HRTEM STUDY OF ION BEAM IRRADIATION INDUCED AMORPHIZATION IN CERAMIC MATERIALS

L.M. Wang†, R.C. Ewing† and W.J. Weber*

†Department of Earth and Planetary Sciences, University of New Mexico, Albuquerque, NM 87131
*Materials Science Department, Pacific Northwest Laboratory, Richland, WA 99352

Radiation damage of nuclear materials (e.g. fast- or fusion-neutron damage in reactor structural components, fission-fragment damage in nuclear fuels and alpha decay damage in nuclear waste forms) has been one of the major challenges faced by the material science community. Ion beam irradiation and implantation experiments have been used extensively in the past few decades not only for simulating these damage processes in materials but also for improving material properties for many technological applications.

As an energetic particle traverses a crystalline target, it loses its energy predominantly through electronic (ionization) and nuclear (elastic collision) interactions with the atoms in the structure. The target atom which receives sufficient energy from the interactions may get displaced from its lattice site and may further displace other target atoms, creating a displacement cascade which is usually a few nanometers in size. After cascade formation, which lasts for only a few tenths of a pico-second, a displacement cascade consists of a very dense cluster of point defects and the region is considered to be amorphous. However, the type and amount of the radiation damage observable after the irradiation depend strongly on the material's ability to recover during the subsequent relaxation and annealing. While self-ion irradiation of simple metallic materials often results in void and dislocation loop formation because of the high mobility of point defects, heavy ion irradiation of intermetallic compounds, covalently bonded semiconductors and complex ceramic materials with mixed bond types is more likely to induce solid-state amorphization†. The amorphization may either occur directly within the displacement cascade (the entire cascade is quenched) or by point defect accumulation (only isolated point defects survive the cooling phase of each cascade), as well as by chemical disordering. In this study, the effects of ion beam irradiation in Ca$_2$La$_8$(SiO$_4$)$_6$O$_2$, SiC, UO$_2$ and other complex ceramic materials are studied with HRTEM and compared at the nanometer scale.

Most HRTEM images taken from 1 to 1.5 MeV Ar$^+$, Kr$^+$ and Xe$^+$ ion irradiated (below 498K) Ca$_2$La$_8$(SiO$_4$)$_6$O$_2$ shows nanometer scale discrete domains which are believed to be the images of the displacement cascades. Computer processing of the image (Fig. 1) indicates that amorphization occurs directly within the displacement cascade in Ca$_2$La$_8$(SiO$_4$)$_6$O$_2$. This can be expected because it is easy to understand that such a complex crystal structure (apatite, $P6_3/m$) is hard to be reconstructed once it is heavily damaged (all the different atoms have to find the unique structural sites). The fact that the size of the cascade image increases with the increasing ion mass and decreases with the increasing temperature agree well with the critical amorphization dose—temperature curves determined by in situ electron diffraction observations during ion irradiation. Although SiC can also be fully amorphized under 1.5 MeV Xe$^+$ ions at room temperature and the HRTEM images taken below the critical dose show mottled contrast (Fig. 2), the lack of the discrete amorphous cascade domains in the image indicates that amorphization occurs by point defect build up in β-SiC. UO$_2$ has never been amorphized and the HRTEM image of UO$_2$ contains no visible damage even after a very high ion dose at near liquid helium temperature (20K) (Fig. 3). The stability or the high damage recovering rate of UO$_2$ must be related to its closest packing and high symmetry nature.

References

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FIG. 1.—(a): Digital HRTEM image of a Ca$_2$La$_8$(SiO$_4$)$_6$O$_2$ crystal containing a collision cascade damage created by 1 MeV Ar$^+$ ion irradiation at room temperature; (b): Fourier transform of (a) indicating the image contains a periodic and an aperiodic component; (c): contributions of the periodic component in (b) have been filtered out in Fourier space; (d): inverse Fourier transform of (c) showing the aperiodic (amorphous) component in the original image (a); (e): intensity distribution across the line in (d) showing the cascade damage in the third dimension; (f): the periodic (crystalline) component in the original image (a) obtained by subtracting (d) from (a) pixel by pixel. The discrete nature of the cascade image and the result of the computer processing indicate that amorphization in Ca$_2$La$_8$(SiO$_4$)$_6$O$_2$ occurs directly with the displacement cascade.

FIG. 2.—HRTEM image of β-SiC irradiated at room temperature with 1.5 MeV Xe$^+$ to 8.5x10$^{13}$ ions/cm$^2$. The mottled contrast and lack of discrete amorphous regions in this image indicate that amorphization occurs by point defect accumulation in β-SiC.

FIG. 3.—HRTEM image of UO$_2$ irradiated at 20K with 1.5 MeV Xe$^+$ to 1x10$^{16}$ ions/cm$^2$. Irradiation damage was not observed even under extreme conditions (low temperature and high ion dose) indicating that cascade damage is easily recovered in UO$_2$. 