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Surface morphological evolution and nanoneedle formation of 18Cr-ODS steel by focused ion beam bombardment

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ABSTRACT

Morphological evolution upon intense energetic particle–matter interactions is of fundamental importance for materials utilized in extreme radiation environment, and controlling surface morphology by radiation also provides a new pathway for exploring non-equilibrium process at surface. In this work, surface morphology and microstructural evolution upon low energy ion irradiation of 18Cr-ODS, a candidate structural material for cladding and first wall of future fission and fusion reactors, were investigated by *in situ* focused Ga^+ ion beam/scanning electron microscopy and *ex situ* transmission electron microscopy. A surface roughening through pore formation, coalescence and eventually nanoneedle formation was induced on 18Cr-ODS steel surface. Cross-section microstructure analysis indicates that the formation of nanoneedle was not a result of grain recrystallization or chemical composition change. Pre-irradiated materials by He^+ and Fe^+ ions displays enhanced kinetics for surface morphological evolution and lower fluences of focused Ga^+ are required for the nanoneedle formation. These results suggest that the surface roughening and morphological evolution of 18Cr-ODS under low energy ion irradiation is an interplay between a curvature-dependent sputtering and defect accumulation near the surface.

1. Introduction

Ion-induced nanoscale structure formation on the material surface has attracted great interest due to both the underlining fundamental material science and its potential as a processing technique for nanodevices, sensors, and magnetic storage media. These appealing morphology induced by focused ion beam (FIB), including nanoripples [1], nanofibers [2], nanowires [3] and nanodots [4], has been reported on a variety of materials under normal incidence or off-normal incidence without sample rotation. These typical microstructures generally formed at temperatures below the melting point of a particular nanostructure, yielding mechanically weak and often thermally and chemically unstable arrays [5,6]. The distinct nanoneedle microstructure induced by FIB on tungsten has been reported [7]. A general consensus about the evolution of nanoneedle structure during ion bombardment is developed as being due to the competition between surface roughening introduced by sputtering and smoothing created by various processes, such as curvature dependent sputtering [8,9], thermal diffusion [10], redeposition and ion induced diffusion [11,12], viscous flow [10,13], and preferential sputtering without actual mass movement [14,15]. The mechanism for nanoneedle formation still remains controversial and needs to be further identified.

The surface roughening and morphological evolution are sensitive to materials and different irradiation conditions such as ion mass, beam energy and direction. The final surface pattern is a result of interplay between surface roughening/smoothening kinetics and defects near the surface. *In situ* observation of morphological evolution of materials exposed to ion irradiation is important not only for the fundamental understanding of ion–matter interaction in probing materials behavior under extreme radiation environment, but also for characteristic structure fabrication applied in quantum, photon and phonon device. As an efficient and direct approach [16,17], focused ion beam (FIB) technology enables the real time *in situ* observation that can provide and display the surface microstructure evolution in a temporal and spatial manner, and also enables the control of irradiation parameters separately and precisely. In this work, the morphological and microstructure evolution of an oxide dispersive strengthen (ODS) alloy, a very important group of structural materials for cladding and first wall of future fission and fusion reactors, were investigated by FIB low energy bombardment. The surface roughening

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process and the mechanism of formation of nanoneedle-structure were investigated, and the impact of the surface/subsurface defects induced by pre-irradiation was also explored.

2. Experiment

The 18Cr-ODS steel used in this work was prepared by mechanical alloying and hot extrusion with the nominal chemical composition of Fe–18Cr–8Ni–0.5Mo–0.3Ti–0.35Y_2O_3 (mass in percent). The sample was mechanically polished by SiC sand-paper from 240 to 1200 grid and finally polished using 0.5 μm diamond paste. The surface roughness was measured before and after ion beam bombardment using a Bruker Dimension Icon AFM instrument. The focused Ga^+ ion beam bombardment experiments were carried out at room temperature without sample rotation using a field emission scanning electron microscope (SEM)/FIB dual-beam system (FEI Nova 200 NanoLab). During the FIB bombardment, a pre-designed rectangular area (e.g., $25 \times 25\ \mu m^2$) was selected with the dwell time of 1 μs and the beam spot overlapping 50% for each FIB patterning. The morphological variation and microstructure evolution of irradiated samples were analyzed by *in situ* SEM imaging and *ex situ* TEM. The TEM samples of the nanoneedles were prepared using FIB *in situ* technology [18] and then investigated using a JEOL 3011 electron microscope operated at 300 kV.

3. Results and discussion

The morphology and topography of unirradiated 18Cr-ODS steel surface were investigated by atomic force microscopy (AFM) as shown in Fig. 1. AFM image was acquired in a contact mode. A few fine scratch lines exist and some nanosized pits and embossments remain on the specimen surface due to the diamond sand damage during the mechanical polishing, which could affect the sputtering during ion beam bombardment. The cross-section analysis of the AFM 3D image shown in Fig. 1b indicates that the surface roughness value (Rz) of the mechanically-polished (MP) 18Cr-ODS steel is about 13 nm. Upon low energy focused Ga^+ bombardment, the morphology of specimen surface, as revealed by *in situ* SEM/FIB imaging, goes through a sequence of evolution: (1) the surface roughening upon low energy ion sputtering; (2) the formation of small-sized pore on the specimen surface, pore enlargement/coalescence with increasing ion fluences; and finally, the formation of nanoneedle-like microstructure. Fig. 1c shows nanoneedles with average length of 3.8 μm formed on the 18Cr-ODS steel surface under 30 keV Ga^+ bombardment at a fluence of 8.74×10^{18} ions/cm^2 (ion flux: 1.09×10^{16} ions/cm^2 s and incident angle: 52°). The nanoneedles are aligned with ion beam direction upon bombardment at an incident angle 52°. With further increasing of ion fluence, the nanoneedles become finer and longer. A similar ion beam bombardment-induced phenomenon was also observed on pure tungsten surface [7], in which morphological evolution induced by ion bombardment strongly depends on the ion beam conditions (energy, incident angle, etc), and the initial surface morphology as a result of dynamic competition between the surface roughening and smoothing process. The difference between tungsten and 18Cr-ODS steel is the different ion fluence threshold for nanoneedle formation at the same bombardment parameters, which can be attributed to the distinction of material and physical parameters, e.g., crystal constant, heat conduction, and surface sputtering ratio.

To gain deep insight into the formation mechanism of nanoneedles induced by focused Ga^+ ion bombardment, we use *in situ* FIB preparation technology to prepare TEM samples on the formed nanoneedles. The bright field TEM image of cross-section nanoneedle is shown in Fig. 2(a). The nanoneedles are induced after 5.46×10^{18} ions/cm^2 Ga^+ ion bombardment under 30 keV, at 7 nA and 52° incident angle. The selected area electron diffraction (SAED) patterns of two Fe grains with different crystal orientations are shown in Fig. 2(b) and (c), respectively. The formed nanoneedles are polycrystalline, as referred by the contrast variation of the bright field TEM image (Fig. 2a) and well defined grain boundary between location grains A and B. The evidence above reveals that the formed nanoneedles are not induced by grain recrystallization and regrowth during Ga^+ bombardment. High resolution TEM image (Fig. 2d) of the nanoneedle tip shows the lattice fringes of the Fe (1 1 0) plane. An amorphous layer with several nanometers width can be found on the edge of needle which can be attributed to the bombardment effect of FIB. The EDS analysis of the nanoneedle tip is shown in Fig. 2(e). Fe, Cr, Ni, Yi and O peaks in the EDS spectrum come from the original chemical composition. The observed Ga and Cu peaks come from the injected Ga^+ ions during FIB bombardment and the supporting copper grid, respectively. Similar EDS results are obtained at several locations of the same nanoneedle tip, even though under the high-angle annular dark-field (HAADF)–STEM (Z-contrast) mode. Quantitative analysis of the EDS spectra before and after irradiation shows no significant chemical composition change for the nanoneedle.

The surface morphological evolution and patterning formation induced by focused ion beam bombardment has been intensely investigated over the past decades [9–15]. Bradley and Harper (BH) provided a linear instability model to describe the evolution behaviors of surface morphology on the base of the Sigmund mechanism and surface diffusion [8]. BH model pointed out that

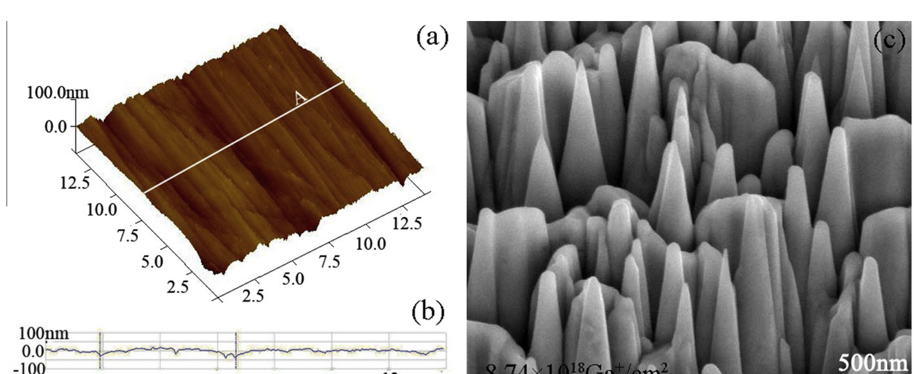

Fig. 1. (a) 3D AFM image showing the morphology and topography of the mechanically polished 18Cr-ODS steel surface and (b) the section analysis of AFM image from line 'A' in (a). (c) SEM image showing the surface morphology under 30 keV focused Ga^+ ion beam bombardment with 8.74×10^{18} ions/cm^2 fluence. The incident angle of ion beam is 52°.