Microstructure damage of titanium films by irradiation with fission fragments

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ABSTRACT

Radiation damage caused by fission fragments to metal surfaces is an important research topic. Thin titanium foils were irradiated with a continuous wave beam of 132 MeV ^{132}Xe^{+29} at the current intensity of 2 pnA. Pre- and post-irradiated surface topologies were investigated using atomic force microscopy and the observed defects were quantified by root mean square roughness, depth profile of the disordered zones, size and areal density of the voids, and discussed as a function of the applied fluencies (1–9) $\times 10^{13}$ Xe/cm^2. The first ellipsoidal dislocation loops appeared at the fluence of 3.0×10^{13} Xe/cm^2 with the areal density of 1.56×10^6/cm^2 that increased to 2.0×10^7 cm^{-2} when the dose rose to 9.0×10^{13} Xe/cm^2. At this point also the first dislocation lines with the density of 1.3×10^7 cm^{-2} were seen. Our results suggest that the fission fragments might maximize large voids and dislocations and increase the degradation in depth resolution.

1. Introduction

The structural changes induced in materials used in the nuclear industry due to radiation damage are of a great interest in nuclear science and engineering. Materials used in nuclear reactors are designed to withstand the extreme conditions of radiation damage during many years of reactor operation. The main physical process that produces heat is the deceleration and stopping of high-energy heavy ions present in the fission products (FP). The energy released in fission occurs primarily in the form of the kinetic energy of the fission fragments having the atomic numbers A = 75–160. The light fragment energies are about 100 MeV while the heavy fragment energies are about 70 MeV [1]. Additional important damage comes from intense fluxes of different energy neutrons, intense fields of beta and gamma radiation from the decaying fission products and alpha decay of the actinides, especially from the minor actinides generated by successive neutron capture during reactor operation. However, the alpha decay of ^{241}Am produces only about 1700 atom displacements, mostly caused by heavy recoil atom of ^{237}Np with a very short range of about 20 nm causing a collision cascade, while the typical products of U-235 fission, Ba and Kr, cause ~40,000 defects by light FP (Kr) and a ~60,000 defects by

heavy FP (Ba), for a total of about 100,000 defects [1]. Accumulated radiation damage leads to amorphization of an originally crystalline phase [2,3], or polygonization (grain subdivision), as occurs in UO$_2$ fuel [1]. In addition to the large number of atom displacements, the accumulated fission damage includes changes caused by precipitation of volatile fission products and chemical changes caused by beta decay of fission products into new elements with different reactivity. Hence, irradiation by fission products is a multifaceted problem that has been of interest to researchers for decades; however, the mechanism of damage during irradiation (reactor operation) and post-irradiation effects are not fully understood yet.

One solution to a better understanding of the complex problem of radiation damage by FP is using accelerator beams with different intensities and energies of heavy ions. We have used irradiation by ^{132}Xe^{29+} at ATLAS, the Argonne Tandem-Linac Accelerator System in Argonne National Laboratory, to study the FP irradiation effects of uranium dioxide targets, using UO$_2$ deposited on thin metallic support backings made of aluminum and titanium foils that are frequently used as a standard or backing material for irradiation targets or as a component of advanced nuclear materials is an important research topic. Atomic force microscopy (AFM), scanning transmission electron microscopy and energy-dispersive X-ray spectroscopy (SEM-EDX), and electron microprobe (EPM) were the diagnostic tools used to characterize the structures of pre- and post-irradiated surfaces. In the present paper we report and discuss the results of our work dedicated to the FP radiation damage of the titanium backing surfaces.

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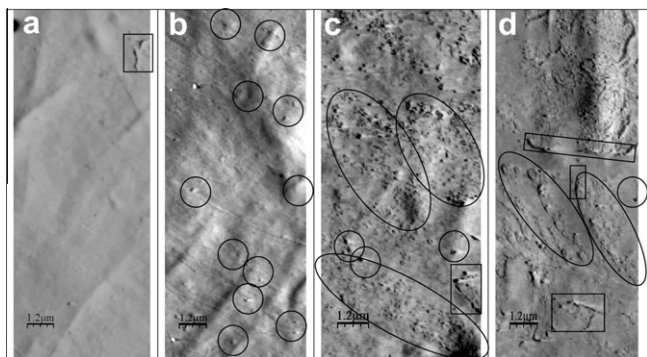
Fig. 1. AFM images of surface damage evolution after irradiation to fluencies of (a) 0, (b) 1.2×10^{13}, (c) 3.0×10^{13} cm^{-2}, and (d) 9.0×10^{13} cm^{-2} (circle = void, ellipse = dislocation loop, rectangle = dislocation line).

2. Experimental

2.1. Sample preparation

The titanium metal foils were purchased from Goodfellow as pre-cut 5×5 cm square with a thickness of 0.904 mg/cm^2 Ti. The irradiation target sample holder was a custom-made, ladder-type frame, made of aluminum, and suited for holding four targets in a vertical row. The outer dimensions of each from four frame windows were 24.1×14.9 mm^2 with the 10 mm diameter hole in the middle.

2.2. Irradiation procedure

Samples were irradiated with the continuous wave particle beam of ^{132}Xe^{+29} with energy of 132 MeV and with the current intensity of 2 pnA. The target ladder was mounted to the feed-through, which allowed the ladder to move along a vertical axis for sample positioning. Beam spot was about 0.1 cm in diameter. Three different fluences, $(1.2, 3.0, \text{and } 9.0) \times 10^{13}$ ions/cm^2, were delivered with the irradiation times of 2, 5, and 15 min, respectively. The pressure in the target chamber was about 5×10^{-7} Torr.

2.3. Surface examination

A detailed surface examination was made using atomic force microscopy (VEECO/Digital Instruments). Samples were scanned using silicon nitride cantilevers in contact mode using a Multimode

AFM with a Nanoscope IIIa controller and an E scanner, calibrated using a silicon grid with 10×10 μm^2 pitch. All images were captured at the scan rate of 1 Hz, using 256×256 pixel resolutions with scan sizes from 50 nm to 15 μm and flattened to remove large degrees of background tilt. The images were analyzed using the WS×M 5.0 version developed by Nanotec Electronica [4].

3. Results and discussion

Typical AFM images of titanium films are shown in Fig. 1. The pre-irradiation titanium surface (Fig. 1(a)) was relatively smooth with small imperfections which can be readily distinguished from the defects on the Xe-irradiated surfaces (Fig. 1(b–d)). The black region in the upper left and the dislocation lines in the upper right in Fig. 1(a) are part of pre-existing crater and shallow furrow features, respectively. As the fluence increased, the areal density of voids, their size and depth, as well as void swelling all increased but the RMS surface roughness, which decreased slightly from 23 ± 1 nm to 20 ± 1 nm and then to 19 ± 2 nm (at a scan size of 15×15 μm^2, see Table 1).

A displacement cascade creates a number of vacancies and interstitial atoms, which can collapse in a dislocation loop.[5] The first dislocation loops become clearly visible after a fluence of 3.0×10^{13} Xe/cm^2, Fig. 1(c) They were found to be similar to elliptical loops seen on n-irradiated titanium, [6]. Increased fluences cause the size of dislocation loops to increase; however, the average b/a value of 0.55 measured for their mean short diameter (b) and mean long diameter (a) remains constant and independent of the size of loops and fluencies.

Dislocation lines appeared more frequently after 9.0×10^{13} Xe/cm^2, Fig. 1(d). It seems that this fluence (9.0×10^{13} Xe/cm^2) is about the value where significant changes in the microstructure of titanium start. Budzynski [7] reported interesting differences in topographies of titanium irradiated with Xe (130 MeV) and swift Kr (240 MeV) ions. The ion fluencies applied were 1×10^{14} Kr/cm^2 and 1×10^{14}, 3×10^{14}, 4.9×10^{14} Xe/cm^2 that are much larger than those in our experiments; the only comparable point of 1×10^{14} Xe/cm^2 is not really discussed by the authors. While their almost two fold-larger energy and lower fluence of the swift-Kr ions caused hillocks, the irradiation with less energetic Xe at larger fluence caused craters. For the fluences used in our experiments, we have observed similar, crown-localized hillocks and craters only in the case when the Ti-foil was coated with a thin layer of uranium.

Table 1
Summary of radiation changes on titanium surfaces.

Swelling and void characteristics					
Fluence [cm^{-2}]	Swelling [%]	Diameter [nm]	Depth [nm]	Areal density [voids/cm^2]	Surface roughness [nm]; at 15×15 μm^2
1.2×10^{13}	0.12 ± 0.02	190 ± 20	10 ± 1	$(5.78 \pm 0.21).10^6$	20 ± 1
3.0×10^{13}	0.32 ± 0.11	198 ± 17	13 ± 2	$(1.60 \pm 0.13).10^7$	19 ± 2
9.0×10^{13}	1.25 ± 0.19	210 ± 20	15 ± 2	$(5.15 \pm 0.44).10^8$	19 ± 2

Dislocation loops				
Fluence [cm^{-2}]	Areal density [cm^{-2}]	Mean short diameter (b) [nm]	Mean long diameter (a) [nm]	Depth [nm]
1.2×10^{13}	n/a	n/a	n/a	n/a
3.0×10^{13}	$(1.56 \pm 0.31) \times 10^6$	59 ± 6	107 ± 4	3.12 ± 0.64
9.0×10^{13}	$(2.00 \pm 0.53) \times 10^7$	300 ± 42	550 ± 26	19 ± 3

Dislocation lines			
Fluence [cm^{-2}]	Areal density [cm^{-2}]	Length [nm]	Depth [nm]
1.2×10^{13}	n/a	n/a	n/a
3.0×10^{13}	Rare	n/a	n/a
9.0×10^{13}	$(1.29 \pm 0.52) \times 10^7$	1400 ± 203	25 ± 2

4. Conclusion

Irradiation of Ti films with 132 MeV Xe^{+29} produced elliptical dislocation loops with the average short/long mean diameter ratio of 0.55 ± 0.01. With increasing the fluence, the size of voids, depth and void density as well as void swelling increased without significant change of the surface roughness values.

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