SURFACE DAMAGE IN TiO_2 AND Al_2O_3 INDUCED BY SWIFT HEAVY IONS

J. O'Connell[1], V.A. Skuratov[2, 3], G. Aralbayeva[4], M. Saifulin[2, 3] A. Dauletbekova[4], M. Zdorovets[5, 6]

[1]*Centre for HRTEM, Nelson Mandela University, Port Elizabeth, South Africa, jacques.O'Connell@nmmu.ac.za*

[2]*Flerov Laboratory for Nuclear Research, Joint Institute for Nuclear Research, Dubna, Russia*

[3]*National Research Nuclear University MEPhI, Moscow, Russia*

[4]*L.N. Gumilyov Eurasian National University, Astana, Kazakhstan*

[5]*Institute of Nuclear Physics, Astana, Kazakhstan*

[6]*Ural Federal University, Yekaterinburg, Russia*

In this paper we summarize results of AFM and TEM studies of surface defects in titanium and aluminum oxide single crystals irradiated with high energy Xe and Bi ions. The formation of nanoscale hillocks subtended by conically shaped low density zones has been observed over a wide range of electronic stopping powers and irradiation temperatures. Analysis of experimental data obtained from targets with thickness less than the projected ion range indicated that the hillocks are produced on both the entrance and exit surface of the specimen. HRTEM/STEM examination has revealed that the hillocks are crystalline in nature. The relationship between the latent track morphology in the near surface region and hillocks is discussed.

Introduction

Shaped as hillocks, surface nanoscale defects induced by single swift heavy ions (SHI) have been observed in a variety of alkali halide and oxide crystals using scanning microscopy techniques. In particular, atomic force microscopy (AFM) studies of some radiation resistant insulators, like Al_2O_3, showed that the mean hillock height on the sapphire surface depends almost linearly on the incident electronic stopping power and increases by a factor of two on average when hillocks start to overlap [1]. This phenomenon occurred at a relatively low fluence of 700 MeV bismuth ions of 10^{11} cm^{-2}. A subsequent increase of ion fluence to 10^{12} cm^{-2} results in a completely disordered surface layer with increased roughness [1]. This implies that the formation of defects such as hillocks, may significantly affect the long term radiation stability of ceramic nuclear fuel due to the large number of fission track recoils crossing the surface of fuel pellets.

Although the AFM itself provides very important information about radiation induced changes in surface relief, the most efficient way to study SHI induced surface radiation damage is to utilize this method in combination with high resolution transmission electron microscopy (TEM). First of all, it is necessary to answer questions concerning the connection of surface hillocks and latent track morphology in the sub-surface region. Another topic that should be investigated is the comparison of surface defects on the entrance and exit surface of SHI irradiated ceramic specimens. This is of considerable practical value for simulation of fission fragment impacts since the typical accelerator-based experiments involve irradiation of an entrance surface, while fission fragments escape from fuel leading to exit hillocks on the surface. Currently, very limited information is available in the literature concerning TEM studies of surface hillocks [2, 3].

The majority of experimental data on surface damage as well as latent tracks in radiation resistant insulators have been acquired from room temperature irradiation studies. This is in stark contrast to actual reactor temperature ranges. Therefore, data from specimens irradiated over a wide temperature range is of interest for both practical applications and the development of realistic physical models of the mechanisms responsible for the creation of hillocks and latent track formation.

Experimental

Sapphire and rutile single crystals purchased from MTI Corporation were used as target materials. The samples were irradiated at the IC-100 and MC400 cyclotrons at the FLNR JINR, in Dubna and the DC-60 cyclotron in Astana at energies ranging from 1.2 to 11 MeV/nucleon. Irradiation temperatures included room temperature, 300°C, 500°C and 700°C. TEM/STEM analysis was performed using a double Cs corrected JEOL ARM 200F operating at 200 kV. TEM specimen preparation was done using an FEI Helios Nanolab 650 with final polishing performed at 1 keV Ga energy. Crystal surfaces were imaged using a SmartSPM 1000 scanning probe microscope.

Results and Discussion

Figure 1 shows a typical AFM micrograph of surface hillocks on a TiO_2 crystal after irradiation with 220 MeV Xe ions at a temperature of 300°C. Although the individual hillocks are clearly visible, their shapes are distorted due to the physical size of the scanning tip. Information regarding the crystallinity of the hillocks is also not available.

Figure 2 shows an HRTEM micrograph of two closely spaced hillocks on the surface of a TiO_2 crystal after irradiation with 220 MeV Xe at 700°C. In this case the crystal structure of the hillocks is clearly visible although the bulk crystal exhibits a different contrast due to the relative height and thickness differences between the bulk and hillock.

Figure 3 shows an HAADF and ABF STEM micrograph pair of the same hillock structures as in figure 2. In both cases the atomic columns are clearly visible in both the bulk crystal and the hillocks. Some defects can be seen running through the hillocks and into the bulk crystal.

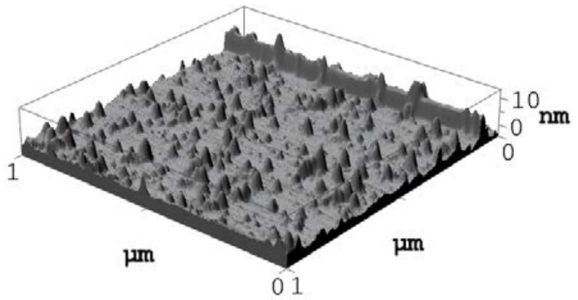

Fig. 1. AFM micrograph of the surface of a TiO_2 crystal irradiated with 220 MeV Xe at a temperature of 300°C

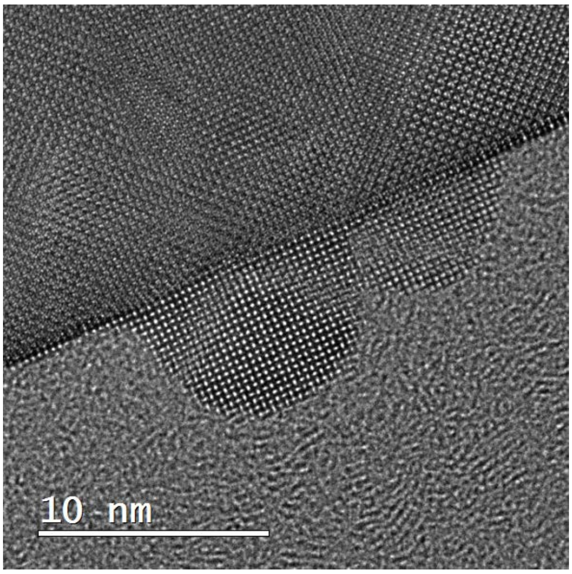

Fig. 2. HRTEM micrograph of the surface of a TiO_2 crystal irradiated with 220 MeV Xe at a temperature of 700°C. The crystallinity of the hillocks is clearly evident

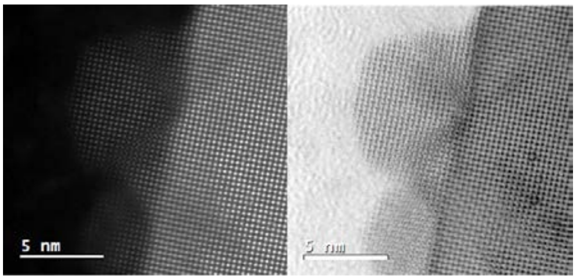

Fig. 3. HAADF (left) and ABF (right) micrographs of the same hillocks shown in figure 2

Figure 4 shows a BF TEM micrograph of the surface of a 1 GeV Bi irradiated TiO_2 crystal. Below each hillock is a conical zone with a relatively low density with respect to that of the surrounding crystal. Beyond this zone the latent tracks appear as discontinuous lines of defects in the crystal.

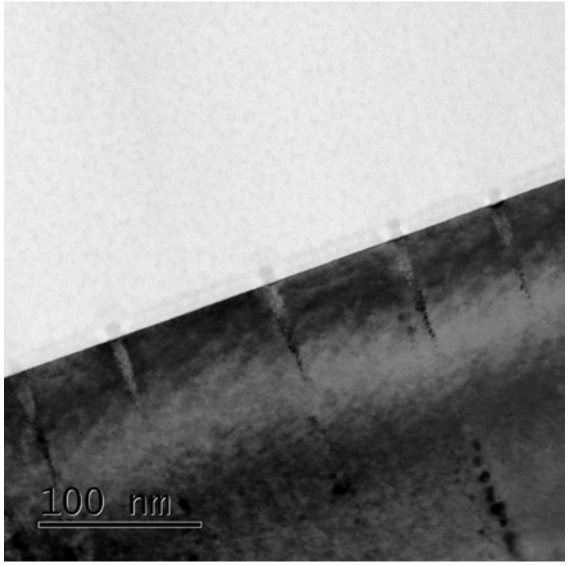

Fig. 4. BF TEM micrograph of surface hillocks on TiO2 irradiated with 1GeV Bi ions at room temperature. Conically shaped low density zones can be observed below each hillock

Conclusion

HRTEM/STEM is a valuable tool for investigation of the morphology of SHI induced latent tracks and surface hillocks. It was found that surface hillocks on TiO_2 and Al_2O_3 are crystalline and epitaxial to the bulk crystal. Below each hillock is a conically shaped zone of lower density than the surrounding bulk crystal. It is assumed that the lost material in these zones form the surface hillock upon expulsion from the bulk. Beyond the conical zone the latent tracks appear as discontinuous lines of defected crystal. There appears to be no difference in morphology between hillock on the entrance and exit surface of a crystal. Irradiation at elevated temperatures produces larger hillocks which is consistent with the prediction of the thermal spike model.

References

1. *Skuratov V.A.*, *et al.* // Surface and Coating Technology. 2006. V. 196. № 1-3. P. 56.
2. *Ishikawa N., Okubu N., and Taguchi T.* // Nanotechnology. 2015. V. 26. P. 35701.
3. *Jacques O'Connell, et al.* // Phys. Status Solidi B. 2016. V. 253. № 11. P. 2144.