

## Radiation Response of Synthetic U-containing Sn-Zr Based Pyrochlore

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A closed nuclear fuel cycles will generate plutonium and minor actinides (Np, Am, Cm) that are required to be immobilized into durable nuclear waste forms for long-term geologic disposal. Isometric pyrochlore,  $A_2B_2O_7$ , is a promising host phase for the incorporation of actinides. Many studies have simulated the alpha-decay damage in a wide range of compositions using ion beam irradiation. In the present study, the response to the radiation damage of U-doped Sn-Zr based pyrochlore has been studied by 1-MeV  $Kr^+$  ion irradiation and characterized by *in situ* transmission electron microscopy (TEM).

Sn-Zr based, lanthanides-mixed pyrochlore doped with U was synthesized by mechanical activation with two-stages sintering of the oxide mixture for 5 hrs at 1350 C. The detailed sample compositions was listed in Table 1, with the general formula  $(REE)_2(Sn-Zr,U)_2O_7$ . The mixed composition of RE oxides corresponds to formula as  $REE_2=La_{0.26}Ce_{0.51}Pr_{0.24}Nd_{0.79}Sm_{0.14}Eu_{0.04}Gd_{0.03}$ . A SEM image of the Sn-Zr sample was shown in Fig. 1a as a representative of the microstructure of pyrochlore with a small amount of impurity. The ordered crystal structure and composition were characterized using a JEOL JEM 2010F electron microscope. (EDS was shown in Fig. 1b) A 1-MeV  $Kr^{2+}$  ion irradiation was performed in the IVEM-Tandem Facility at the Argonne National Laboratory over the temperature range from 50 to 700K. The microstructure evolution was monitored intermittently by taking selected area electron diffraction (SAED) pattern during irradiation.

A series of SAED patterns of Sn-pyrochlore and Zr-pyrochlore irradiated by 1-MeV  $Kr^{2+}$  ions at different ion fluencies at room temperature (RT) and 50K respectively was shown in Fig 2.  $(REE)_2(Sn_{1.93}U_{0.07})O_7$  pyrochlore was susceptible to radiation-induced amorphization at RT after a fluence of  $6 \times 10^{14}$  ions/cm<sup>2</sup>, while  $(REE)_2(Zr_{1.93}U_{0.07})O_7$  was radiation resistant without amorphization even at 50K after a fluence of  $2.5 \times 10^{15}$  ions/cm<sup>2</sup>, instead forms a disordered fluorite structure. The critical amorphization was plotted as a function of temperature as shown in Fig. 3. The critical temperature of Sn-pyrochlore, above which the complete amorphization does not occur, is 650K. The mixed Zr-Sn based pyrochlore shows an intermediated response with a critical temperature of 510K.(summarized in Table 1) The relation of radiation resistance and cation ionic radius ratio shows that the material with greater resistance to irradiation-induced amorphization has structure closer to the fluorite structure (with smaller  $r_A/r_B$ ).

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Table 1. Summary of the characteristics of synthesized U-doped pyrochlore samples and their irradiation response data. ( $A_2$ :La<sub>0.26</sub>Ce<sub>0.51</sub>Pr<sub>0.24</sub>Nd<sub>0.79</sub>Sm<sub>0.14</sub>Eu<sub>0.04</sub>Gd<sub>0.03</sub>).

sample	stoichiometry	$r_A/r_B$	$F_c$ at RT (ions/cm <sup>2</sup> )	$T_c$
Zr	A <sub>2</sub> (U <sub>0.07</sub> Zr <sub>1.93</sub> )O <sub>7</sub>	1.544	2.5X10 <sup>15</sup>	<50K
Sn-Zr	A <sub>2</sub> (U <sub>0.07</sub> Zr <sub>0.93</sub> Sn)O <sub>7</sub>	1.587	1.3X10 <sup>15</sup>	510K
Sn	A <sub>2</sub> (U <sub>0.07</sub> Sn <sub>1.93</sub> )O <sub>7</sub>	1.630	0.75X10 <sup>15</sup>	650K

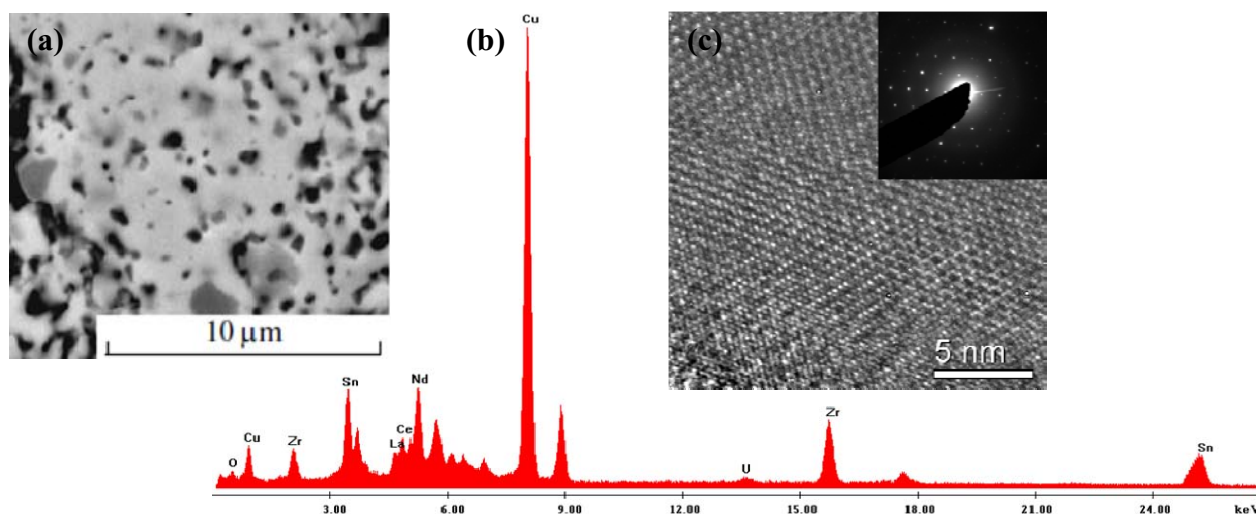


Fig. 1 (a) SEM image of microstructure of (REE)<sub>2</sub>(U<sub>0.07</sub>Zr<sub>0.93</sub>Sn)O<sub>7</sub> pyrochlore and corresponding energy-dispersive X-ray spectrum (b) and HRTEM image (c).

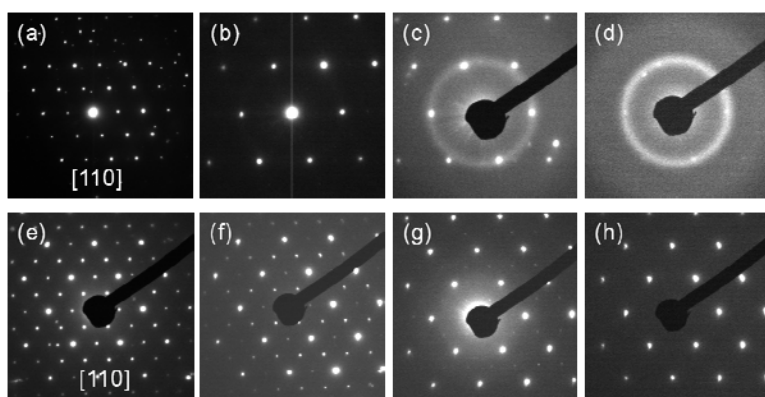


Fig. 2 A series of SAED patterns for Sn-pyrochlore irradiated by 1-MeV Kr<sup>2+</sup> ions at different ion fluencies of (a)original, (b)2.5x10<sup>14</sup>, (c)4x10<sup>14</sup> and (d)6x10<sup>14</sup> ions/cm<sup>2</sup> at RT, and for Zr-pyrochlore irradiated at 50K: (e)original, (f)1.75x10<sup>14</sup>, (g)4x10<sup>14</sup>, (h)1.25x10<sup>15</sup> ions/cm<sup>2</sup>.

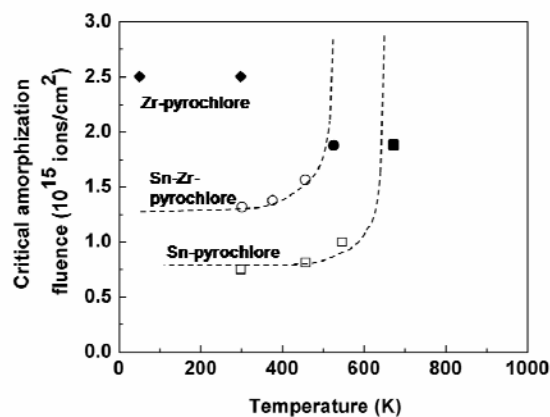


Fig. 3 The amorphization fluences of U-doped pyrochlore as a function of temperature.