TEM OBSERVATION OF SHI TRACK INTERACTION AND INDUCED STRAIN IN ZIRCONIA AT INTERMEDIATE FLUENCES

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The SHI irradiation of m-zirconia at low fluences in the non-overlapping regime has been shown to produce single ion tracks containing tetragonal (t-zirconia) segments by direct observation by imaging in a transmission electron microscope (TEM). The stability at RT of the high temperature t-zirconia phase is a function of a numerous thermodynamical parameters which include particle size and shape, interfacial energy, oxygen vacancies and strain energy. The absence of strain at low SHI fluence indicates the possibility the presence of oxygen vacancies to be the predominant factor for RT stabilization. However, the increase of fluence into the intermediate range, up to the onset of the overlapping regime, corresponds to the increased strain in the material between the tracks as shown by TEM imaging. At a fluence of 1 x 10^{13} cm^{-2} the tracks are no longer visible due to the excessive strain in the material. Regions of different phases are observed which possibly indicate the onset of phase evolution at this fluence. It is therefore proposed that the stabilizing factor for non-overlapping fluences is due to the presence of oxygen vacancies. However, with increasing fluence into the overlapping regime these oxygen vacancies are possibly annealed out with the predominant factor for the stabilization of the t-zirconia phase being due to the increased strain in the m-zirconia.

Keywords: SHI; track; overlap; zirconia; TEM; strain.

Introduction

Several authors have reported on the SHI irradiation studies for monoclinic zirconia and have shown that the subsequent transformation to the tetragonal phase (m→t) was stabilized at room temperature (RT) after exposure to suitable irradiation fluences[1-3]. This tetragonal transformation has been quantitatively demonstrated using techniques such as Raman spectroscopy and X-ray diffraction (XRD) for SHI irradiation fluences down to 2 x 10^{12} for heavy ions and 5 x 10^{13} cm^{-2} for light ions[4]. The "apparent" fluence threshold for the m→t transformation is probably due to the detection limit (~ 1-2 vol%) for the phase fraction determined by the Raman and XRD spectroscopic techniques. However, O'Connell *et al* reported on the direct observation of individual tetragonal ion tracks formed in monoclinic zirconia (ZrO_2) as a result of 167 MeV Xe irradiation (fluence 2 x $10^{10}$$cm^{-2}$) at room temperature[5].

In this work we report on the direct observation, by transmission electron microscopy (TEM), of the response by monoclinic zirconia to ion irradiation at intermediate fluences covering both the single track and track overlapping regimes.

Experimental

Sections of single crystalline m-zirconia were irradiated with 150 MeV Xe ions at a fluence in the range 4 × 10^{10} – 1 x 10^{13} cm^{-2} at room temperature. To simulate the effect of track overlap, the resultant interaction of tracks for the m-zirconia specimen irradiated from two different opposite directions both at 45º to the surface (each at a fluence of 2 x 10^{10} cm^{-2}) is shown schematically in figure 1. Electron transparent lamellae were prepared by a FIB lift out technique using an FEI Helios NanoLab 650 FIBSEM and analysed using a double Cs corrected JEOL ARM200F operated at 200 kV.

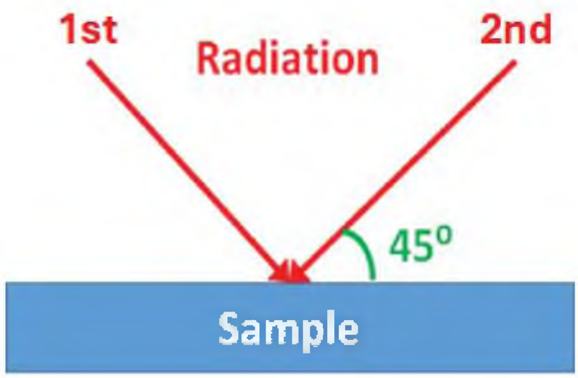

Fig. 1. Schematic of zirconia specimen irradiated from two different directions at 45º to the surface

Results and discussion

TEM imaging (Fig. 2) of single ion tracks produced in m-zirconia irradiated by Xe ions at a fluence of 2 x 10^{10}cm^{-2} are shown to produce tetragonal segments aligned at 9^0 along the track and each interfaced at a junction. The localized strain field appears to be restricted to the region surrounding the junctions without any visible strain in the rest of the surrounding material. The simulation of track overlap annealing effect, is illustrated by the resultant interaction of tracks for the m-zirconia specimen irradiated from two different opposite directions, both at 45^0 to the surface (each at a fluence of 2 x 10^{10} cm^{-2}) are observed by TEM imaging (Fig. 3). It was noted that the tracks from the second dose are more continuous which demonstrates the annealing effect of the second dose on the first dose of ions. Based on the observed saturation track density, each track should have an effective annealing radius of around 6-7 nm. This corresponds to a fluence of approximately 1.5×10^{12} cm^{-2} for the onset of significant track overlap.

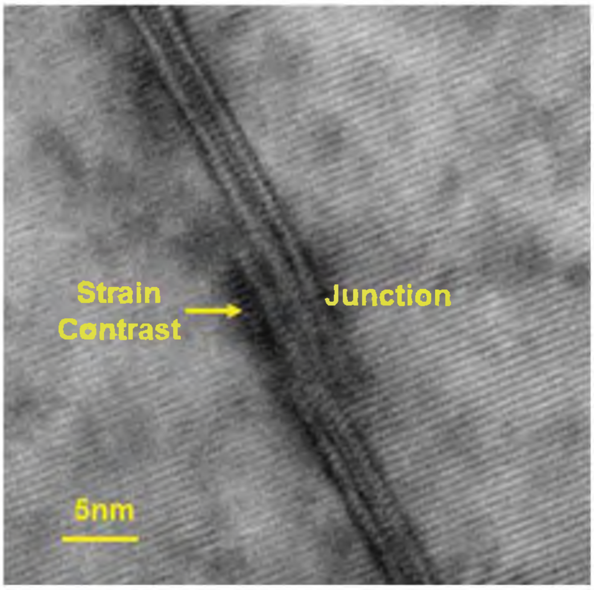

Fig. 2. TEM BF image of strain at the junction between two t-zirconia segments in a single ion track

Radiation of the material with increased fluences from 5×10^{11} cm^{-2} to 1×10^{13} cm^{-2} corresponds to the observation of increased material strain and the tetragonal tracks being difficult to view at exact image focus. At the onset of the track overlap regime, high resolution cross-sectional TEM image (Fig. 4) of zirconia irradiated at 1 x 10^{13} cm^{-2} reveals highly strained material resulting in the tracks not being visible at focus.

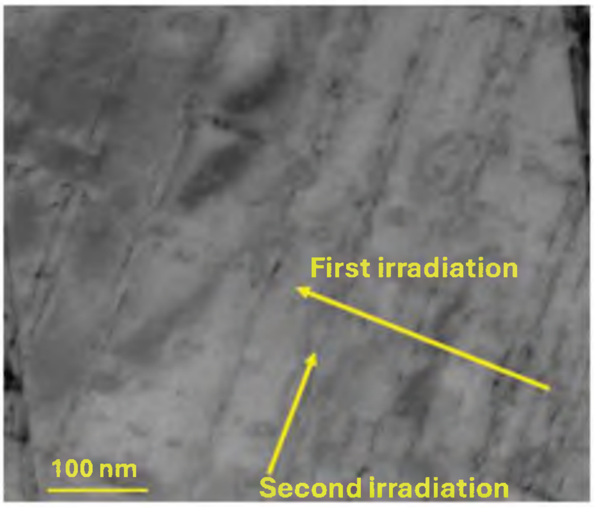

Fig. 3. TEM BF image of overlapping track simulation

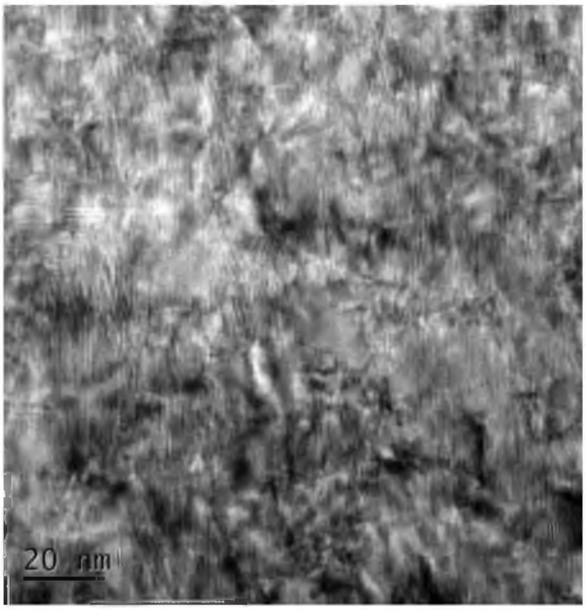

Fig. 4. High resolution cross-sectional TEM image of zirconia irradiated at 1 x 10^{13} cm^{-2} (Zone axis $[100]_m$)

However, detailed inspection, at higher magnification, of the strained areas shown in figure 4 reveal three different regions observed by HRTEM imaging and the FFT in figure 5(b) corresponding to the monoclinic $[100]_m$ zone axis. The FFT for the regions shown in figure 5(a) and (c) reveal missing reflections with respect to (b) which is an indication of a non-homogeneous structural evolution. This is possibly due to an intermediate phase, resulting a highly strained sub-lattice, before the t-zirconia phase fraction becomes detectable by other techniques such as XRD and Raman spectroscopy.

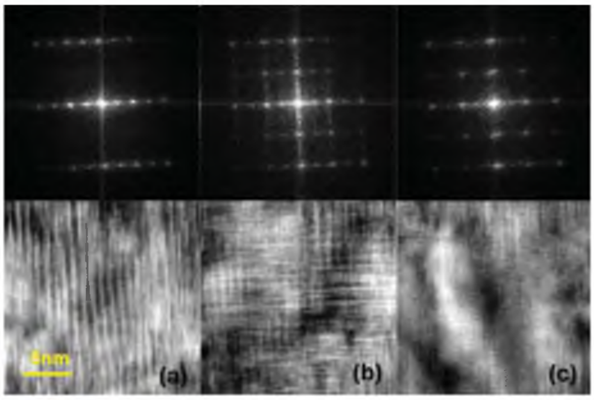

Fig. 5. HRTEM images and corresponding FFT for three different phases observed in the zirconia for a fluence of 1×10^{13} cm^{-2}

Conclusions

Direct observation of the track interaction by TEM suggests a track diameter of the order 6-7 nm. This corresponds to a fluence of approximately 1.5×10^{12} cm^{-2} for the onset of significant track overlap. Material strain increases with fluence until the tracks are no longer visible at 1×10^{13} cm^{-2}. This corresponds to the observation of a non-homogeneous structural evolution of the material. The increased strain in the m-zirconia at intermediate fluences could possibly be the dominant factor for the RT stabilization of the t-zirconia phase compared to the oxygen vacancy stabilizing and minimal strain at low fluence. Future investigations using XRD and Raman to determine the strain in the irradiated m-zirconia may assist in understanding the phase evolution of the material.

References

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