Evidence for thermal alteration of fission tracks in Phalaborwa baddeleyite

Lee, M.[1], O'Connell, J.[1] and Skuratov, V.[2]

[1] Nelson Mandela University, South Africa, [2] Joint Institute for Nuclear Research, Russian Federation

Large deposits of baddeleyite (ZrO_2) were found in the Phalaborwa complex in South Africa, found mainly in the foskorite ore zone and to a lesser extent in the carbonatite ore body [1]. Previous reports [2] have commented on the lack of a detailed microstructural analysis for baddeleyite which would possibly provide information on the nature of radiation tracks in baddeleyite. It was previously assumed that uranium was 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 [3]. 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 [2,4]. In this paper we will show the existence of straight lines of defect loops originating from UO_2 inclusions in natural zirconia. These defects are believed to be due to recoil ions from uranium fissions and subsequently thermally annealed. This is demonstrated through the simulation of such fission fragment impact through swift heavy ion irradiation and in situ annealing. The resultant tracks are then compared to those previously shown for tracks in as-received baddeleyite.

A Phalaborwa baddeleyite xenocryst was cut into sections and polished. A section was irradiated with 167 MeV Xe ions to a fluence of 2×10^{10} cm^{-2}. These ions agree well in mass and energy to U fission fragments. Irradiation was performed at room temperature. TEM lamellae were prepared for as received material and irradiated material in cross section using an FEI Helios Nanolab FIB. The lamellae were examined using a double Cs corrected ARM 200F operating at 200 kV in both TEM and STEM mode. The irradiated samples were annealed in situ using a Dens^{TM} Wildfire heating holder. 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. Figure 2 shows a BF TEM micrograph for Xe latent tracks in the same material. Individual tracks have a rectangular cross section (approximately 2.5 nm) and consists of defects aligned at a slight angle to the incident radiation as shown by the high magnification BF TEM micrograph in figure 3. The BF TEM micrograph in figure 4 shows the defects loops in the in situ annealed sample which are similar to those observed in the as-received material shown in figure 1. It is therefore concluded from the radiation simulation results that there are indeed radiation tracks in baddeleyite which have been thermally annealed.

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4. Degueldre, D. and Hellwig, Ch. (2003) J. Nucl. Mater. 320, 96.

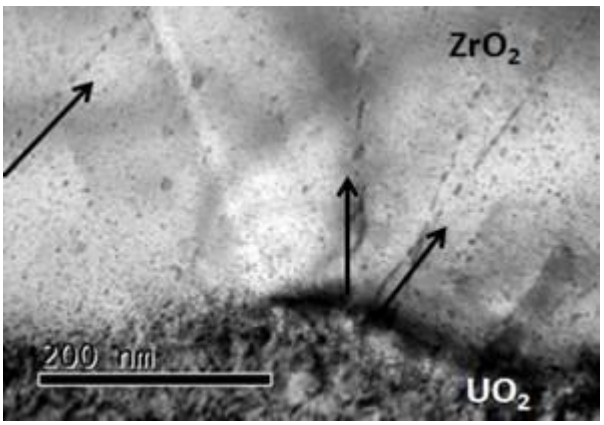

Fig 1. BF TEM micrograph of as received material showing the lines of defect loops in ZrO_2 originating from the UO_2 inclusion

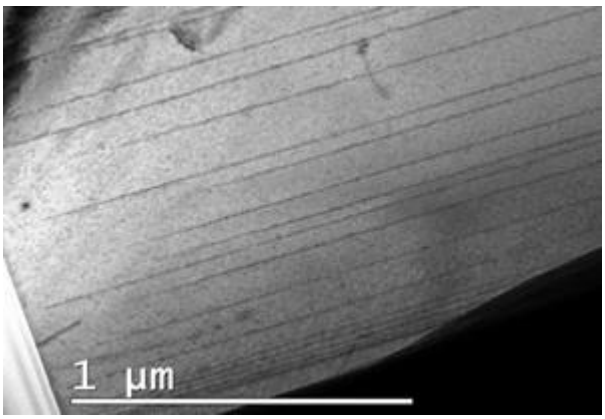

Fig 2. BF TEM micrograph showing latent tracks in the Xe irradiated material

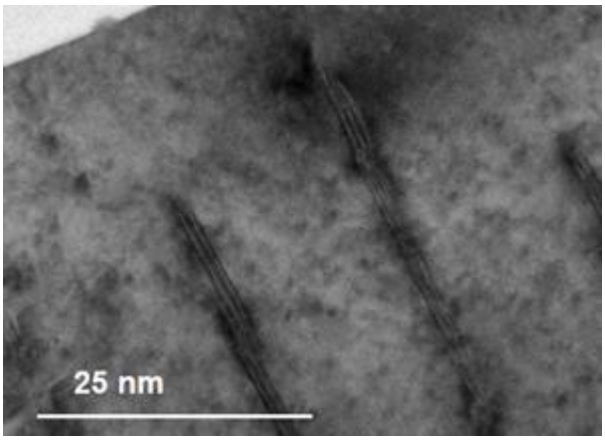

Fig 3. BF TEM micrograph showing a rectangular cross section for the latent tracks in the irradiated material.

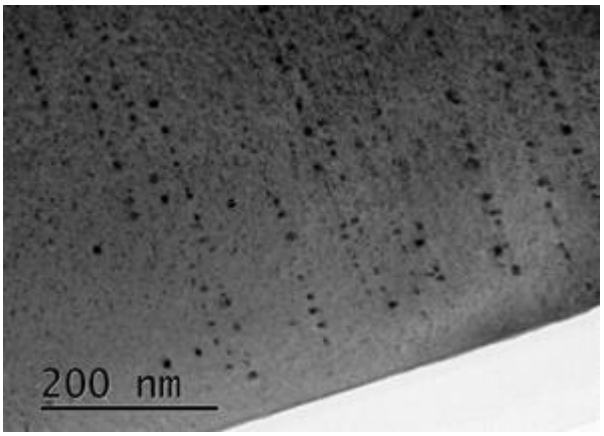

Fig 4. BF TEM micrograph showing lines of defect loops in the irradiated zirconia after annealing at 500 °C.