiarity of all samples investigated by the X-ray structure method is the presence of two solid solutions in the composition of the steel (Table 3, figure 6).
### Table 3. Phase composition and lattice parameters before and after irradiation

| Sample number | Number of irradiation pulses | Phases found     | Lattice parameter $a$, Å | Change of the lattice parameter after irradiation $\Delta a$, Å |
|---------------|------------------------------|------------------|--------------------------|------------------------------------------------------------|
| 1             | 0                            | Fe, Cr           | 2.8823                   |                                                            |
|               |                              | Cr$_{0.7}$Fe$_{0.3}$ | 2.8724                   |                                                            |
| 2             | 2                            | Fe, Cr           | 2.8678                   | –0.0145                                                   |
|               |                              | Cr$_{0.7}$Fe$_{0.3}$ | 2.8779                   | +0.0055                                                   |
| 3             | 9                            | Fe, Cr           | 2.8777                   | –0.0046                                                   |
|               |                              | Cr$_{0.7}$Fe$_{0.3}$ | 2.8685                   | –0.0039                                                   |

In the virgin sample 95% belongs to the phase enriched by Fe as it is in the ferritic steel of the type 434 SS and 5% — a phase enhanced with Cr (80%). It seems that this structure is a consequence of a spinodal decomposition, i.e. of those irreversible process that determined by local fluctuations of the concentrations of the mixture components. At the same time a spinodal decomposition take place uniformly throughout all the material in opposite to the metastable phase decay connected with the creation of nuclei. It is possible to suppose that the prolonged thermal treatments in the temperature range 450-500°C taking place during the manufacturing of the ODS steel help to the formation of the high-chromium phase. Besides it is known that a separation of carbide, nitride or solicited phases during the initial stage of a spinodal decomposition accelerates its process. It takes place as early as during a process of the ODS steel manufacturing directly after the hot isostatic pressing of this material [17].

It the sample irradiated by 2 pulses of deuterium plasma and fast deuterons this ratio and phase composition are preserved. However, the lattice parameters are slightly decreased due to elimination of impurities from the surface (Table 3). In the sample irradiated by 9 pulses of the above-mentioned radiations the phase composition is changed due to a release of a noticeable amount of energy that is resulted in melting. The phase enriched by chromium is practically disappear that is evidenced in favor of a formidable increase of temperature during irradiation and of redistribution of doping elements and height difference resulting in dilution of the phase enriched by Cr.

In figure 5 one may see a striped structure with extended granules and regular separations of the phase particles in the form of relatively large white spots having a size up to 50 μm and more. They can be defined as large particles of the complex oxide Y$_4$Al$_2$O$_9$ [11].

Below (figures 7, 8 and 9, Tables 4, 5 and 6) the characteristic images of the samples, their roughness and heights differences obtained with a semi-contact (tapping-mode) atomic force microscopy are presented.

![Figure 7](image-url) Images of the virgin sample obtained with the help of the semi-contact (tapping-mode) AFM method.
Table 4. The surface roughness and heights differences estimations in the virgin polished sample obtained by an AFM method

|      | 5x5     | 20x20   |
|------|---------|---------|
| $R_{a}$, nm | $R_{max}$, nm | $R_{a}$, nm | $R_{max}$, nm |
| 5.767 | 31.63  | 9.399  | 73.81  |
| 5.193 | 28.25  | 9.253  | 65.14  |
| 4.206 | 31.19  | 7.892  | 62.48  |
| 7.471 | 42.98  | 10.14  | 104.5  |
| 6.949 | 37.43  | 10.48  | 71.51  |
| 5.9±1.2 | 34±5  | 9.4±0.9 | 75±15  |

Comparison of Tables 4-6 shows that the 9-fold irradiation results in a certain increase of roughness and heights differences compared with the virgin sample and the sample irradiated by 2 pulses.

Figure 8. Images of the the sample irradiated by 2 pulses of deuterium plasma and fast deuterons obtained with the help of the semi-contact (tapping-mode) AFM method.

Table 5. The surface roughness and heights differences estimations in the sample irradiated by 2 pulses of deuterium plasma and fast deuterons obtained by an AFM method

|      | 5x5     | 20x20   |
|------|---------|---------|
| $R_{a}$, nm | $R_{max}$, nm | $R_{a}$, nm | $R_{max}$, nm |
| 4.718 | 35.38  | 7.102  | 80.26  |
| 6.959 | 42.57  | 8.404  | 69.25  |
| 6.758 | 48.02  | 7.843  | 83.65  |
| 3.84  | 29.77  | 6.366  | 60.4   |
| 6.046 | 40.62  | 9.632  | 81.26  |
| 4.958 | 30.47  | 7.613  | 70.34  |
| 5.5±1.1 | 38±7  | 7.8±1.0 | 74±8   |

Figure 9. Images of the the sample irradiated by 9 pulses of deuterium plasma and fast deuterons obtained with the help of the semi-contact (tapping-mode) AFM method.
Table 6. The surface roughness and heights differences estimations in the sample irradiated by 9 pulses of deuterium plasma and fast deuterons obtained by an AFM method

|        | 5×5     | 20×20   |
|--------|---------|---------|
| $R_a$, nm | 4.718   | 6.959   |
| $R_{max}$, nm | 35.38   | 42.57   |
| $R_a$, nm | 6.758   | 3.84    |
| $R_{max}$, nm | 48.02   | 29.77   |
| $R_a$, nm | 6.046   | 4.958   |
| $R_{max}$, nm | 40.62   | 30.47   |
| $R_a$, nm | 5.5±1.1 | 9.632   |
| $R_{max}$, nm | 38±7    | 7.8±1.0 |

5. Conclusions

The phase composition of the steel under investigation in its virgin state demonstrates a presence of the two solid solutions: a phase enriched by Fe with the lattice parameter $a = 2.8823$ Å in quantitative terms 95%, and a phase with an enhanced content of chromium (80%Cr) with the lattice parameter $a = 2.8724$ Å in numerical terms 5%. This is evidence of a partial spinodal decomposition of the solid solution.

Irradiations by 2 pulses of the powerful streams of high-temperature plasma and fast deuterons in the PF-1000U facility with targets placed at a distance of 50 cm from the anode face does not result in a noticeable change in its phase composition whereas at 9 pulses a formidable modification of the phase structure takes place. A practical disappearance of the phase enriched with Cr is observed, and the second phase with enhanced content of Fe is appeared. It is resulted from the absorption of the large amount of energy on the surface of the steel sample and from the surface layer melting.

As a result of the selective evaporation from the surface its roughness is increased to some extent with the number of pulses.

A structure of the steel reinforced by the particles of the complex oxides observed after irradiations by powerful streams of high-temperature plasma and fast deuterons is preserved. It evidences in favor of its high stability in the conditions of the above irradiations.

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