THERMAL ANNEALING AND SIMULATION OF FISSION FRAGMENT SELF-IRRADIATION OF BADDELEYITE

Michael Lee[1)], Jacques O'Connell[1)] and Vladimir Skuratov[2)]
[1)]*Centre for HRTEM, Nelson Mandela University, University Way, Port Elizabeth South Africa, michael.lee@nmmu.ac.za, jacques.O'Connell@nmmu.ac.za*
[2)]*Flerov Laboratory for Nuclear Research, Joint Institute for Nuclear Research, Dubna, Russia, skuratov@jinr.ru*

The discovery of loop type defects along straight lines originating from uranium inclusions in natural zirconia (baddeleyite) was the first evidence for radiation tracks in this material. The tracks were assumed to be a result of U fission fragments. The physical processes such as change in pressure and temperature before and after the volcanic emplacement process was unknown. However, the crystalline transition from tetragonal to monoclinic in the range 700 -1100 C is well known. It was therefore necessary to use the same material and expose it to 167Mev Xe ions to simulate the U fission fragment tracks. The as-irradiated bulk material was subsequently annealed at 800 C and thin sections were vacuum heated in-situ up to 500C. The microstructure of the material, before and after annealing was investigated by transmission electron microscopy (TEM). The results showed that the loop like structures along the fission tracks could be produced after annealing. The application of high resolution TEM demonstrated the presence of a localized tetragonal phase transformation, which was quenched into the ion track material being surrounded by monoclinic bulk material (the stable isomorph at room temperature).

Introduction

Previous reports [1] have commented on the lack of a detailed microstructural analysis for natural zirconia (baddeleyite) which would possibly provide information on the nature of primary and secondary physical alteration mechanisms, and hence the geothermal history of the material. It was previously assumed that uranium, which is used for geological dating, is in solid solution with the zirconia. However, it has been shown that the uranium oxide is not in solid solution and forms inclusions within the zirconia matrix [2]. Considering the age of the material it should be expected that there should be evidence for radiation damage in the zirconia from the uranium oxide inclusion. It was found that zirconia found in the presence of natural radioactive uranopyrochlore deposits did not reveal any thermal or radiation induced structural damage [1,3]. However, we previously demonstrated the existence of straight lines of defects originating from UO_2 inclusions in natural zirconia [4], which are believed to be due to recoil ions from uranium fissions. In this work we firstly simulated such fission fragment impact through swift heavy ion irradiation followed by thermal annealing (bulk and in situ) by comparing the microstructure of resulting latent tracks to those shown previously. Secondly, the presence of quenched high temperature isomorphs (tetragonal or cubic) in the proximity of the ion tracks was investigated.

Experimental

A thin section (~2mm) of a Phalaborwa baddeleyite xenocryst was irradiated with 167 MeV Xe ions at the FLNR cyclotron complex in Dubna, Russia to a fluence of $2{\cdot}10^{10}$ cm^{-2}. These ions agree well in mass and energy to U fission fragments. Irradiation was performed at room temperature and TEM lamellae were prepared in cross section and plan view geometry using an FEI Helios Nanolab FIB. These lamellae were examined using a double Cs corrected ARM 200F operating at 200 kV in both TEM and STEM mode. Two methods used for thermal annealing were used: (1) An as-irradiated bulk sample was annealed in a quartz tube furnace for 20 minutes at 800°C under argon flow (2) a TEM lamella containing as-irradiated material was annealed in situ under vacuum in the TEM. The in situ heating of the sample was performed in the TEM using a Dens Solutions Wildfire holder.

Results and Discussion

Figure 1 shows a BF TEM micrograph of straight lines of loop-like defects extending into a ZrO_2 crystal from a UO_2 inclusion in as-received material. It is assumed that these defects were produced by fission recoils originating from the UO_2.

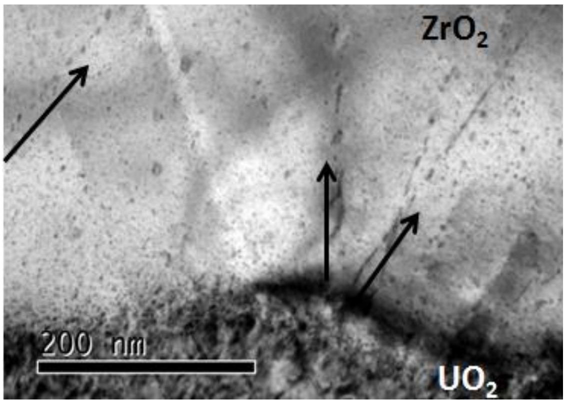

Fig. 1. Lines of loop-like defects originating from A UO_2 inclusion in natural ZrO_2

Figure 2 shows two BF TEM micrographs of latent tracks in the same material in cross-section (a) and plan view (b). Individual tracks have a rectangular cross section and consist of defects aligned at a slight angle to the incident radiation. The high magnification BF TEM micrograph of the latent track shown in figure 3 demonstrates the discontinuity of the tetragonal tracks. This is due to the a-c mismatch between the tetragonal and monoclinic phase.

In figure 4 the HAADF (a) and BF (b) STEM micrographs are shown for of one such latent track along the ion direction. The track core consists of a rectangular (in cross section) region where a phase transformation has occurred. Figure 5 shows similar

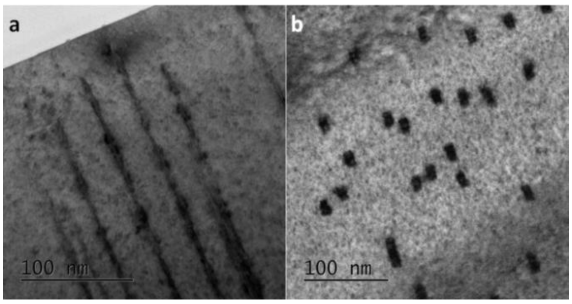

Fig. 2. BF TEM micrographs in cross-section (a) and plan view (b) of 167 MeV Xe latent tracks in natural ZrO_2

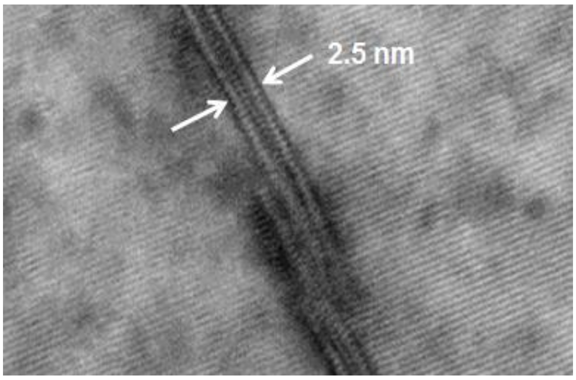

Fig. 3. BF TEM micrograph showing magnified view of a latent track in ZrO_2 as shown in figure 2(a)

structures as in figure 1 after annealing at 800°C for 20 min.

Conclusion

Latent track geometry in the natural and simulated systems was found to be distinctly different with the simulated system exhibiting more continuous tracks with a larger diameter. The simulated tracks were also rectangular in cross-section unlike the loop-like structures found in the natural system.

The discrepancy is believed to be due to the thermal history of the natural system. Room temperature irradiation is known to quench in defect

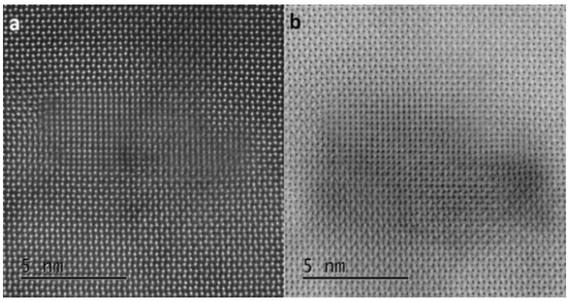

Fig. 4. BF TEM micrograph showing magnified view of a latent track as shown in figure 2(a)

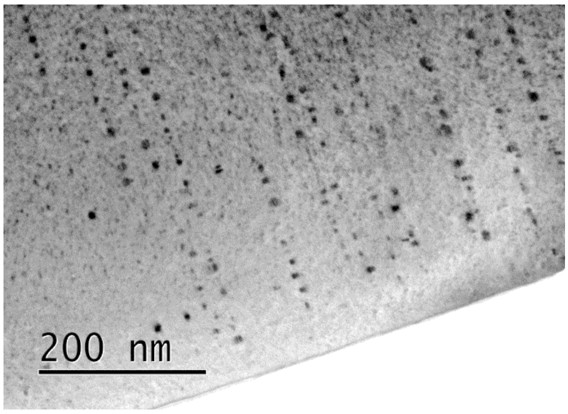

Fig. 5. BF TEM micrograph of latent tracks after annealing at 800°C

structures that will at least partially anneal out during subsequent heat treatment as shown in the annealed specimens. Post annealing the ion track morphology of the simulated system closely resembled that of the natural system.

References

1. *Lumpkin G.R.* // J. Nucl. Mater.1999. V. 274. P. 206.
2. *Lee M.E., O'Connell J.H. and Goosen W.E.* // Proc. Microsc. Soc. South. Afr. V. 45. P. 43.
3. *Degueldre D. and Hellwig C.H.* // J. Nucl. Mater. 2003. V. 320. P. 96.
4. *Lee M.E., O'Connell J.H and Janse van Vuuren A.* // Proc. Microsc. Soc. South. Afr. 2015. V. 45. P. 43.