MICROSTRUCTURE AND PHASE TRANSFORMATIONS IN THE ODS ALLOYS IRRADIATED BY SWIFT HEAVY IONS

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Microstructure of KP4 ODS alloy irradiated with 700 MeV bismuth ions at 300K has been studied using high resolution transmission electron microscopy. No latent tracks have been observed in $Y_4Al_2O_9$ particles in KP4 irradiated with Bi ions. Small oxides (~ 5 nm) in KP4 alloy remain crystalline at Bi ion fluence 1.5×10^{13} cm^{-2}, while subsurface regions in large (~ 20 nm) particles faced to the beam entrance became amorphous.

Introduction

Being considered as promising candidate materials for future reactors, oxide dispersion strengthened (ODS) steels are subject of extensive irradiation testing with various radiation sources. Recent TEM examinations of the ODS alloys irradiated with high energy Xe and Kr ions have revealed strong sensitivity of oxide particles to dense ionization [1-3]. In particular, some nanoparticles in a Fe–13Cr–1.5Mo steel (DT2203Y05 or DY) reinforced by yttrium and titanium oxides were found to undergo amorphization after 92 MeV Xe ion irradiation to fluence 3.2×10^{14} cm^{-2} at room temperature, whereas no dissolution seemed to occur [1].

Another, the EM10 ODS alloy with silicium-magnesium mixed oxides, studied in this work, exhibited similar effect – amorphization of some nanoparticles. The amorphous precipitates in both materials are coexisting with other precipitates which remain crystalline. Also, no dissolution of oxide nanoparticles has been evidenced in TEM studies of a Fe18Cr1W – $0.5Y_2O_3$ alloy irradiated at the same conditions [2]. At the same time, the nanoparticles lost contrast by HRTEM that can be associated with their amorphization.

The experiments started in [1,2] were continued in work [3], where the single impact regime (10^{12}, 74 MeV Kr ions×cm^{-2}) at room temperature was used to eliminate the role of the high electronic energy deposition under swift heavy ion irradiation in structural transformations of oxide particles in the DT2203Y05 ODS alloy. This material was chosen due to presence of large sized (d > 50 nm) oxide particles that could enable the observation of ion tracks, which typical radii are in the few nm range in most oxides. Indeed, the TEM examination has revealed amorphous tracks in large (Y, Ti) oxides with density close to the Kr ion fluence, thus confirming that these tracks are formed as a result of dense ionization.

Since the sensitivity of nanoparticles in the ODS alloys was found to be dependent on many parameters, like the nature of the oxides, ion fluence, irradiation temperature, this stimulates farther research with involving of new materials and variation of irradiation conditions.

In this paper we present the results of TEM studies of Fe-15Cr-4Al-2W-0.35Y2O3 ODS (KP4) ODS alloy irradiated with 700 MeV Bi ions.

Experimental procedure

The materials used in our studies was KP4 Fe-15Cr-4Al-2W-0.35Y2O3 ODS alloy (Kyoto University). High energy ion irradiation at room temperature were performed at the U-400 (Bi) cyclotrons at FLNR JINR, Dubna. Ion beam homogeneity better than 5% on irradiating specimen surface has been reached using beam scanning in horizontal and vertical directions. Average Bi ion flux was 10^9 cm^{-2}s^{-1}. Ion flux has been continuously monitored by measuring of ion beam current from Faraday cup, which bottom serves as target holder. Accuracy of ion flux measurements is 15%. The heating of Bi irradiating samples was no more a few degrees of centigrade.

The samples used in our irradiation experiments were the bulk materials in comparison with pre-thinned specimens studied in works [1-3]. Cross-sectional TEM specimens have been prepared using FEI Helios Nanolab 650 FIB technique. Milling was done with 30keV Ga ions and polishing down to 500eV. Cleaning the FIB lamella at 500 V gallium beam energy provided a damage-free surface of TEM targets.

All samples were characterized within depth about 1 μm from the surface by using a JEOL JEM 2100 LaB_6 transmission electron microscope operating at 200 kV. Additionally to XTEM targets, several plain-view specimens, prepared by mechanical polishing, dimple grinding and the ion-beam milling technique, have been analyzed also.

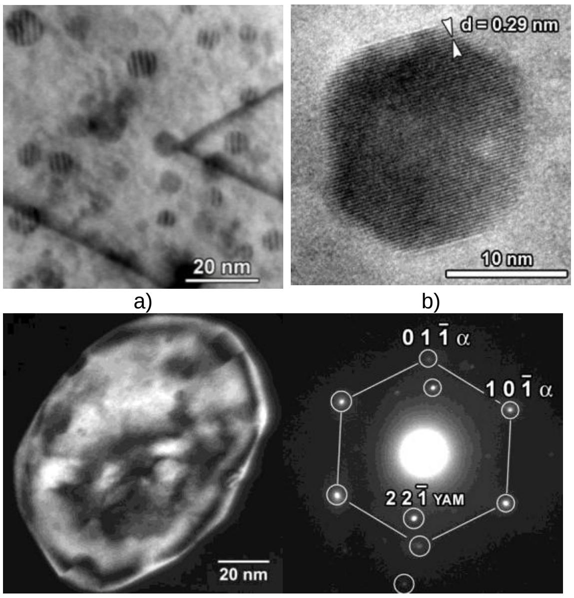
Results and discussion

Considering the irradiation effects we are, first of all, interested in structural response of nanoparticles to dense ionization induced by high energy heavy ions and will not discuss the radiation damages in ferritic matrix. The first conclusion, which can be done after analysis of Bi ion irradiated KP4 specimens, is absence of specific features observed for metal NPs in oxide matrices bombarded with swift heavy ions [6,7] like shaping along ion beam direction or formation of satellites around of NPs followed by their partial dissolution. Taking into account that typical track diameter in insulators is in the range 2-10 nm [8], the Bi ion fluences 1.5×10^{12} and 1.5×10^{13}

cm^{-2} correspond to different irradiation conditions – regimes of individual(I) and multiply overlapped (II) ion track regions. It is suggested that ion tracks start to overlap when $\Phi\times\pi R^2 = 1$, where Φ is ion fluence and R – track radius.

The nanoparticle population in KP4 is composed of $Y_4Al_2O_9$ (YAM: yttrium aluminum monoclinic) oxides with sizes ranging from 2 nm to 30 nm, with a mean particle size of 5 nm as demonstrated in Fig. 1a. Images of majority YAM particles shown in this figure have moire fringes, which are only slightly misaligned to each other, indicating on the same orientation in a matrix.

An example of high-resolution image of single YAM nanoparticle is given in Fig. 1b. Like in [4,5], where the results of detailed analysis of KP3 (KP4) ODS alloys are presented, small nanoparticles (< 10 nm) are coherent or semi-coherent with the matrix, while larger oxides (~ 20 nm) tend to be incoherent with the matrix due to amorphous shell. The particle density in our KP4 samples is on the order of ~ $10^{16}cm^{-3}$. Fig. 1c, d show dark field image of for relatively large YAM particle and corresponding SAD pattern, proving its single crystalline structure. Several stacking faults are seen in this image only.



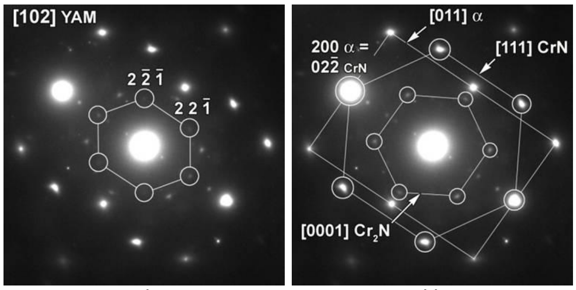
a) b)

c) d)

Fig. 1. Two beam bright-field image showing moire contrast in YAM nanoparticles in KP4 steel (a). High-resolution TEM image of single YAM nanoparticle (b). Dark-field image (c) and corresponding SAD pattern from relatively large YAM particle (d).

Typical SAD from single grain and results of diffraction analysis are given in Fig. 7a-c. Diffraction reflections from the α-Fe matrix are denoted by index α. The measured lattice parameter of matrix a = 0.289 nm is a bit higher than those known for pure α-Fe, probably due to the presence of Cr in a solid solution. One of the SAD patterns shown in Fig. 2c was indexed as fcc crystal structure coinciding with the pattern from [111] zone axis of CrN (CODdata_1008956: Acta Crystallographica B (24,1968-38,1982), a=4.148 Å, the measured value is 4.09 Å). Another pattern belong to hexagonal Cr_2N ($P\overline{3}m$, a = 4.81 Å, measured value is 4.9 Å, c = 4.48 Å; CODdata_4311894: Inorganic Chemistry 43, p.7050-7060, (2004), [001] zone axis).

Dark-field images of KP4 in CrN and Cr_2N reflexes look like homogeneously distributed small (< 1 nm) spots.

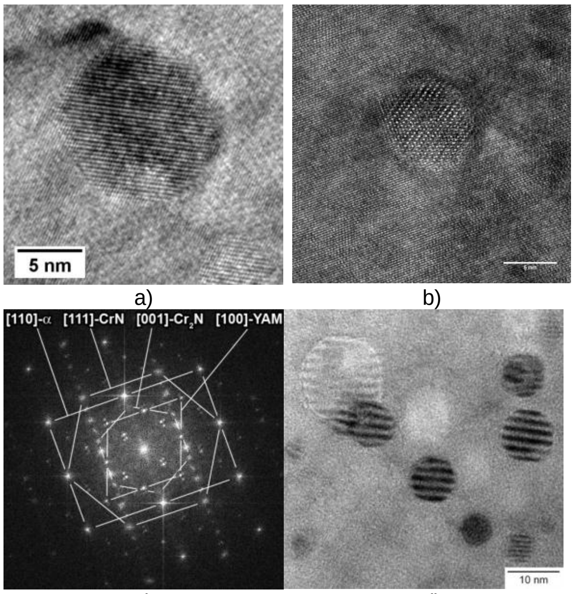


a) b)

Fig. 2. Diffraction analysis of single grain microstructure in KP4 alloy showing the presence of CrN and Cr_2N phases.

No Bi ion induced latent tracks, as well as crystalline to amorphous phase transition, have been observed in $Y_4Al_2O_9$ oxides in KP4 steel. Typical HRTEM images of YAM particles from samples irradiated by 6.0×10^{12} cm^{-2} and 1.5×10^{13} cm^{-2} Bi ions are given in Fig. 3a, b. They show no difference in morphology and microstructure of YAM NPs in comparison with those in unirradiated material.

It is confirmed also by FFT image from micrograph presented in Fig. 3b, demonstrating all crystalline phases in KP4 sample irradiated with 1.5×10^{13} cm^{-2} Bi ions (Fig. 3c) and image of YAM particles obtained from the target exposed to this ion fluence having moire fringeslike in virgin material (Fig. 3d).

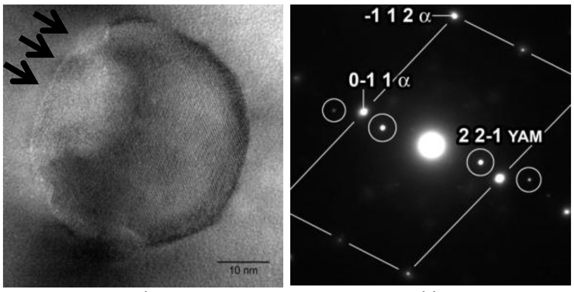


a) b)

c) d)

Fig. 3. HRTEM images of small (< 10 nm) YAM nanoparticles in KP4 sample irradiated with 6.0×10^{12} (a) and $1.5\times10^{13}cm^{-2}$(b,d) Bi ions. SAD pattern from the region shown in b.

The difference in structural response between small and large YAM nanoparticles to Bi ion bombardment is demonstrated by Fig. 4a, b. As was found, Bi ions induce characteristic "erosion" of part of large (diameter more 20 nm) particles faced to the beam direction and transforming it into amorphous state (sometimes this effect was observed also in the back side of the NP). At the same time, the rest of the NP remains crystalline as seen from the corresponding SAD pattern.



a) b)

Fig. 4. Typical HRTEM image of large (>20 nm) YAM nanoparticle in KP4 sample irradiated with $1.5\times10^{13}cm^{-2}$ Bi ions and corresponding SAD pattern. The arrows indicate the ion beam direction.

Similar effect was never seen in small (~ 5 nm) NPs. This strongly implies that above structural changes are dependent on coherency of NPs with the matrix and role of the interfacial region is very important in radiation damage formation via dense electronic excitations in nanooxides embedded in metal matrix.

Conclusion

Dense electronic excitations in the wakes of high energy Bi ions induce significant changes in microstructure of oxide nanoparticles in KP4 ODS alloys.

It was found that $Y_4Al_2O_9$ particles have demonstrated a high resistance to amorphization. No latent tracks have been observed in $Y_4Al_2O_9$ particles.

Small oxides (~ 5 nm) in KP4 alloy remain crystalline at Bi ion fluence 1.5×10^{13} cm^{-2}, while subsurface regions in large (~ 20 nm) particles faced to the beam entrance became amorphous.

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