

Effects of Fission Product Accumulation in Cubic Zirconia*

The disposition and disposal of plutonium from the dismantlement of nuclear weapons and from the reprocessing of the commercial nuclear fuel have led to increased interest in the possibility of "burning" actinides in non-fertile or so-called inert-matrix fuels (e.g., fuels without ^{238}U).^{1,2} This strategy prevents the further production of plutonium by neutron capture reactions and subsequent beta decay. The recently adopted strategy in selecting materials for inert matrix fuel has been to consider not only the properties of the material as a fuel, but also the waste form properties of the inert-matrix fuel. Such an approach would allow direct disposal without reprocessing after once-through burnup.

Yttria stabilized cubic-zirconia (YSZ) is a promising candidate material for both inert matrix fuel and waste form based on its high solubility for actinides, high chemical durability and its reported exceptional stability under radiation.^{3,4} Because the incorporation of fission and other transmutation products during burnup may significantly affect the radiation response and the chemical durability of cubic zirconia, the solubility and mobility of the fission product nuclides in the inert matrix fuel at high temperatures (reactor fuel conditions) and low temperatures (repository conditions) are important.

In this study, we have investigated the effects of fission product incorporation on the microstructure of YSZ (with 9.5 mol. % of yttria) by ion implantation (using 70-400 keV Cs^+ , Xe^+ , Sr^+ and I^+ ions) and transmission electron microscopy (TEM). The ion implantation was conducted in a temperature range between 300 to 873 K to doses up to 1×10^{21} ions/ m^2 . *In situ* TEM was conducted on pre-thinned TEM samples to follow the microstructure evolution during ion implantation using the IVEM-Tandem Facility at Argonne National Laboratory. Cross-sectional TEM was performed after implantation of the bulk samples to reveal the depth-dependent microstructure induced by ion implantation.

In situ TEM during the 70 keV Cs^+ implantation at the room temperature revealed a high density of defect clusters on the nanometer scale after $\sim 2 \times 10^{20}$ Cs/m^2 . The defect clusters with characteristics of interstitial type dislocation loops are interpreted to be the result of planar precipitates of Zr and/or O interstitials displaced from their original lattice site by the collisional events. Amorphous domains in thin regions of the specimen were observed after 1×10^{21} Cs/m^2 with high resolution TEM (HRTEM) and nanobeam electron diffraction (Fig. 1).

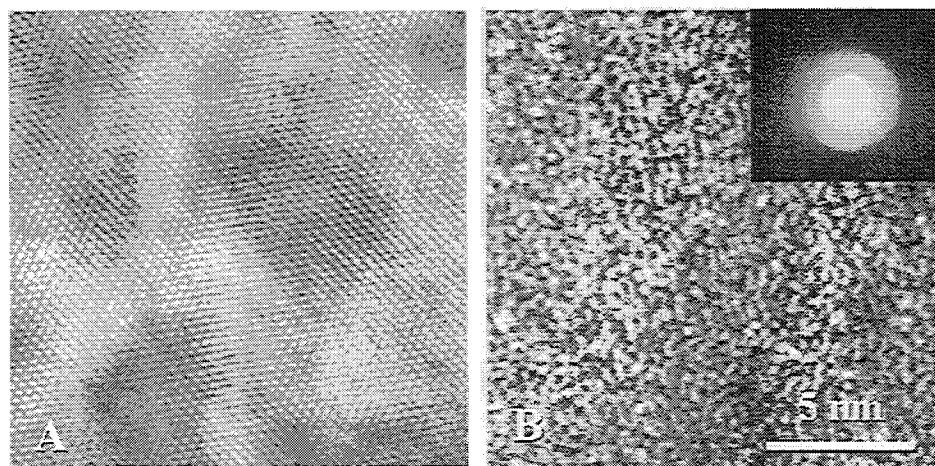
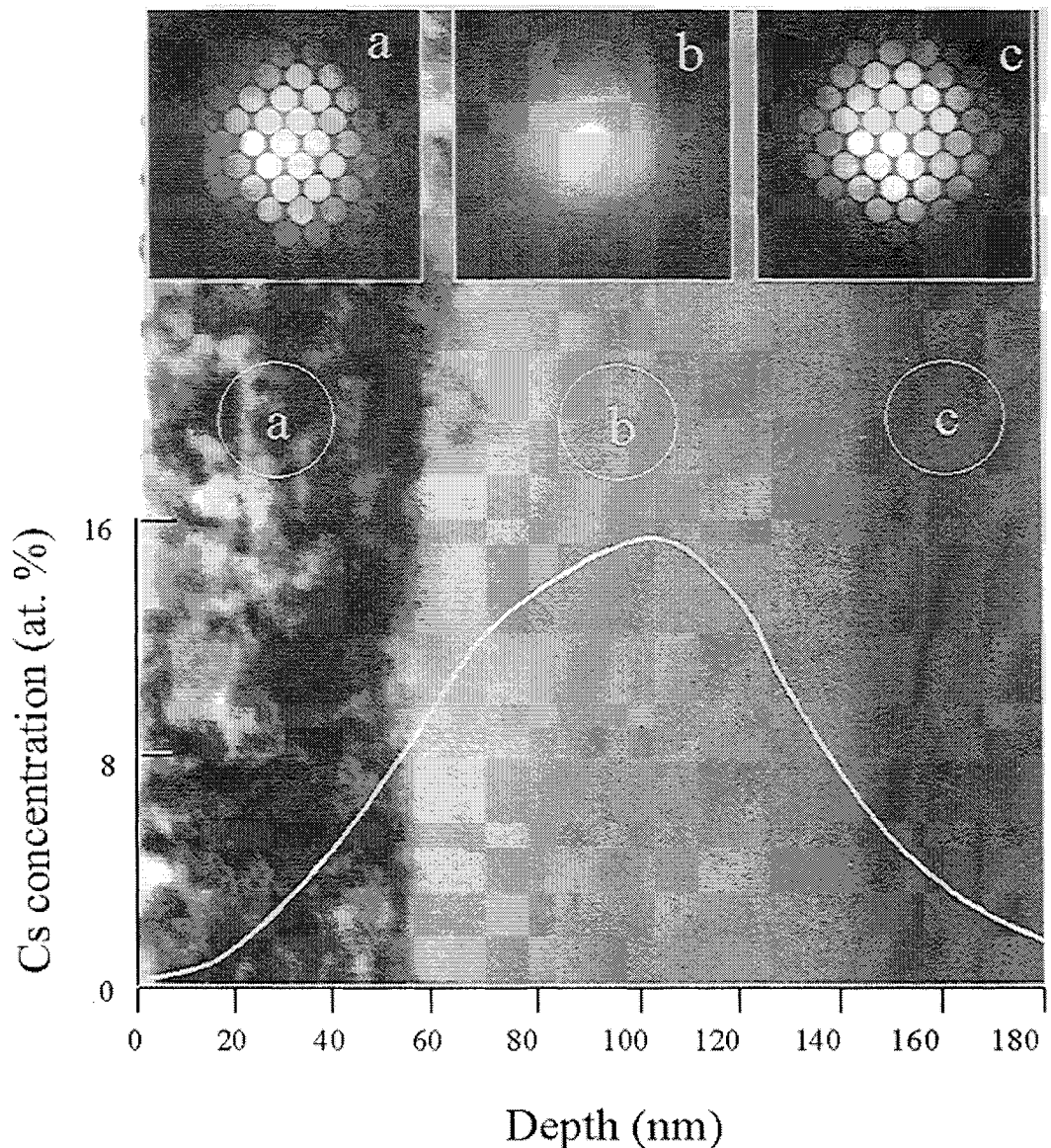


Figure 1. Plan-view high resolution TEM micrographs from various regions of a cubic zirconia sample (stabilized by 9.5 mol. % of yttria) after 70 keV Cs^+ implantation to 1×10^{21} Cs/m^2 at room temperature. (A) from a region with ~7 at.% Cs; (B) from a region with ~11 at.% Cs.

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Cross-sectional TEM of a specimen after 400 keV Cs⁺ implantation to 1x10²¹ Cs/m² at the room temperature has revealed an amorphous band in a depth range where Cs concentration is greater than 8 at.% (Fig. 2). Although the front edge of the amorphous layer overlaps with the displacement damage peak that reached 330 displacement per atom (dpa), we suggest that the amorphization is mainly due to the incorporation of Cs rather than from the displacement damage as the center of the amorphous layer overlaps with the peak Cs concentration. This interpretation is consistent with previous results of radiation damage studies in YSZ that reached damage levels as high as 680 dpa but without amorphization.³ Amorphization of YSZ is caused by the large size incompatibility and low mobility of cesium ions in the YSZ structure at room temperature, reflecting a relatively low solubility of Cs in YSZ. Nevertheless, the Cs concentration at which amorphization of YSZ occurred (~8 at. %) is well above the value that will likely be reached in an inert fuel matrix (~5 at. % assuming a 30 at.% Pu loading).

Figure 2. Cross-sectional bright-field TEM micrograph with associated electron diffraction pattern showing the formation of an amorphous band in a cubic zirconia sample (stabilized by 9.5 mol. % of yttria) after 70 keV Cs⁺ implantation to 1x10²¹ Cs/m² at room temperature. The overlay of the Cs concentration profile is based on the results of analytical TEM that has been normalized by the results of a Monte Carlo computer simulation using the SRIM code.



Various types of nanometer-scaled defect clusters (e.g., dislocation loops or small gas bubbles) were apparent in samples implanted by the other three ions after 1×10^{20} ions/m². However, other than Xe bubbles, no secondary phase precipitates were apparent in YSZ implanted with Xe, Sr or I ions even after 1×10^{21} ions/m². No amorphization was observed after 400 keV I⁺ implantation to 1×10^{21} ions/m² at 973 K, even though iodine has a larger ionic radius than cesium, due to the relatively high mobility of iodine in YSZ at the high temperature.

References

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