Neutron Total Scattering and X-ray Diffraction Analysis of Radiation Effects in A3BO7 Weberite-type Complex Oxides

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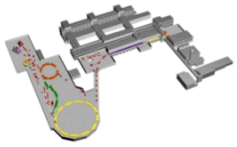
Motivation

- Nuclear technology requires materials which maintain structural stability in harsh radiation fields
- Irradiation-induced structural disorder in weberite-type (A_3BO_7) oxides is not studied

Methods

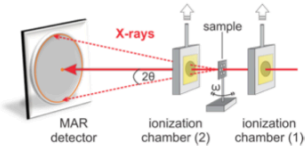
Swift Heavy Ion Irradiation at GSI

- 2.2 GeV ^{197}Au (electronic energy loss)
- Fluence: $5\times10^{12} - 8\times10^{12}$ cm^{-2}



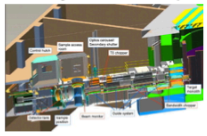
X-ray Diffraction at APS

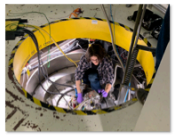
- High-resolution, intense light sources



Neutron Total Scattering at NOMAD

- Hight-resolution, neutron total scattering with access to both diffraction and pair distribution analysis techniques





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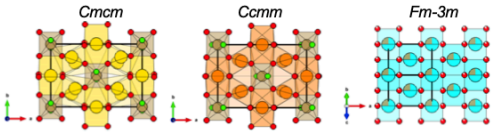
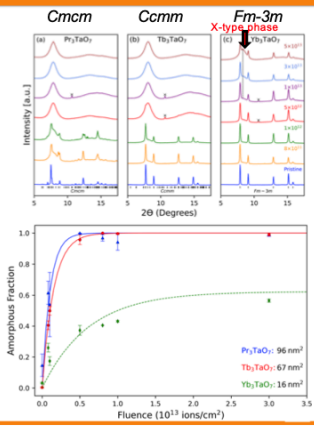
Materials

- **A_3TaO_7** (A = Pr, Tb, Dy, Y, Yb)
- Tuning the structure by changing the A-site cation
- 2 different tilt systems (*Cmcm*, *Ccmm/C222$_1$*) and 1 disordered structure (*Fm-3m*)

A-cation Effect on Long-range Structure

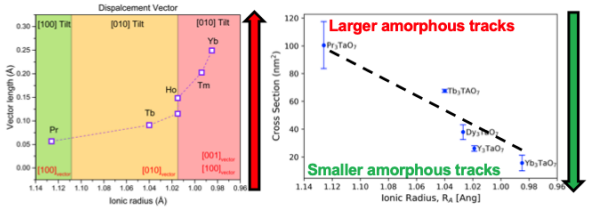
- A-site is tuned to produce different A_3BO_7 structures
- The radiation responses are dramatically different
- Ion-induced amorphization
- Increase in diffuse scattering beneath Bragg peaks
- Rapid, complete amorphization of Pr_3TaO_7, Tb_3TaO_7
- Yb_3TaO_7 remain partially crystalline (~40%)
- Formation of **X-type phase**

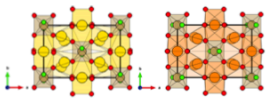




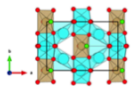
Radiation Resistance as a Function of Disorder

- Radiation resistance in **A_3TaO_7** system is correlated with the pre-existing level of local disorder (i.e., local polyhedra distortions away from ideal long-range lattice positions)
- Structures with intrinsically larger level of pre-existing disorder show an improved amorphization resistance





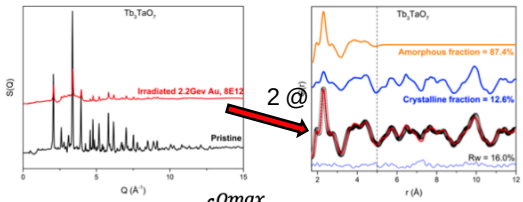
Local motifs in $(Pr,Tb)_3TaO_7$



Local motif in Yb_3TaO_7

Radiation Resistance of Short-range Structure

- Average, long-range weberite-type structure in Tb_3TaO_7 shows significant amorphization **(~80-90% amorphous)**
- Local, short-range structure is highly resistant
- No significant local modifications due to large difference in charge states of A^{3+} and B^{5+}
- Cation sites cannot mix



$$G(r) = A * Q_{max} \int_{Qmin}^{Qmax} Q(S(Q)-1)Sin(Qr)dQ$$

Conclusions

- Radiation response of complex oxides is heterogeneous across different length scales
- Local atomic arrangement must be studied to obtain comprehensive understanding
- In A_3TaO_7 system, materials with pre-existing level of disorder display a higher radiation stability over the long range
- Local weberite-type motif is present even in almost completely amorphous samples

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