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International Nuclear Energy Research Initiative Project No. 2010-006-E

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Idaho National Laboratory

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State-of-the-Art Post-Irradiation Examination of Advanced Nuclear Fuels

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Collaborators: Colorado School of Mines, Los Alamos National Laboratory, Massachusetts Institute of Technology, University of Central Florida

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Research Objectives

Safe and effective implementation of new closed nuclear fuel cycle concepts employing fast reactors with advanced fuel forms necessitates improved understanding of the behavioral characteristics of the proposed advanced fuels. The fuel systems under consideration include minor actinide (MA) transmutation fuel types such as advanced mixed oxides (MOX), advanced metallic alloys, inert matrix fuels (IMFs), and other ceramic fuels (nitrides, carbides, etc.) for fast neutron spectrum conditions. Most of these advanced fuel compounds have already been the object of past examination programs, which included irradiations in research reactors. The knowledge derived from previous experience constitutes a significant, albeit incomplete, body of data. Today's new or upgraded experimental tools can extend the scientific and technological knowledge associated with the new generation of nuclear reactors and fuels. More rapid progress can be achieved through effective synergy with advanced (multiscale) modeling efforts.

This project is undertaking further investigations using some of the state-of-the-art experimental techniques now available. The objectives are three-fold:

- Extend the available knowledge on properties and irradiation behavior of high burnup and minor actinide-bearing advanced fuel systems.
- Upgrade/develop advanced modeling tools using experimental data and expertise on the irradiation behavior of nuclear fuels, establishing a synergy between experimentation and multiscale modeling and code development.
- Promote the effective use of international resources—i.e., information exchange among leading experimental facilities—to characterize irradiated fuel.

Research Progress

In FY 2012, Idaho National Laboratory (INL) researchers worked towards developing high spatial resolution instrumentation that will help determine thermal conductivity and mechanical properties. They used a focused ion beam (FIB) to prepare and perform electron backscatter diffraction (EBSD) studies on irradiated fuel samples in conjunction with computational modeling and simulation efforts. Their experimental findings will be further coupled with the computational modeling and simulation efforts. Team members were successful in their efforts to transport FIB-prepared samples to the Institute for Transuranium Elements (ITU), supporting the planned joint characterization of irradiated silicon carbide (SiC).

Properties of Advanced Fuels and Synergy between Methodologies

Advanced characterization techniques such as transmission electron microscopy (TEM) and EBSD have been widely utilized for non-radiological materials; however, the extreme difficulty of working with radioactive samples has inhibited their application to nuclear investigations. These advanced techniques provide the ability to obtain critical experimental data on the evolution of microstructure, dislocation density, grain orientation, and composition in irradiated materials from the atomic to the mesoscale. Using FIB, the INL team has taken high-burnup fuel irradiated in the Fast Flux Test Facility (FFTF) reactor and produced site-specific samples for TEM and EBSD examination from this highly radioactive and friable material, obtaining first-of-a-kind images. In parallel, the team has developed the MARMOT code, INL's flagship mesoscale simulation code[1] that predicts the microstructural evolution in fuel under a variety of reactor and experimental conditions.

Both the experimental methods and the modeling and simulation techniques improve understanding of the evolution of the fuel's microstructure and defining its impact on important material properties. By combining the two methods, the INL team has developed a coordinated approach that greatly reduces the limitations of each method in isolation. Experimental microstructures, characterized both before and after irradiation, are fully reconstructed in the MARMOT code. Figure 1 shows experimental high-burnup mixed oxide fuel microstructures that have been computationally reconstructed and used as the simulation initial conditions. Using the resulting simulation, researchers can determine the effective thermal conductivity of various areas within the fuel pellet and predict grain boundary migration and fission gas segregation. The reconstruction includes the topology, grain orientation, crystal structure, and chemical composition.

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[1] 1. M. Tonks, D. Gaston, P. Millett, D. Andrs, and P. Talbot, "An object-oriented finite element framework for multiphysics phase field simulations," *Comp. Mat. Sci.*, 51 (2012) 20–29.

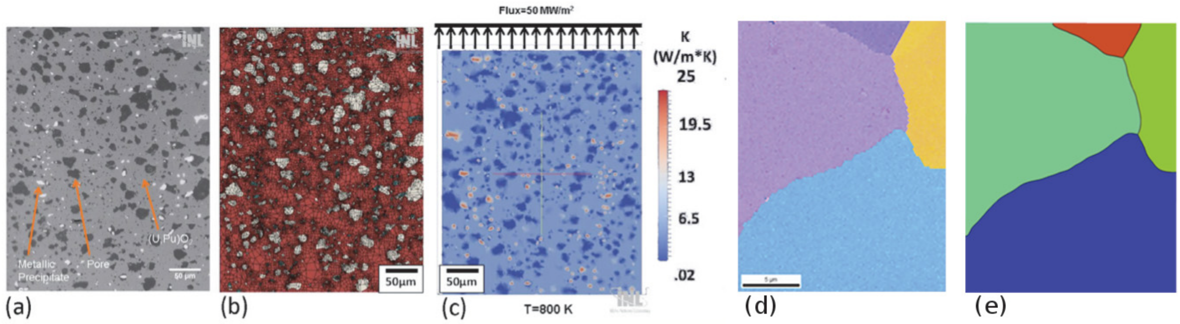

Figure 1. Examples of reconstructed microstructures: (a) optical micrograph of high-burnup ACO-3 metal oxide fuel, (b) reconstructed simulation mesh, (c) heat conduction simulation setup, with initial values for thermal conductivity overlaid on the mesh, (d) EBSD scan of ACO-3 fuel, and (e) reconstruction of the microstructure in the MARMOT code.

A postdoctoral researcher from ITU worked on the INL team and implemented a physics-based fission gas behavior model that he had previously developed into the BISON fuel performance code . This model was developed for analyzing the coupled phenomena of fission gas swelling and release in UO_2 fuel during irradiation with an emphasis on modeling grain-face gas bubble development and the related dependence of the fission gas swelling and release on the local hydrostatic stress. This is of special importance when analyzing fuel behavior during power ramps and pellet-cladding mechanical interaction conditions.

ITU will provide INL (and EPRI) researchers with the final version of the Power_Condense computer program, including a new graphical interface. The program reduces the number of data points in a reactor power history, which can greatly reduce computational time when this data is imported into modeling codes. Data produced during reactor operation, such as linear heat rate, coolant temperature, or reactor state, are routinely stored in short time intervals, often 15 minutes or less. Because irradiation times can last several years, a huge amount of data is produced.

Advanced Instrumentation

The INL team is developing two advanced laser-based techniques to study the mechanical and thermal properties of nuclear fuel: the Thermal Conductivity Microscope and the Mechanical Properties Microscope.

Thermal Conductivity Microscope (TCM). This laser-based instrument measures thermal diffusivity and thermal effusivity directly, allowing users to determine thermal conductivity. This method alleviates the need to measure the specific heat separately, which is rather difficult under remote-handling conditions. The instrument is being designed so that it can be remotely operated and maintained, will function in a radiation environment, and can make measurements on nuclear fuel and other radioactive samples. Once operational, the TCM will provide micron-level thermal property information commensurate with the microstructure heterogeneity in nuclear fuel.

The project team achieved the following key accomplishments pertaining to the design, construction, and testing of the TCM:

- Developed an experimental protocol appropriate for measuring thermal conductivity of high-burnup nuclear fuel and fresh nuclear fuel.
- Completed a study on the suitability using a fiber laser for sample heating.
- Identified a figure of merit that can be used to judge data reliability.

The researchers used the TCM to obtain values for the effusivity, thermal diffusivity, and thermal conductivity of metallic systems; results compare very well with those known from the literature. Of particular note is the rather good correspondence between literature values and TCM-determined values for ceramic materials, as the data in Table 1 show.

Table 1. TCM-determined and literature values for SiO_2 and CaF_2 effusivity, thermal diffusivity, and thermal conductivity

	Sample A (SiO_2)	Sample B (CaF_2)
Phase lag (deg)	60.4	46.3
Effusivity ($J/m^2\ s^{1/2}\ K$), measured	1490	4570
Effusivity ($J/m^2\ s^{1/2}\ K$), literature	1436	4989
Effusivity error	<4%	~8%
Diffusivity (m^2/s) measured	9.80×10^{-7}	3.25×10^{-6}
Diffusivity (m^2/s) literature	9.5×10^{-7}	3.4×10^{-6}
Diffusivity error	3%	4%
Conductivity (W/(m K), measured	1.47	8.24
Conductivity (W/(m K), literature	1.4	9.2
Conductivity error	5%	10%
Effusivity ($J/m^2\ s^{1/2}\ K$), measured including $R_{th} = 5 \times 10^{-9}\ m^2\ K/W$	1460	4180

Mechanical Properties Microscope (MPM). This unique laser ultrasound instrument is designed to operate in a hot cell environment via remote control manipulation. In FY 2012, researchers initiated mockup testing of the MPM to measure mechanical properties of nuclear fuel. The MPM will provide micron-level mechanical property information commensurate with microstructure heterogeneity. The MPM's spatial resolution enables results to be tied directly to other electron/photon microstructural imaging technologies. This aspect is essential to understanding microstructure's role in determining mechanical properties of nuclear fuel.

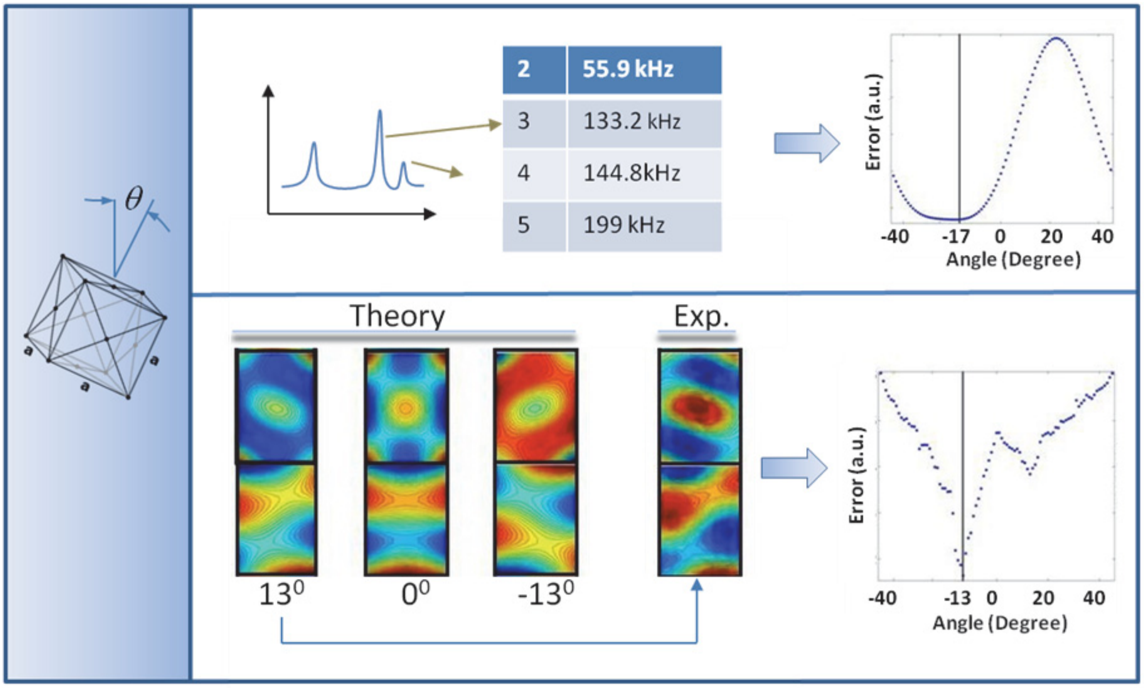

Figure 2. Cover illustration for the November 2012 issue of IEEE-UFFC. The MPM was used to measure crystallographic orientation of a single crystal sample. Laser resonant ultrasonic spectroscopy (LRUS) uses lasers to excite and detect resonant modes. The crystallographic orientation of a copper sample determined using the eigenmode method (bottom) is compared with the traditional eigenfrequency method (top).

Researchers completed a number of important steps on the path to implementing remote MPM operation: obtaining final approval of the remote equipment qualification plan for Phase I testing, finalizing construction of the prototype instrument, and completing initial mockup testing. In addition, the team took first-time measurements of the elastic constants of a cast uranium–molybdenum fuel ingot and compared the data to a textured uranium–molybdenum fuel plate imposed by rolling (see Table 2).

Table 2. Comparison of elastic constants of a cast uranium–molybdenum fuel alloy and a rolled textured sample

	Isotropic Ingot	Anisotropic rolled foil	Percent change
Young's Modulus (GPa)	102	71	30
Shear Modulus	36	25	30

Joint Studies

One of the more logistically difficult project tasks is a combined characterization effort of irradiated fuel samples. The collaborators have discussed several irradiated and non-irradiated fuel materials and are exploring the viability of sharing Euratom's METAPHIX irradiation test samples with U.S. researchers. The transport of samples between INL and ITU remains a difficult issue.

Nonetheless, the project team has made progress with the cooperative study of irradiated SiC. Four β-SiC samples have been investigated, three of which were irradiated in the Massachusetts Institute of Technology reactor, under the umbrella of the Advanced Test Reactor National Scientific User Facility. Colorado School of Mines performed Hall coefficient measurements in 2011 to determine electrical carrier densities and impedance. In CY 2012, INL employed FIB techniques to prepare atom probe tomography, transmission electron microscopy, and nano- and micro-indentation measurement samples, as well as samples for shipment to ITU for micro-indentation and TEM measurements. These samples have been successfully shipped and are currently under examination at ITU.

Along with materials, the two research teams continue to exchange information and knowledge. Drawing on ITU experience, INL is working to implement an electron probe microanalyzer (EPMA) for highly radioactive fuels and materials. The two teams reviewed the coating device for preparation of EPMA samples, how INL could adapt the design, and how to use software for microprobe measurement and data processing. In addition, INL remains involved in the ITU-initiated informal Cameca shielded SX 100 group of users, who share experiences and problem-solving ideas.

Planned Activities

In FY 2013, researchers plan to extend the EPMA information exchange to the nuclearization of a FIB apparatus. A FIB will be delivered to ITU in early 2013 and their researchers will visit INL to discuss details of the device's installation.

Researchers will maintain interactions related to post-irradiation examinations, including operation of an EPMA. They will also continue to develop advanced techniques such as micro X-ray diffraction, FIB, thermal conductivity measurements, and mechanical property measurements.

The project team will move forward with joint studies on irradiated SiC. Team members will also continue their efforts to resolve issues associated with transport of high-level radioactive materials, including fuels. For the project to deploy its full scope—including joint characterization of materials—a viable means of transport to facilitate sample exchange is essential. Until a solution is found, the scope of the joint work will remain forcibly limited.