Center for Thermal Energy Transport under Irradiation (TETI)
EFRC Director: David Hurley
Lead Institution: Idaho National Laboratory
Class: 2018 – 2026

***Mission Statement**: To accurately predict, from first principles, thermal energy transport in actinide materials in extreme environments.*

In nuclear fuel, irradiation-induced defects effectively scatter thermal energy carriers (electrons and phonons), greatly reducing the capacity of the fuel to transport heat to the coolant for eventual electricity generation. For example, in oxide fuels, the thermal conductivity decreases by as much as 70% over the operational lifetime of the fuel. This reduction significantly impacts fuel performance, safety margins, and the amount of usable energy. However, in some special cases, microstructure evolution can lead to local increases in thermal conductivity. For oxide fuels, examples include a reduction in phonon scattering associated with the transformation of faulted loops to perfect loops (a change in the strain field), transformation of a loop ensemble into a line segment (change in defect dimensionality), and defect segregation at interfaces (cooperative effects). Indeed, the myriad of defect types and interactions in nuclear fuel under irradiation naturally leads to the supposition that the deleterious losses in thermal conductivity can be mitigated by controlling defect evolution. The Center thus adopts the **vision** that a first-principles understanding of electron and phonon transport addressing the complexity of irradiation-induced defects will provide the necessary tools to control thermal transport in nuclear fuel.

Our vision will be examined from the perspective of two thrusts. The first will tackle phonon mediated thermal transport in advanced oxide fuels (thorium oxide - ThO_2 and thorium/uranium mixed oxide $Th_{1-x}U_xO_2$). The second thrust will emphasize electron and phonon mediated thermal transport in advanced nitride fuels (uranium nitride - UN and thorium nitride - ThN). Both thermal energy transport phenomena contain rich physics that are not well understood and can be investigated using simple systems.

To meet our vision we have defined four research goals that represent significant challenges in the field of thermal transport. These goals are enumerated below:

1. *Extend computational and experimental framework to temperature extremes*

   This will be crucial for developing a fundamental understanding of fuels at operating temperatures. The emphasis will be on fully extending our computational effort beyond 3rd order anharmonicity. To do this we will compare computed and measured phonon dispersion relationships, linewidths, elastic constants, Raman spectra, and thermal conductivities in perfect single crystals of $Th_{1-x}U_xO_2$ measured in extreme environments to reveal the significance of beyond 3rd order anharmonicity.

2. *Accurately measure electron-phonon coupling*

   Electron-phonon coupling is a controlling mechanism in electron-mediated thermal transport. The emphasis will be on using measurement orthogonality (multiple investigative methods spanning different aspects of the solution space) to accurately gauge the impact of electron-phonon coupling. This approach will involve first-principles modeling in tandem with INS/IXS and ARPES to investigate electron-phonon coupling from an electron and phonon perspective in ThN.

3. *Characterize the spectrum of defects and model defect carrier interactions*

   This is critical to developing accurate, first principles informed models of thermal energy transport under irradiation. The emphasis will be on using measurement orthogonality to characterize the size and distribution of sub-nanometer defects (defects that cannot be statistically characterized using

TEM techniques) in oxide and nitride fuels. This will be followed by a first-principles treatment of scattering of energy carriers by irradiation-induced defects.

4. *Understand defect segregation at interfaces and thermal transport across interfaces*

   Defect segregation at interfaces is a prototypic defect evolution mechanism that has been shown to have a net beneficial impact on thermal transport. Additionally, from a thermal transport perspective, interfaces can be studied in isolation using new experimental tools, allowing for measurements to be compared directly to atomistic prediction. Our emphasis will be on naturally occurring grain boundaries in oxide fuels and epitaxially grown heterointerfaces in nitride fuels.

Tackling the computational complexity is a far-reaching challenge. At the atomistic scale, the approach will involve using density functional theory (DFT) and beyond DFT methods able to capture many-body interactions to understand the role of 5*f* electrons on phonon and electron transport, defect formation, and scattering mechanisms. At the mesoscopic-length scale, thermodynamic modeling, molecular dynamics, and kinetic Monte Carlo will be used to understand defect interaction and evolution. Phonon-scattering mechanisms will be investigated using both perturbative and Green's function approaches and their impact on thermal transport will be captured using the Boltzmann transport equation (BTE).

These modeling approaches will be complemented by a well-defined set of electron- and phonon-structure measurements and transport measurements in ion-irradiated model fuels having well-characterized microstructures. Sample synthesis routes include hydrothermal and flux growth of large single crystals as well as heterointerfaces assembled using molecular beam epitaxy. Inelastic neutron and X-ray scattering will be used to measure phonon dispersion and lifetime. Angularly resolved photoemission spectroscopy, and low-temperature magnetic field measurements (de Haas-van Alphen and Shubnikov-de Haas) will be used to obtain the electronic structure of nitride fuels. Transmission electron microscopy, positron annihilation spectroscopy, and optical spectroscopy will be used to characterize the spectrum of defects produced by ion irradiation. Thermal wave microscopy combined with coherent acoustic wave spectroscopy will be used to make spatially resolved thermal transport measurements across heterointerfaces and isolated grain boundaries as well as the damage plateau in ion-irradiated samples. Scanning transmission electron microscopy and electron energy loss spectroscopy will be used to measure localized, phonon modes associated with interfaces.

Center for Thermal Transport under Irradiation (TETI)	
Idaho National Laboratory	David Hurley (Director), Boopathy Kombaiah, Krzysztof Gofryk, Zilong Hua, Amey Khanolkar, Kaustubh Bawane, Chao Jiang, Linu Malakkal, Brelon May, Shuxiang Zhou
Oak Ridge National Laboratory	Michael Manley
Airforce Research Laboratory	Mathew Mann
Columbia University	Chris Marianetti
The Ohio State University	Marat Khafizov
University of Texas at San Antonio	Elizabeth Sooby
Arizona State University	Farida Selim
University of Wisconsin	Yongfeng Zhang
The Pennsylvania State University	Miaomiao Jin

Contact: David Hurley, Director, david.hurley@inl.gov
(208) 360 6374, https://teti.inl.gov