## Ion Irradiation of $ThO_2$ and $UO_2$ Single Crystals

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The goal of this project was to investigate the impact of irradiation induced defects on thermal transport in nuclear fuel materials. Considering previous irradiation of $CeO_2$ and $UO_2$ using light ions, two irradiation temperature regimes were considered [1,2]. The first room temperature regime targeted a microstructure dominated by point defects. The second irradiation, performed at 600°C, targeted a microstructure dominated by interstitial dislocation loops. Two MeV protons were selected to develop a uniform damage profile into the top 20 µm of the sample for spatially resolved thermal conductivity measurement [3].

**Experimental or Technical Approach**

Single crystals of $ThO_2$ and $UO_2$ obtained using the hydrothermal growth approach were provided by the Air Force Research Laboratory. Irradiations were performed utilizing the ion beam accelerator at Texas A&M University using 2 MeV protons up to estimate doses of 0.2 and 0.8 dpa at room temperature and at 600°C, respectively. Extensive characterization has been performed on the irradiated single crystal thoria sample. Transmission electron microscopy (TEM) characterization of extended defects induced by ion irradiation was performed using facilities located at Idaho National Laboratory's Irradiated Materials Characterization Laboratory (IMCL) and included FEI Quanta 3D FEG Dual Beam focused ion beam for TEM lamellae preparation and Thermo Scientific Titan Themis TEM. Subsequently, additional microstructure characterization of point defects was conducted using Raman spectroscopy, photoluminescence spectroscopy, and optical spectroscopic ellipsometry. The impact of defects on thermal conductivity was measured using a modulated thermoreflectance approach [4]. Microstructure evolution was analyzed using rate theory modeling [5,6]. Thermal conductivity was analyzed using both classical models for thermal transport [1,2] with input from first principles calculations [9,10].

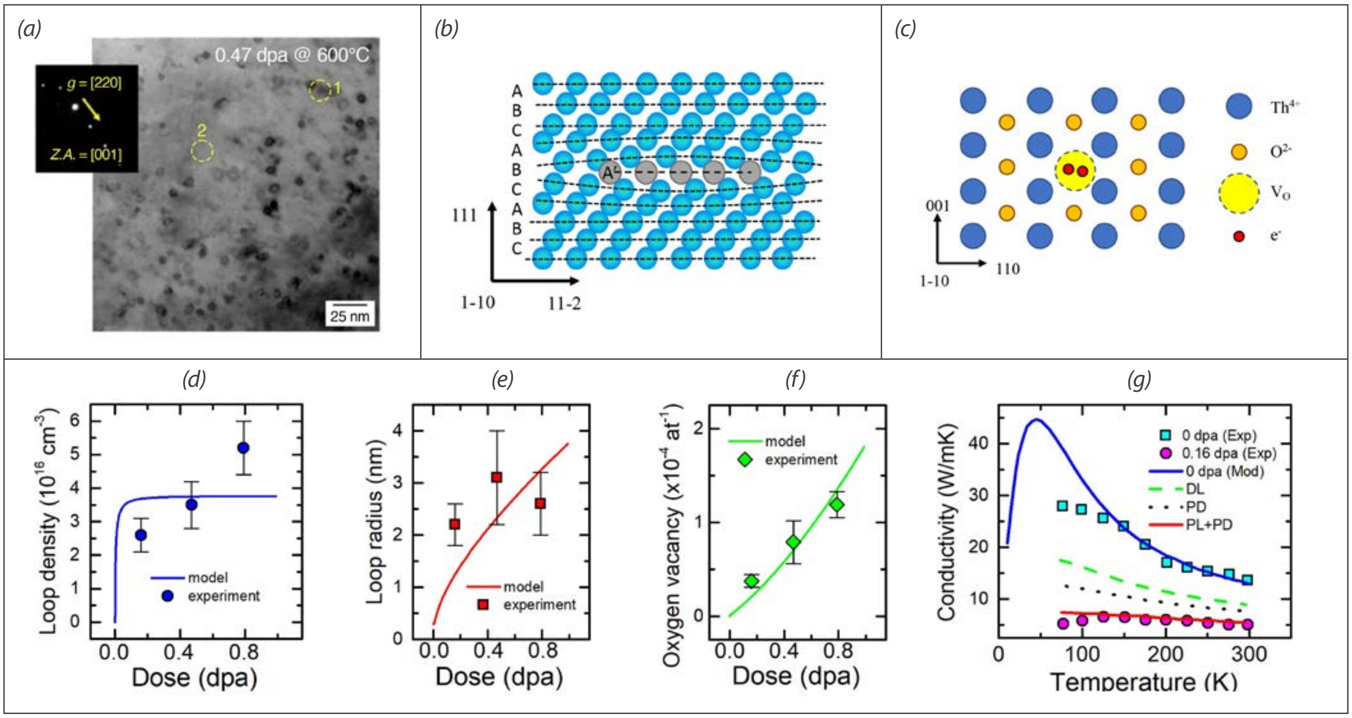


*Figure 1. Characterization of proton irradiated single crystal $ThO_2$. (a) Bright field TEM image revealing dislocation loops [8]. (b) 1/3(111){111} loop structure revealed by $\vec{g} \cdot \vec{b}$ invisibility criterion analysis [8]. (c) The proposed structure of oxygen vacancy visible in optical spectroscopy. Evolution of (d) dislocation loop density, (e) loop diameter, and (f) oxygen vacancy concentration obtained from experimental characterization [10]. Solid lines are result of rate model predictions parametrized using these results. (g) Thermal conductivity reduction analysis [10].*

## Results

Bright field image combined with rel-rod dark field TEM image analysis was used to observe and quantify the diameter and density of dislocation loops in the high-temperature irradiated samples [7]. Using standard $\vec{g} \cdot \vec{b}$ analysis and inside-outside contrast methods, the loops were determined to be 1/3(111){111} oriented interstitial Frank faulted loops [8]. Characterization performed using Raman and optical spectroscopies revealed an abundance of point defects and small defect clusters in both room- and high-temperature irradiated samples. These observations suggested that at 600°C some of the point defects are not sufficiently mobile to ensure their recombination or absorption by extended defects [7]. This is in contrast to the loop dominated microstructure observed in polycrystalline ceria irradiated at 700°C [1]. The abundance of point defects in thoria is consistent with the rate theory modeling analysis of microstructure [10]. Thermal conductivity analysis revealed that the conductivity reduction as a function of dose is due to point defects in room temperature irradiated samples as expected. However, for the high temperature irradiated samples, the conductivity is limited by phonon scattering by extended defects as well as by point defects on thorium sublattice.

**Discussion/Conclusion**

Ion irradiation and extended defect characterization enabled by this NSUF project revealed information on microstructure evolution in $ThO_2$ under irradiation. The experimental data is critical to furthering our fundamental understanding of the behavior of irradiated nuclear fuels.

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**Publications**


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## Distributed Partnership at a Glance

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| Idaho National Laboratory | Irradiated Materials Characterization Laboratory |
| **Collaborators** | |
| Idaho National Laboratory | Lingfeng He (co-principal investigator), David Hurley (co-principal investigator) |