TEM studies of ion-irradiation tolerance in Gd_2TiO_5

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Abstract: Gd2TiO5 has been identified as a potentially useful material for nuclear based applications such as burnable poison or constituent in waste-forms. A polycrystalline, single phase sample of Gd_2TiO_5 has been fabricated via the oxide-route reaction. The single phase status and chemical stoichiometry were confirmed using SEM backscattered images, EDS, and laboratory XRD. The structure was confirmed to be orthorhombic, *Pnma,* space group number 62. The suitability of use for this material in a radioactive environment was tested by *in-situ* measurement of the materials resistance to transformation from the crystalline to amorphous state via 1MeV Kr^{2+} ion-irradiation. The *in-situ* ion-irradiation was carried out using the intermediate voltage electron microscope (IVEM)-Tandem facility at Argonne National Laboratory. The critical dose, Dc, required to render the Gd_2TiO_5 completely amorphous was determined and the result is compared with other similarly tested materials. Further bulk gold ion-irradiation at various accelerating energies was carried out at the Australian National University using the TANDEM accelerator with damage penetration depth characterised using cross-sectional TEM.

Introduction: Heavy ion beam irradiation is now routinely used as a method for simulating recoil damage caused by alpha-decay in actinide containing materials [1] and neutron damage within fission and fusion reactor systems.

High dose irradiation may lead to phase changes and as damage accumulates, certain materials may undergo a crystalline to amorphous phase transformation. By observing the transition from crystalline to amorphous with increasing ion irradiation dose, the critical dose of amorphisation (D_c) can be established typically via x-ray and electron diffraction methods.

Atomic- and/or microscopic-scale changes in structure lead to larger scale effects in the material properties such as an increase in volume (swelling) linked to the generation and agglomeration of crystalline defects, decreased thermodynamic stability and changes in physical properties (e.g., hardness, elastic modulus, fracture toughness). Fundamental studies are essential to acquire an understanding of ion irradiation induced defect accumulation mechanisms and microstructure damage evolution.

Ceramics may be used within nuclear reactors in various applications; protective cladding, part of the fuel matrix, burnable poisons or as structural materials. Ceramics are also used for radioactive waste immobilisation. For these applications the ceramic must be highly durable, able to accommodate

a large percentage of actinides and be tolerant to the effects of radiation damage.[2]

The Ln_2TiO_5 (Ln = lanthanides and Y) series is based on three different structure types at room temperature. With decreasing ionic radius the Ln cation, the structure changes from orthorhombic to hexagonal to cubic, the latter having some similarities to pyrochlore and defect fluorite compounds. Pyrochlores make up part of the polyphase ceramic found within the commercial product Synroc (Synthetic Rock). This product has been proposed for the immobilisation of nuclear waste due to its ability to incorporate large percentages of actinides into its structure. Despite their appearance within waste forms or potential use as burnable poison there is still relatively little literature on these particular materials and their response to exposure to radiation.[3-6]

Figure 1 shows a series of bright field images of Gd_2TiO_5 with increasing Kr^{2+} ion dose along with corresponding selected area diffraction patterns. The images show a diminishing in diffraction contrast with increasing dose. The diffraction patterns show an increase in diffuse scattering halo and reduction in diffracted spot intensity representing an increase in amorphous phase with increasing dose.

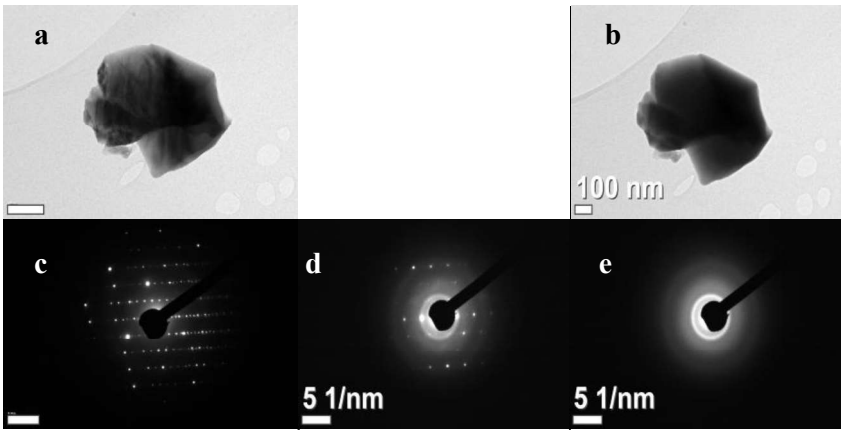

Fig. 1 (a and c) Unirradiated Gd_2TiO_5 crystalline grain and SAD. (d) Irradiated to 9.375×10^{13} ions cm^{-2} with 1MeV Kr^{2+} ions showing diffuse halo in SAD indicating introduction of amorphous phase. (b and e) Irradiated to 2.8125×10^{14} ions cm^{-2} showing completely amorphous material.

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