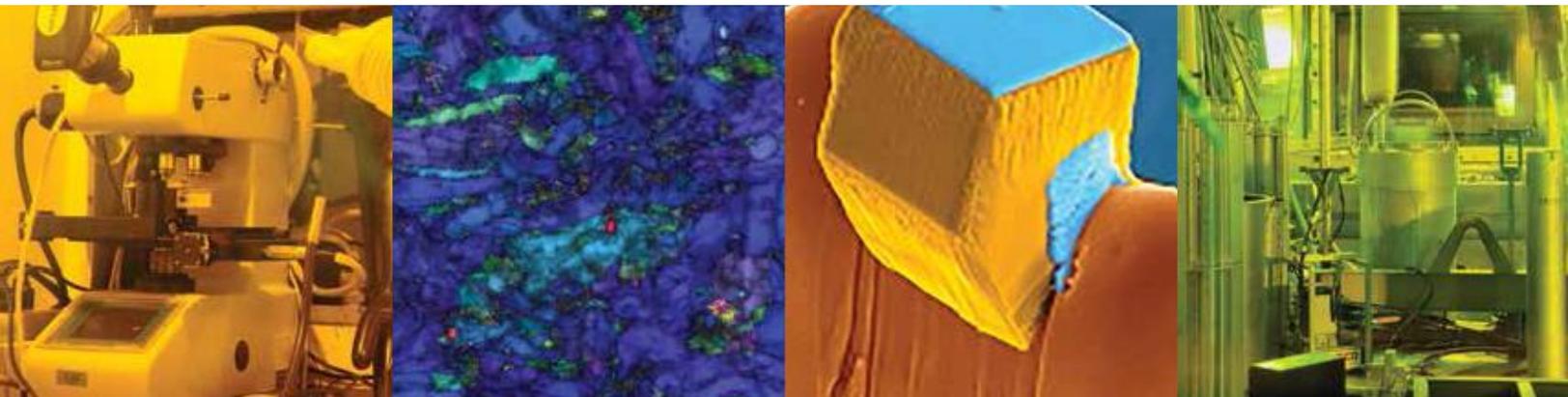


# ADVANCED FUELS CAMPAIGN

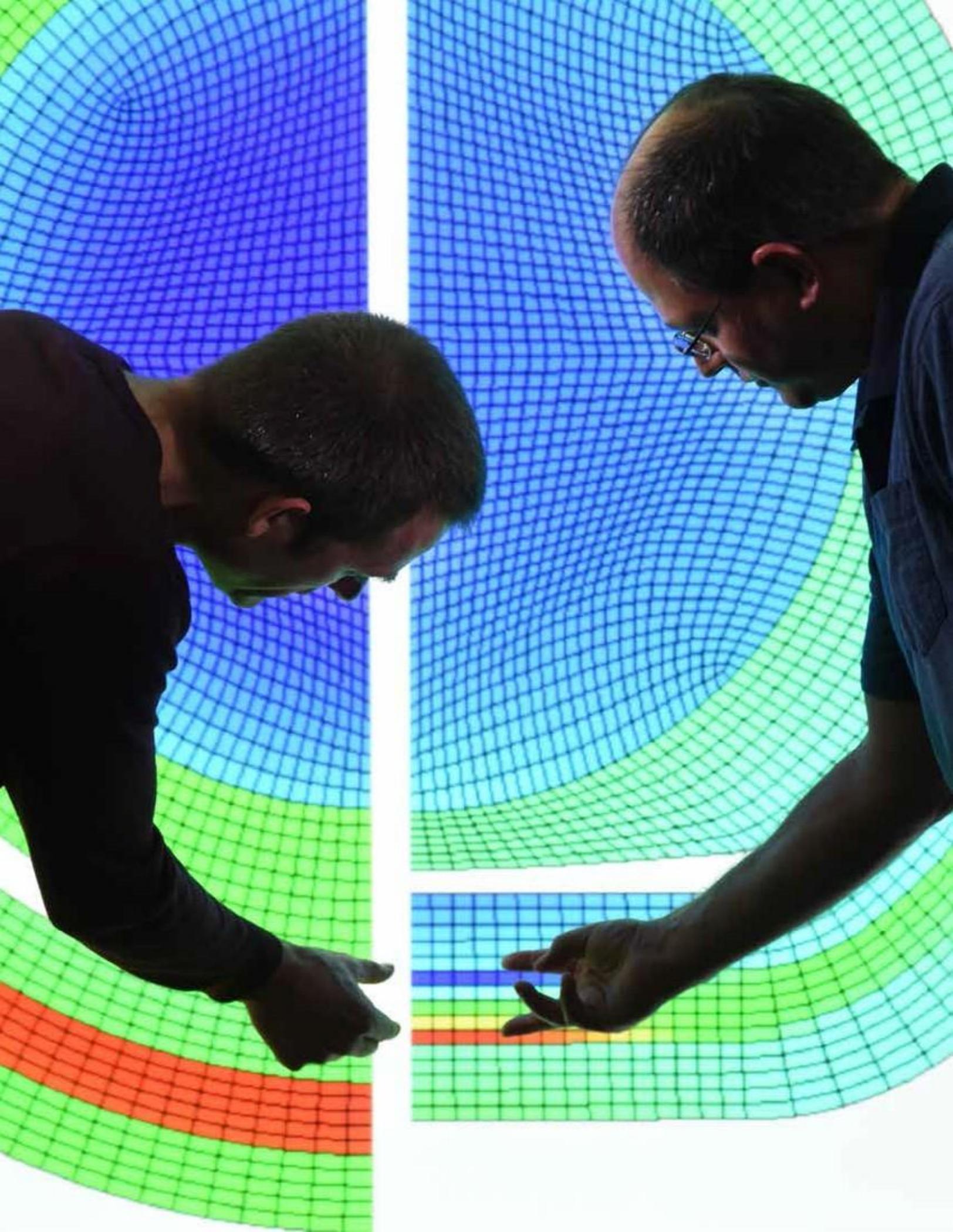
## 2016 Accomplishments



**AFC** Advanced Fuels Campaign

November 2016

FCRD-FUEL-2016-000116



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## Fuel Cycle Research and Development

# Advanced Fuels Campaign 2016 Accomplishments

INL/EXT-16-40127

Revision 0

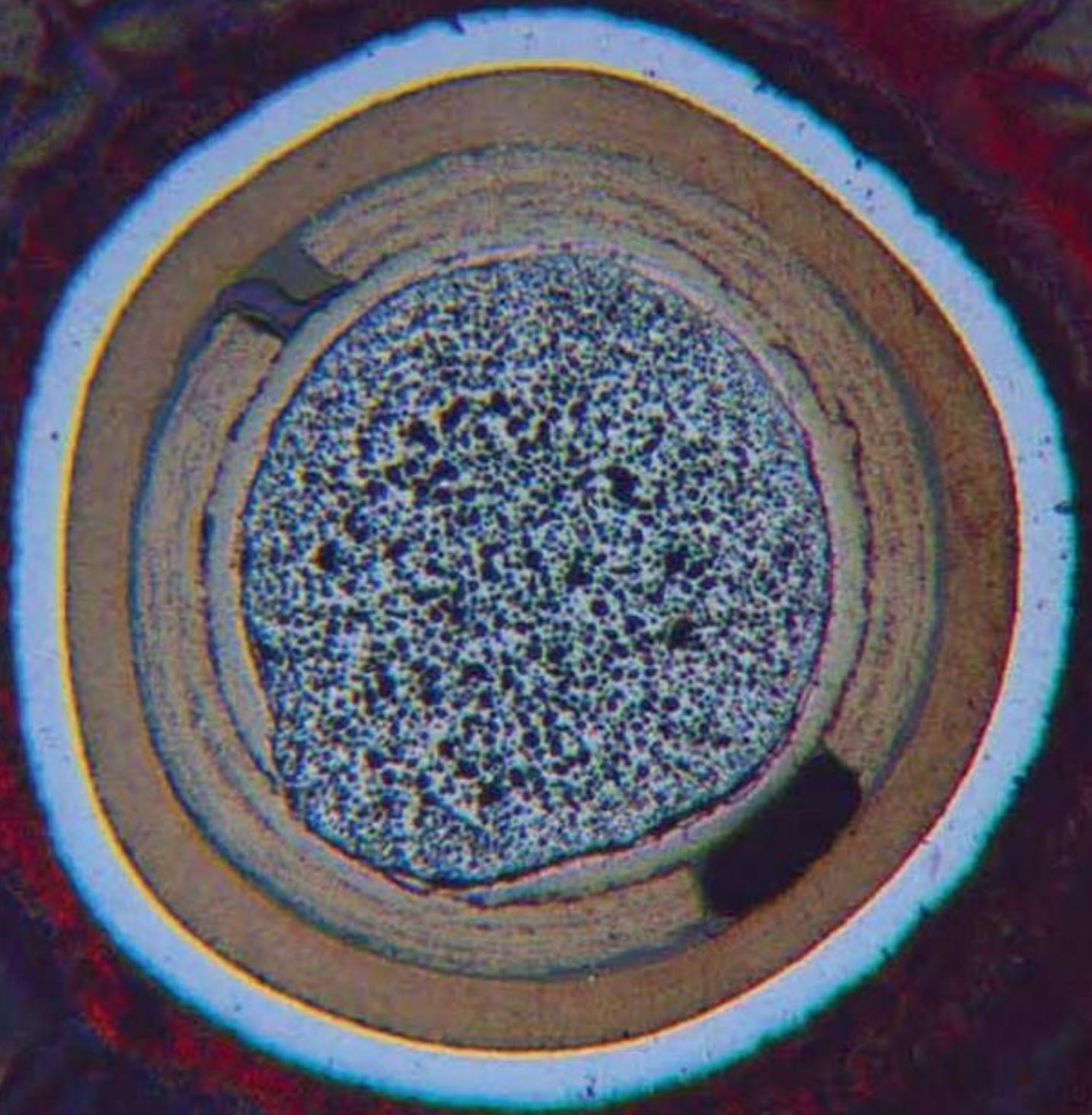
November 2016

Compiled and edited by:

\_\_\_\_\_  
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FCRD AFC National Technical Director



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# AFC MANAGEMENT AND INTEGRATION

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- 1.1 The Advanced Fuels Campaign Team
- 1.2 From the Director (Campaign Overview)
- 1.3 International Collaborations



# 1.1 THE ADVANCED FUELS CAMPAIGN TEAM

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## 1.2 FROM THE DIRECTOR

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The mission of the Advanced Fuels Campaign (AFC) is to perform research, development, and demonstration (RD&D) activities for advanced fuel forms (including cladding) to enhance the performance and safety of the nation's current and future reactors; enhance proliferation resistance of nuclear fuel; effectively utilize nuclear energy resources; and address the longer-term waste management challenges. This includes development of a state-of-the-art research and development (R&D) infrastructure to support the use of a "goal-oriented science-based approach."

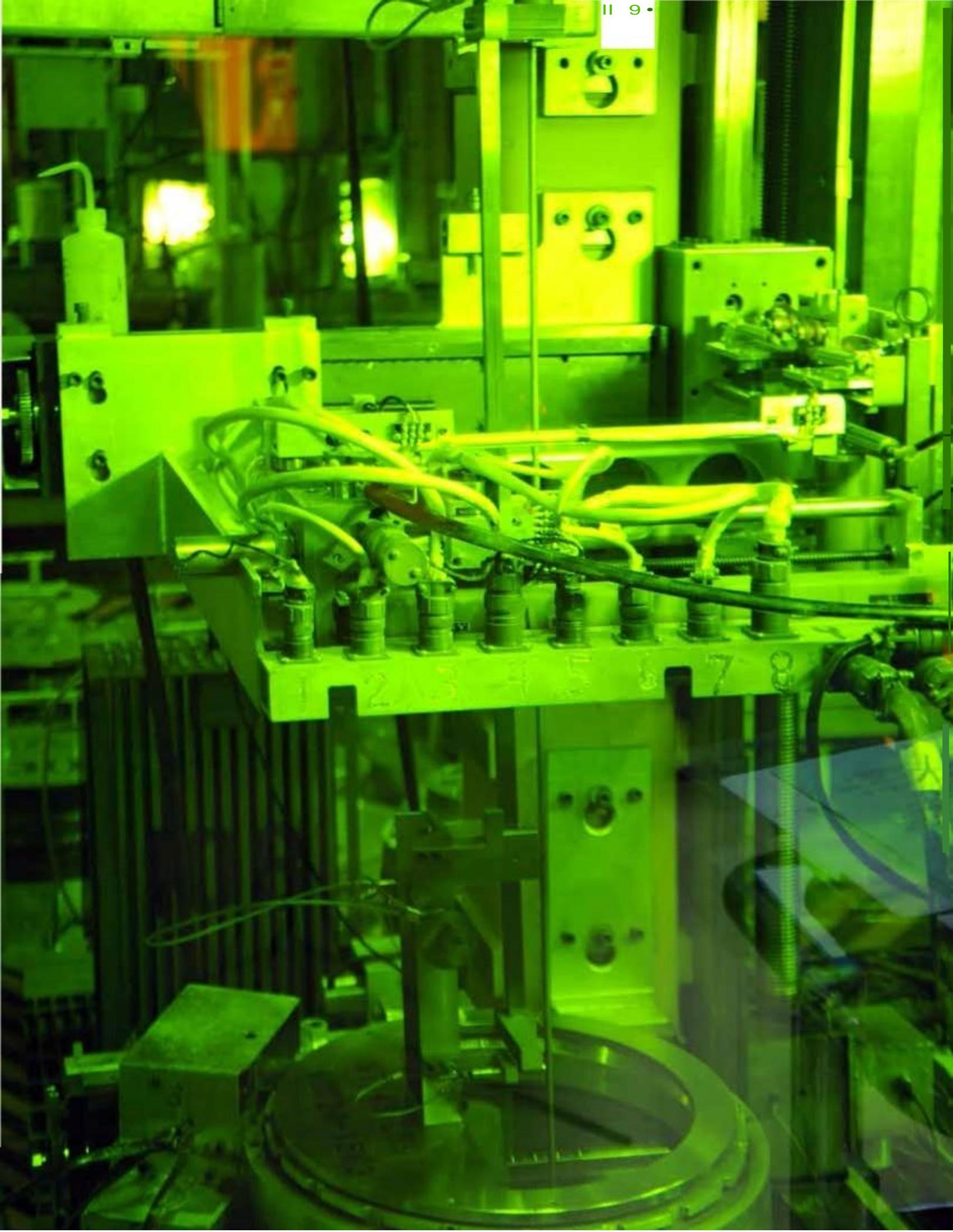
In support of the Fuel Cycle Research and Development (FCRD) program, AFC is responsible for developing advanced fuel technologies to support the various fuel cycle options defined in the Department of Energy (DOE) Nuclear Energy Research and Development Roadmap, Report to Congress, April 2010.

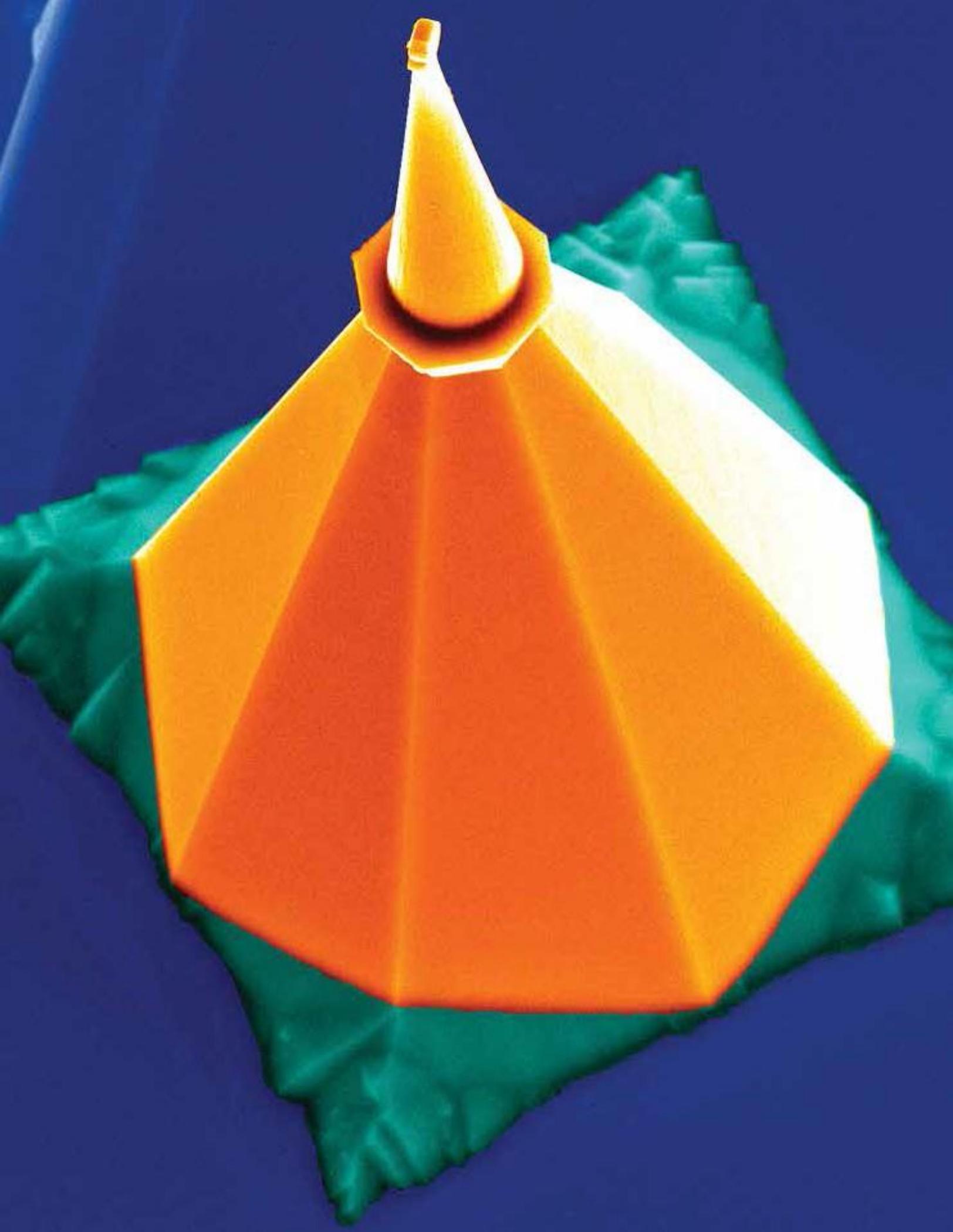
AFC uses a "goal-oriented, science-based approach" aimed at a fundamental understanding of fuel and cladding fabrication methods and performance under irradiation, enabling the pursuit of multiple fuel forms for future fuel cycle options. This approach includes fundamental experiments, theory, and advanced modeling and simulation. The modeling and simulation activities for fuel performance are carried out under the Nuclear Energy Advanced

Modeling and Simulation (NEAMS) program, which is closely coordinated with AFC. In this report, the word "fuel" is used generically to include fuels, targets, and their associated cladding materials.

Light-water reactor (LWR) fuel with enhanced accident tolerance is another R&D area under AFC. These fuel systems are designed to achieve significantly higher fuel and plant performance to allow operation to significantly higher burnup, and to provide enhanced safety during design basis and beyond design basis accident conditions. The overarching goal is to develop advanced nuclear fuels and materials that are robust, have high-performance capability, and are more tolerant to accident conditions than traditional fuel systems.

AFC management and integration activities in fiscal year (FY-16) included continued support for international collaborations, primarily with France, Japan, the European Union, Republic of Korea, and China, as well as various working group and expert group activities in the Organization for Economic Cooperation and Development Nuclear Energy Agency (OECD-NEA) and the International Atomic Energy Agency (IAEA). Three industry-led Funding Opportunity Announcements (FOAs) and two university-led Integrated Research Projects (IRPs) funded in 2013, made significant progress in fuels and materials development. All are closely integrated with AFC and accident-tolerant fuels (ATF) research.





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Accomplishments made during FY-16 are highlighted in this report, which focuses on completed work and results. The key FY-16 technical area outcomes are highlighted below.

- **International Coordination and Collaboration:** Bilateral agreements are supported with France, Japan, the European Union, the Republic of Korea, and China. The emphases in all collaboration activities are advanced LWR fuels with enhanced accident performance, metallic fuel development, irradiation testing and data analyses, and development of characterization and postirradiation examination (PIE) techniques. Two joint irradiation projects have been developed with the Halden Reactor Project (Norway) in advanced LWR fuels, an instrumentation qualification test in the Advanced Test Reactor in advance of the ATF-2 loop test and a creep test of FeCrAl and silicon carbide (SiC) samples in the Halden reactor. Activities are supported under four multinational agreements and arrangements, the Gen IV Sodium Fast Reactor project arrangement, the OECD-NEA, the European Atomic Energy Community (EURATOM), and coordinated research projects under IAEA. These multinational agreements allow the review and coordination of fuel development activities world-wide.
- **Advanced LWR Fuels with Enhanced Accident Tolerance:** The primary focus is to continue fundamental RD&D on several promising ATF

concepts; establish screening attributes and metrics for ATF concepts; and coordinate research activities between DOE laboratories, industry FOA teams, university IRP teams and Nuclear Energy University Program (NEUP) investigators. Critical testing capability is required for ATF development. This includes high-temperature steam oxidation testing (recently developed specifically for ATF), material property measurements, and irradiation testing.

- **Advanced Reactor Fuels:** Primary RD&D areas include advanced fabrication technology development; fabrication and characterization of minor actinide (MA) and lanthanide-bearing fuels; fundamental property measurements and fuel-cladding chemical interactions (FCCI) testing; and irradiation performance testing.
- **Capability Development:** Primary RD&D areas include advanced modeling and simulation (M&S) of fuel performance and fabrication processes; characterization technique development; and unique in-pile and out-of-pile material property measurements. Included in FY-15 was the establishment of experimental transient testing capabilities for the Transient Reactor Test (TREAT) reactor. Three principal test modes are currently under development, a static capsule test capability, a water test loop, and a sodium test loop.

## 1.3 INTERNATIONAL COLLABORATIONS

Advanced Fuels Campaign (AFC) researchers are very active in international collaborations with Korea, France, Japan, China, Russia, EURATOM, and OECD-NEA. These interactions and collaborations are managed through a combination of participation in Generation IV Global International Forum projects, International Nuclear Energy Research Initiative (INERI) projects, and participation in bilateral and trilateral government-to-government agreements. The ceramic fuels areas have collaborations primarily under the headings of Advanced Fuels within the US/Japan bilateral and the GenIV SFR. There is also collaboration on Field Assisted Sintering of Nuclear Fuels under a US/EURATOM/INERI arrangement.

### **GenIV-Sodium Fast Reactor Arrangement on Advanced Fuels**

*K.J. McClellan, kmcclellan@lanl.gov, LANL*

The Sodium Fast Reactor Advanced Fuel (SFR-AF) arrangement started in 2007 with a targeted duration of 10 years within the frame of the Generation IV Sodium Fast Reactor program. The primary objective is to investigate high burn-up Minor Actinide bearing fuels as well as cladding and wrapper materials capable of withstanding high neutron doses and temperatures. The project has been structured in 3 steps: evaluation of advanced fuels and materials options, Minor-Actinide bearing fuels evaluation, and assessment of high burn-up capability of advanced fuel(s) and materials. Participants in the arrangement include the DOE, Commissariat à l'Énergie Atomique (CEA), JAEA, KAERI, EURATOM, China and Russia with the

latter two having joined in December 2015. In FY16 program management board completed the Advanced Sodium Fast Reactor (SFR) Fuel Type Recommendation milestone which confirmed the prior Advanced Fuel Comparison report on fuel types and noted that the final SFR fuel type selection for each member country is dependent upon multiple domestic factors. The specifics of each country's experience, infrastructure and policies are critical determining factors in addition to the technical aspects in determining a preferred fuel type; the country-specific recommendation along with the reasoning was presented for each member country. Changes in the representatives and/or alternate representatives were made for EURATOM, France, and Russia in 2016.

### **US/Japan CNWG Collaboration on Advanced Fuels**

*K.J. McClellan, kmcclellan@lanl.gov, LANL*

Cooperative research under the Advanced Fuels area of the Fuel Cycle R&D and Waste Management Sub-Working Group is performed under the general areas of properties, performance and analysis. The goal of this effort is to perform collaborative R&D for evaluation of basic properties and irradiation behavior of advanced fuels. The objectives of the collaboration are to expand the basic properties and performance data and to improve understanding of advanced fuels with an emphasis on employment of advanced experimental techniques. Through incorporation of new minor actinide - mixed oxide fuel (MA-MOX) irradiation data the effort will also enable development and application of advanced modeling and

simulation tools for design and performance analysis of oxide fuels. In FY16, technical expert meetings were held in Japan and in the US at Idaho National Laboratory (INL) to advance specific tasks on basic properties of fuels, development of PIE data, and modeling and simulation of irradiated transmutation MOX fuel. Several joint publications from the fuel properties activities were prepared during the period. A key accomplishment in FY16 was negotiation of a Bison license for JAEA which will allow the collaboration to advance in jointly developing a MA-MOX Bison model for fuel performance.

Another key aspect of the collaboration was a visiting JAEA scientist, Shinya Nakamichi, working at Los Alamos National Laboratory (LANL) on basic fuel properties. A highlight of the research by the current visiting scientist follows. For the near term, Mr. Nakamichi will be the last visiting scientist at LANL as the next CNWG visitors will reside at INL to support the Bison model development.

### **Sintering Behavior of U-Ce-O**

*J.T. White, jtwhite@lanl.gov, LANL; S. Nakamichi, JAEA-Tokai*

Collaboration between the DOE and Japan on characterization of the properties of nuclear materials under the Civil Nuclear Energy Research and Development Working Group (CNWG) is essential to advancement of mutual capabilities in this area. CNWG targets to co-develop nuclear fuel technology by leveraging facilities and resources belonging to both Japan and the US. Japan Atomic Energy Agency (JAEA)

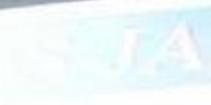




# JAPAN

TMI-2 Knowledge Transfer and Relevance  
to Fukushima Meeting

April 2011, 2012  
March 2013, 2014



## TMI-2 Knowledge Transfer to Fukushima Meeting

Idaho Falls, Id.  
Oct 11-13, 2013



Speaker at podium

Panel of speakers at a table

Panel of speakers at a table

Attendees at long tables with laptops, papers, and water bottles

research focuses on (U,Pu)O<sub>2</sub> as mixed oxide (MOX) fuel. Although current experimental work on MOX or other oxide transmutation fuels under DOE programs is minimal, the effect of Ce on the thermochemistry, thermophysical properties, and fabrication behavior of oxide fuel represent a common interest to both programs given the utility of Ce as a surrogate for Pu as well as its importance as a fission product.

Many studies have been reported on (U,Ce)O<sub>2</sub> as UO<sub>2</sub> with a fission product and a surrogate material of (U,Pu)O<sub>2</sub> since Ce is one of most prevalent fission products forming full solid solution over 1000°C. Furthermore, its 3+/4+ valence mirrors that of Pu, making it a suitable thermochemical surrogate. Fabrication of MOX remains an area of mutual interest given its status as the transmutation fuel of choice in Japan. No fuel form has been definitively chosen for US utilization, but MOX remains under consideration. The role of stoichiometry and specifically oxygen-to-metal ratio (O/M) on fuel properties such as thermal conductivity has been previously studied under the CNWG. However, recent work in Japan has identified O/M as a major contributor to the densification behavior of MOX. Further exploration of the role of O/M on densification of (U,Ce)O<sub>2</sub> was led by a Shinya Nakamichi, a guest researcher stationed at LANL from March 2015 until March 2016.

Dilatometer experiments were performed at LANL and JAEA-Tokai to assess the shrinkage behavior of both (U,Pu)O<sub>2</sub> and (U,Ce)O<sub>2</sub> as a function of O/M. Dynamic gas atmospheres were used to adjust and maintain

stoichiometric, hypostoichiometric, and hyperstoichiometric compositions. Thermogravimetric analysis was used to monitor and verify the O/M of each composition as a function of temperature. X-ray diffraction was also used to ensure that a solid solution was maintained as a function of both O/M and secondary cation content.

A strong dependence of shrinkage rate on O/M was found for both (U,Pu)O<sub>2</sub> and (U,Ce)O<sub>2</sub> compositions. Figure 1 illustrates the shrinkage of (U<sub>0.8</sub>,Pu<sub>0.2</sub>)O<sub>2</sub> on O/M as assessed by varying the heating rate. Figure 2 plots the shrinkage rate of these data. Feedstocks of UO<sub>2</sub>, CeO<sub>2</sub>, and PuO<sub>2</sub> were used in this study; it is hypothesized that the two disparate peaks observed in Figure 2 correspond to fission reaction between the UO<sub>2</sub>-UO<sub>2</sub> particles while the second corresponds to reaction between the UO<sub>2</sub> and PuO<sub>2</sub> particles. The same behavior is observed in UO<sub>2</sub>-CeO<sub>2</sub>. It is hypothesized that O/M plays a strong role in determining the point where UO<sub>2</sub> and the secondary cation species begin to react to form a solid solution.

Further study and assessment of this phenomenon will have significant implications in the ability to control sintering and microstructure of mixed oxide transmutation fuels. In addition to the effect on sintering rate, O/M control during densification was observed to strongly affect the final density and microstructure of both (U,Pu)O<sub>2</sub> and (U,Ce)O<sub>2</sub> pellets. Sintering under hypostoichiometric conditions was observed to limit grain growth; this allows porosity to remain located on grain boundaries where

their elimination is possible. Excessive grain growth as occurs at higher O/M instead was observed to trap porosity at grain boundary interiors, thus limiting ultimate attainable densities.

### **US-France Advanced Nuclear Fuels R&D Collaboration**

Conceptual design work continued on the Americium-Bearing Blanket (AmBB) experiment planned for irradiation in the Advanced Test Reactor at INL. The concept proposes to investigate the possibility of transmuting Americium in the breeder blankets of future sodium fast reactors, and would put 10-15% Am into either depleted UO<sub>2</sub> or depleted U-Zr blanket rods. Such AmBB rods would operate in low power and low temperature regimes for extended periods of time where no performance data currently exists. At year end, the DOE-CEA agreement for this experiment in ATR had not been signed, so final design and fabrication activities have been deferred until FY18.

### **OECD-NEA Expert Group on Accident Tolerant Fuels for LWRs**

The Organization for Economic Cooperation and Development/Nuclear Energy Agency (OECD/NEA) Nuclear Science Committee approved the formation of an Expert Group on Accident Tolerant Fuel (ATF) for LWRs (EGATFL) in 2014. Chaired by Kemal Pasamehmetoglu, INL Associate Laboratory Director for Nuclear Science and Technology, the mandate for the EGATFL defines work under three task forces: (1) Systems Assessment, (2) Cladding and Core Materials, and (3) Fuel Concepts. Scope

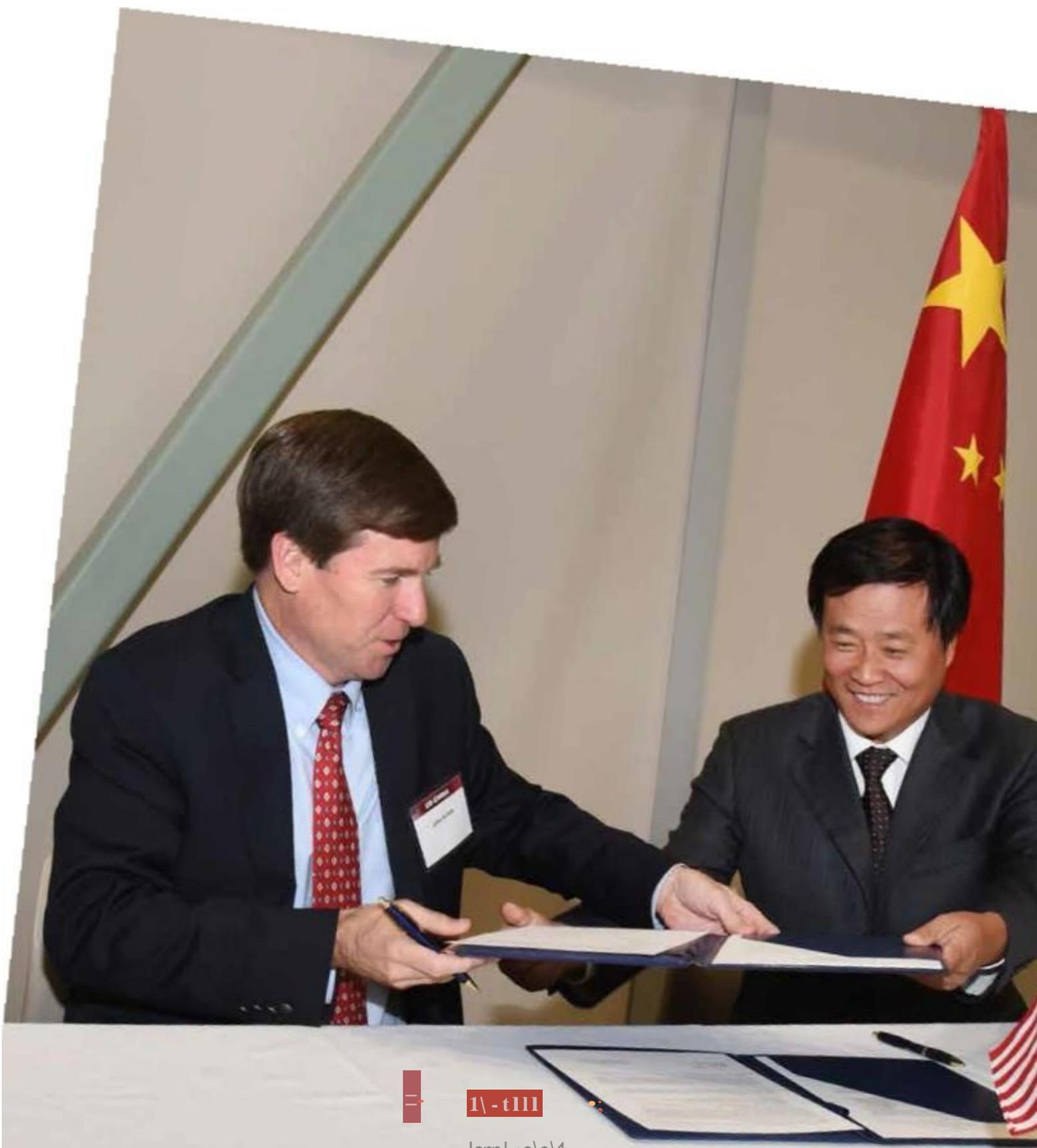
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for the Systems Assessment task force (TF1) includes definition of evaluation metrics for ATF, technology readiness level definition of illustrative scenarios for ATF evaluation, and identification of fuel performance and system codes applicable to ATF evaluation. The Cladding and Core Materials (TF2) and Fuel Concepts (TF3) task forces are working to identify gaps and needs for modeling and experimental demonstration; define key properties of interest; identify the data necessary to perform concept evaluation under normal conditions and illustrative scenarios; identify available infrastructure (internationally) to support experimental needs; and make recommendations on priorities. Where possible, considering proprietary and other export restrictions (e.g., International Traffic in Arms Regulations), the Expert Group will facilitate the sharing of data and lessons learned across the international group membership. The Systems Assessment task force is chaired by Shannon Bragg-Sitton (Idaho National Laboratory [INL], U.S.), the Cladding Task Force is chaired by Marie Moatti (Electricite de France [EdF], France), and the Fuels Task Force is chaired by Masaki Kurata (Japan Atomic Energy Agency [JAEA], Japan). The original Expert Group mandate was established for June 2014 to June 2016. In April 2016 the Expert Group voted to extend the mandate one additional year to June 2017 in order to complete the task force deliverables; this request was subsequently approved by the Nuclear Science Committee. All three task forces are expected to publish their respective deliverable reports in summer 2017.

### **IAEA Coordinated Research Project on Accident Tolerant Fuels for LWRs (ACTOF)**

The Fuel Performance and Technology Technical Working Group (FPTTWG) within the International Atomic Energy Agency (IAEA) established a coordinated research project (CRP) on ATF for LWRs (ACTOF) in 2015 (CRP-T12030). CRPs are typically initiated with a technical workshop, followed by a solicitation for proposals on potential projects under the CRP. Each CRP runs approximately 4 years, with a joint plan for the work established based on proposals submitted by various member institutes/organizations. Studies under that joint plan are typically managed through a series of consultants meetings and small contracts. A technical meeting on ATF for LWRs was initially held in October 2014 at ORNL to launch the ACTOF CRP. Focused on nuclear fuel performance and safety, the objective of ACTOF is to support options for the development of nuclear fuel with improved tolerance of severe accident conditions through experiments to acquire data on new fuel and cladding materials and modeling of new fuel designs using ATF materials. ACTOF is expected to provide information to IAEA Member States to support decision making on ATF choices and to provide data, analyses, and advanced techniques to understand and predict the integral performance of ATF designs under normal, transient, and severe accident conditions. The first Research

Coordination Meeting (RCM) on ACTOF was held in November 2015 and was attended by 14 organizations across 11 countries, including Westinghouse and Battelle Energy Alliance (BEA, with INL as the participating laboratory) in the U.S. The Westinghouse contribution to the CRP will include information associated with the design and development of U3Si2 and Un-U3Si2 composite fuel, SiC composite cladding, Ti2AlC coated Zr cladding, and SiC wrapped Zr cladding. The INL contribution will provide implementation of material models and properties for FeCrAl and U3Si2 in INL's fuel performance code BISON, validate models against experiments, perform simulations of fuel rod behavior with ATF cladding and/or fuel (under normal and accident conditions), and perform sensitivity studies on critical material properties using BISON interfaced with DOE uncertainty quantification tools. Additional participants currently include Karlsruhe Institute of Technology (KIT) in Germany, VTT Technical Research Center in Finland, A.A. Bochvar Institute (VNIINM) in Russia, Bhabha Atomic Research Centre (BARC) in India, and Korea Atomic Energy Research Institute (KAERI) in Korea. Proposals will still be accepted until the next RCM, tentatively scheduled for Sprint/Summer 2017.



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## 1.4 SHOWCASE CAPABILITIES

### Irradiated Materials Characterization Laboratory – Idaho National Laboratory

The Irradiated Materials Characterization Laboratory (IMCL) is the newest nuclear energy research facility at Idaho National Laboratory's Materials and Fuels Complex. This unique, 12,000-square-foot facility incorporates many features designed to allow researchers to safely and efficiently prepare and conduct microstructural-level investigations on irradiated fuel.

IMCL focuses on microstructural, thermal, and mechanical characterization of irradiated nuclear fuels and materials. IMCL's unique design combines advanced

characterization instruments in an environment with tight controls on vibration, temperature, and electromagnetic interference with customizable radiological shielding and confinement systems. The shielded instruments allow characterization of highly radioactive fuels and materials at the micro- and nanoscale at which irradiation damage processes occur. Enabled by its modular design, IMCL will continue to evolve and improve capability to meet the national and international user demand for high-end characterization instruments for the study on nuclear fuel and materials.

Combined with INL's advanced computer modeling techniques, this understanding will enable advanced fuel designs, and reduce the time needed for fuel development and licensing. IMCL includes a shipping bay, high-efficiency ventilation, and a monitored exhaust stack.

#### Current capabilities

- Unshielded focused ion beam (FIB) for preparing minute samples for further testing
- Unshielded Electron Probe Micro-Analyzer (EPMA) for precision quantitative composition analysis



Irradiated materials are prepared for examination right to left: the Shielded Sample Preparation Area (SSPA), the Shielded Transfer Cell (STC), the glovebox, and the fume hood

- Titan transmission electron microscope (TEM)

**Key capabilities available in Fiscal Year 2017**

- Shielded hot cell for sample preparation
- Glovebox and hood for sample preparation and facility support
- Shielded Electron Probe Micro-Analyzer
- Shielded plasma FIB for preparing block samples for 3-D reconstruction, micromechanical testing, and microscale thermal property testing
- Shielded dual Beam FIB for TEM lamella preparation



- Scanning Electron Microscopes (SEMs) for microstructural characterization
- Key capabilities available in Fiscal Year 2018

- Shielded thermal property measurement cell - Laser Flash Thermal Diffusivity - Differential Scanning Calorimetry - Thermal Conductivity Microscope
- Space for future user-defined capability



**Key capabilities available in Fiscal Year 2018**

- Shielded thermal property measurement cell - Laser Flash Thermal Diffusivity - Differential Scanning Calorimetry - Thermal Conductivity Microscope
- Space for future user-defined capability

*The focused ion beam is used to remove a micron-sized cube of irradiated fuel for microanalysis. The cube is prepared as a transmission electron microscope sample mount ready to be sent for analysis.*

## Transuranic Breakout Glovebox – Idaho National Laboratory

The transuranic breakout glovebox (TRU breakout box) was installed in the Fuel Manufacturing Facility at Idaho National Laboratory in FY16. This glovebox will provide INL with the capability to handle larger quantities of highly radioactive material and to retrieve material that is currently inaccessible due to the containers in which it is stored. Facility hazard categorizations as well as the lack of adequate radiological shielding currently limit the amount of highly radioactive material that can be retrieved and utilized. The TRU breakout glovebox provides a place wherein large quantities of high rad material may be broken down into quantities that are allowable in facilities with lower hazard categorization limits. In addition, the design of the glovebox is such that personnel performing the work have greater shielding from radiation exposure. The box contains a specially designed 3013 can opener which will allow for the retrieval of previously irretrievable material. The 3013 container is a configuration of three nested metal containers, all of which are welded closed. Since it was originally designed as a permanent storage solution it is difficult to open;



Figure 1: TRU Breakout Glovebox with 3013 can opener installed



Figure 2: TRU Breakout Glovebox with 3013 can opener installed



Figure 3: 3013 Can opener cutting the outermost container of a 3013 mock-up set



Figure 4: Inner nested can of 3013 mock-up set being removed from outer can

however, some of the radiological material once deemed waste and placed in 3013 containers is now of interest for research and development activities and it is necessary to open these containers. The TRU breakout glovebox provides an ideal place to open these types of containers and

the can opener had demonstrated the ability to open all three components of the 3013 system. The TRU breakout glovebox and 3013 can opener provide a valuable capability for material retrieval and repackaging that will open multiple areas

## Robot at the X-ray Powder Diffraction Beamline at the National Synchrotron Light Source – II, Brookhaven National Laboratory

Scientists at the U.S. Department of Energy's (DOE) Brookhaven Lab have developed an automated system for acquiring data from radioactive samples rapidly and safely at the National Synchrotron Light Source II (NSLS-II), a DOE Office of Science User Facility located at Brookhaven. NSLS-II is one of the world's most advanced synchrotron facilities, providing scientists with state-of-the-art research tools to foster new discoveries that will help power and secure the nation's future.

At NSLS-II, scientists have built the only synchrotron beamline in the world equipped with an automated system optimized for nuclear applications. With a robot that can be fully automated and remotely controlled, a new system at NSLS-II is enabling safe, unmanned, high-throughput manipulation to collect measurements from relatively large quantities of samples irradiated for years in facilities such as test reactors. These quantities can be characterized quickly, so results are statistically representative for large material databases. They can also be incorporated into material models and used for model verification—enabling scientists to address a gap in nuclear materials research and provide greater access to synchrotron data for the entire nuclear community.

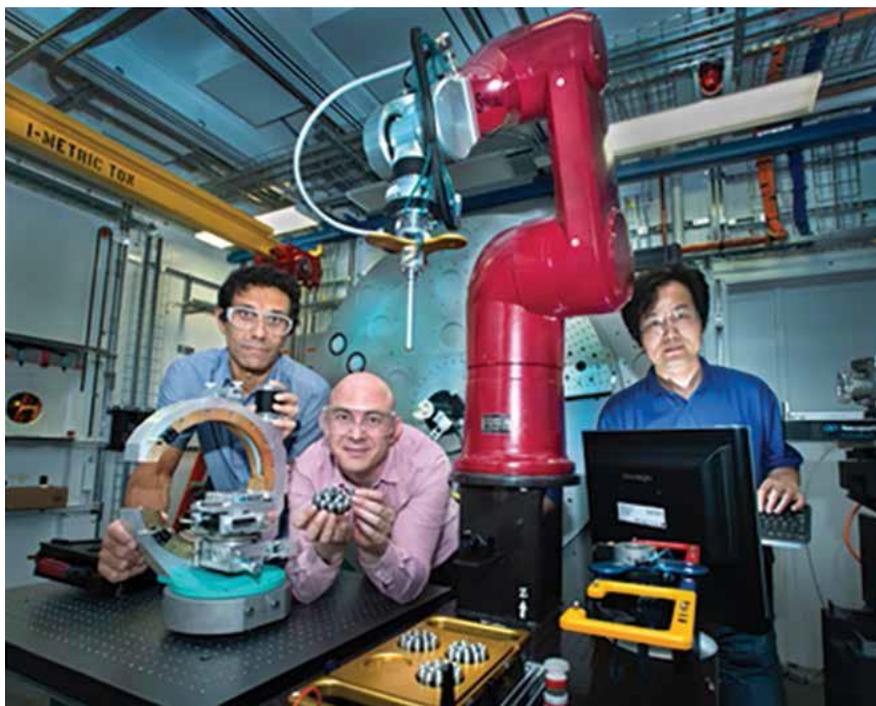


Figure 1. The red robotic arm enables safe, unmanned manipulation of relatively large quantities of samples. It places samples from a sample magazine into the beam for data collection



Figure 2. XPD beamline at the NSLS-II.



Holder with magnetic closure ring for rapid loading of radioactive samples



This radiation-shielded magazine safely holds samples for analysis

Brookhaven scientists also developed sample holders and a carousel to contain, store, and manipulate samples as well as the computer software required to exploit these capabilities. The robot picks up a sample from the carousel, exposes it to an x-ray beam as data are collected, returns it to the carousel, and then moves on to the next sample, closing all the samples in a radiation-shielded box when the work is completed.

#### Current capabilities of XPD

- Energy Range : 30 - 80 keV (tunable)
- Spatial Resolution: ~mm horizontal beam (powder averaging) down to ~20  $\mu$  m
- Energy Resolution  $\Delta E/E = 1.0 \times 10^4 - 1.0 \times 10^{-3}$
- Time Resolution: ~ms
- Intensity:  $2.5 \times 10^{14}$  photons/s/mm<sup>2</sup>
- Techniques: powder diffraction, PDF, SAXS, tomography

#### Current capabilities for radioactive materials

- High-throughput capability
- Radioactivity limit of 100 mrem/hr. @ 30cm (can have one sample or 20 samples in the magazine depending on activity)
- Scripted data collection and analysis for powder diffraction and small angle X-ray scattering experiments
- Powder diffraction can determine crystal structure, grain size, quantify lattice defects and strain in the sample
- Small angle X-ray scattering can determine size, shape and volume fraction of irradiation-induced precipitates

## Mechanical Testing at Radiochemical Processing Laboratory – Pacific Northwest National Laboratory

**R**adiochemical Processing Laboratory (RPL) is PNNL's Hazard Category II Non-Reactor Nuclear Facility and is located in the 300 area on the Hanford site, WA. RPL's mission is to provide core capability in nuclear science and technology, advancing innovative radiological material processes and solutions for environmental, nuclear energy and national security initiatives. RPL facilitates the physical infrastructure required to conduct diverse radiological research activities in multiple laboratories that house 20 gloveboxes and 17 hot cells. Current post-irradiation examination (PIE) capabilities for radioactive materials like fuels and structural materials include:

- Thermal and physical properties measurement (thermal diffusivity, UV, Raman, NMR spectroscopy)
- Radiochemical analysis (mass spectroscopy, radionuclide separation, analytical methods for determining high-concentration to trace-level analytes)
- Mechanical testing (tensile, high/low cycle fatigue, and fracture toughness)
- Microstructural characterization (OM, SEM, FIB, TEM, STEM and AFM)



*Mechanical testing is performed in MEC-2 hotcell (right) at RPL after radioactive specimens are received and prepared for testing in MEC-1 hotcell (below). This new facility is designed to carry out highly efficient mechanical testing with temperature control in inert environment.*



*Inside the test frame in MEC-2 hotcell: A cradle-type fracture testing grip is loaded on the load train (left) with the three-zone furnace opened. Two mostly used testing grips are shown below, respectively for three-point bend fracture testing and uniaxial tensile testing.*



Mechanical testing system is among the recently renewed capabilities at RPL. The key equipment for mechanical property testing is the Instron® 8801 servohydraulic testing system whose frame is installed in MEC-2 hotcell and equipped with a three-zone Mellen furnace that can heat specimens up to 1100 °C in inert environment. The test frame is a double-acting lower base servohydraulic actuator with a force capacity of ±100 kN and a usable stroke of 150 mm. Its load

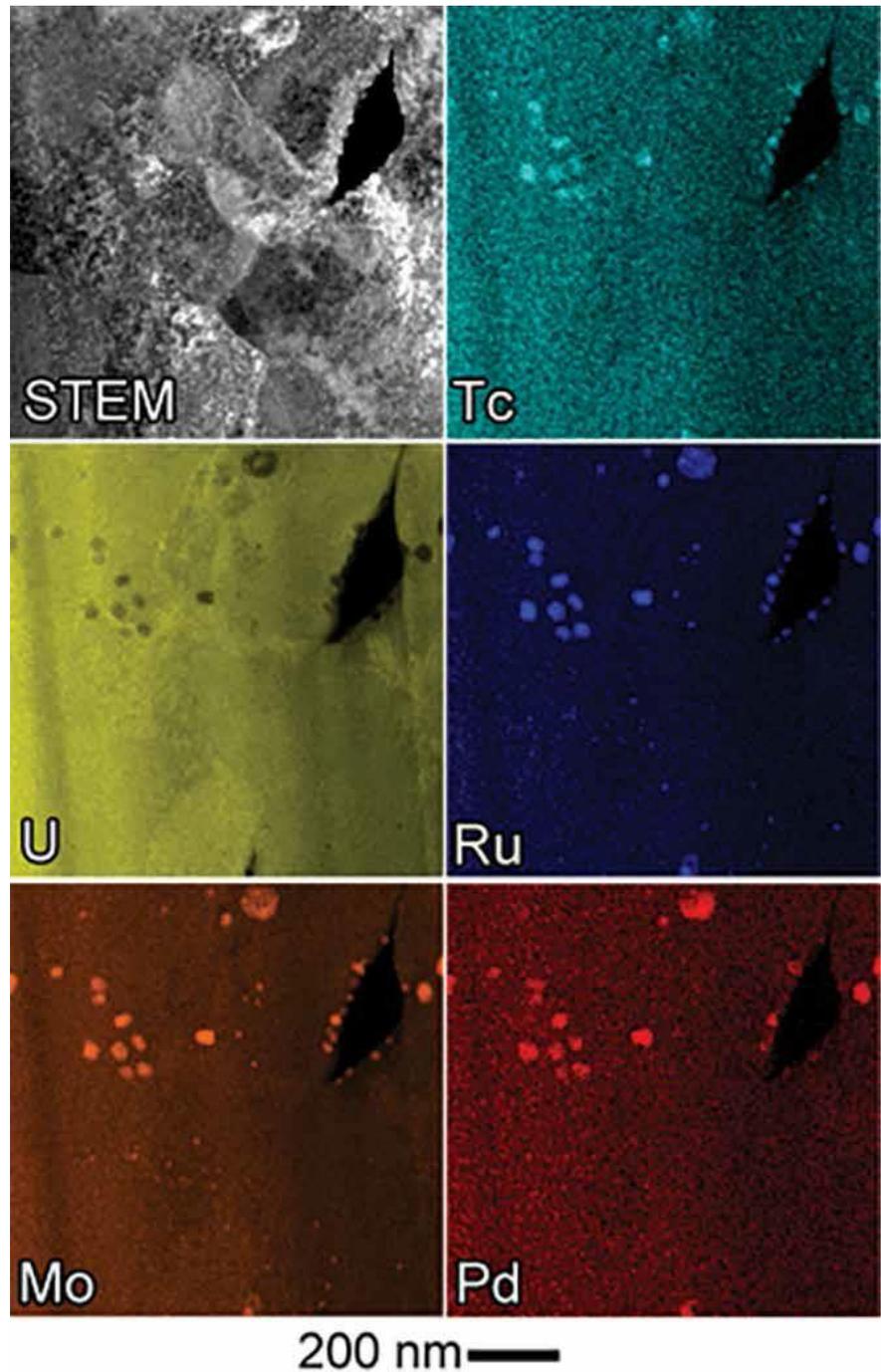
train can hold the cradle-type grips that uniquely developed for highly efficient mechanical tests, including uniaxial tensile, high and low cycle fatigue, fracture resistance (J-R), and bending tests in controlled environment. Among these testing capabilities, the procedure of static J-R fracture testing and fracture toughness calculation requires the most complicated steps and significant background knowledge. A streamlined procedure from specimen loading to fracture toughness determination has been established for the testing and evaluation of irradiated materials in the facility. In pursuit of a fast fracture toughness evaluation process, the precracking and crack growth testing steps were highly simplified, in which no external gage attachment is required, and the fracture mechanics calculation was modified correspondingly. This fracture toughness testing and evaluation technology was proven to be particularly effective for processing irradiated miniature specimens, which have been widely used for PIE of nuclear materials.

## Advanced Electron Microscopy of Irradiated Fuels and Materials – Oak Ridge National Laboratory

The FEI Talos F200X scanning/transmission electron microscope (S/TEM) is the state-of-the-art in nanochemical analysis of metals and ceramics for engineering applications. This instrument provides high efficiency collection of chemical analysis signal via X-rays from small (~1 nm) areas of specimens. This instrument is used to study the effects of irradiation on nuclear reactor structural materials, fuel cladding, and fuel materials.

ORNL scientists use the Talos to determine the changes in many different materials after irradiation conditions or other extreme environments (corrosion, loss-of-coolant accident simulations, etc.). The images on the left show metal refractory particles in high-burnup UO<sub>2</sub> reactor fuel, imaged using the Talos' unparalleled chemical mapping capabilities. Sub-10 nm precipitates are visible.

*Refractory metal precipitates in high-burnup structure in UO<sub>2</sub> irradiated in a light-water reactor.*



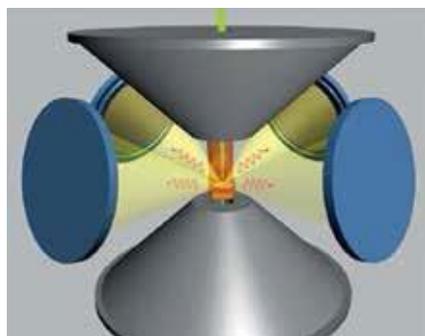
The FEI Versa3D DualBeam focused ion beam/scanning electron microscope (FIB/SEM) is a multipurpose nanoscience workstation and analytical tool. The FIB is, essentially, a “nano-sandblaster” used for high-precision nanofabrication. Using the ion beam nanofabrication, the Versa3D can produce site-specific, high-quality specimens for S/TEM and atom probe tomography. The Versa3D’s electron column provides a high-current, high-resolution probe ideal for micro and nanoanalysis, using X-ray mapping and electron backscatter diffraction.

ORNL scientists use the Versa3D tool to prepare specimens for other electron microscopes, and to determine chemical and structural changes caused by irradiation or other extreme environments.

### Capabilities

- FEI Talos F200X scanning/transmission electron microscope (S/TEM) provides 200 keV electron beams with high probe currents (>5 nA probe in 2-3 nm probe size) and can be focused to a very fine probe for atomic-resolution imaging (<160 pm resolution demonstrated at 100 pA probe current).
- The Talos’s SuperX four-sector X-ray detector captures 0.9 sr solid angle for high efficiency X-ray mapping.
- The FEI Versa3D DualBeam focused ion-beam/scanning electron microscope (FIB-SEM) tool provides <1 to 30 keV beam energies for both gallium and electron beams.

- The Versa3D is primarily used for S/TEM sample preparation from specific sites, such as grain boundaries or other defects.
- X-ray and crystallographic mapping capabilities make the Versa3D a power analytical electron microscope.



FEI Talos SuperX system



FEI Versa3D



FEI Talos F200X

## Capabilities & Characterization Techniques of Interest Provided by the Fuels Research Laboratory – Los Alamos National Laboratory

The Fuels Research Laboratory (FRL) at Los Alamos National Laboratory (LANL) is a unique facility which executes experimental research to support numerous technical areas within the Advanced Fuels Campaign. The primary focus of the FRL is development of structure-processing-property relationships for ceramic and high density fuels that support both accident tolerant fuel (ATF) and transmutation fuel development. In addition, the facility is authorized to handle enriched uranium. This has allowed the FRL to fabricate test articles for ongoing and upcoming irradiations at ATR and elsewhere.

The FRL was constructed beginning in 2008 to fill a key gap in the national infrastructure and AFC; at the time, capabilities for fundamental research on uranium ceramic fuels were extremely limited. The facility performed its first radiological operations in 2011, first enriched uranium operations in 2014, and currently provides state of the art experimental infrastructure for a team of roughly twelve students, postdocs, and staff. The facility is accessible to both U.S. citizens and foreign nationals, making it the ideal collaborative site to accommodate short term and long-term student visitors as part of this study.



*Glovebox lines in FRL used to condition feedstock*

Capabilities of the FRL were initially designed to emphasize synthesis and assessment of ceramic oxide nuclear fuels. Thermophysical properties are among the most important to the design and qualification of nuclear fuels. The thermal conductivity of a material is determined by calculating the product of the thermal diffusivity ( $\alpha$ ), specific heat capacity ( $cP$ ), and density ( $\rho$ ). Each of these parameters can be investigated experimentally through laser flash analysis ( $\alpha$ ), differential scanning calorimetry ( $cP$ ), and dilatometry ( $\rho$ ). All experimental

equipment in the FRL is capable of measurements to temperatures up to 2000C and under a wide range of atmosphere conditions. Additionally, a melt point system was developed that has been calibrated to above 3000C and possesses the ability to perform measurements in electron beam sealed crucibles to maintain the required thermochemical conditions.

Synthesis capabilities extend from basic oxide processing through oxide derivative routes (e.g. carbothermic reduction - nitridation for conversion of  $UO_2$  to  $UN$ ), and are augmented

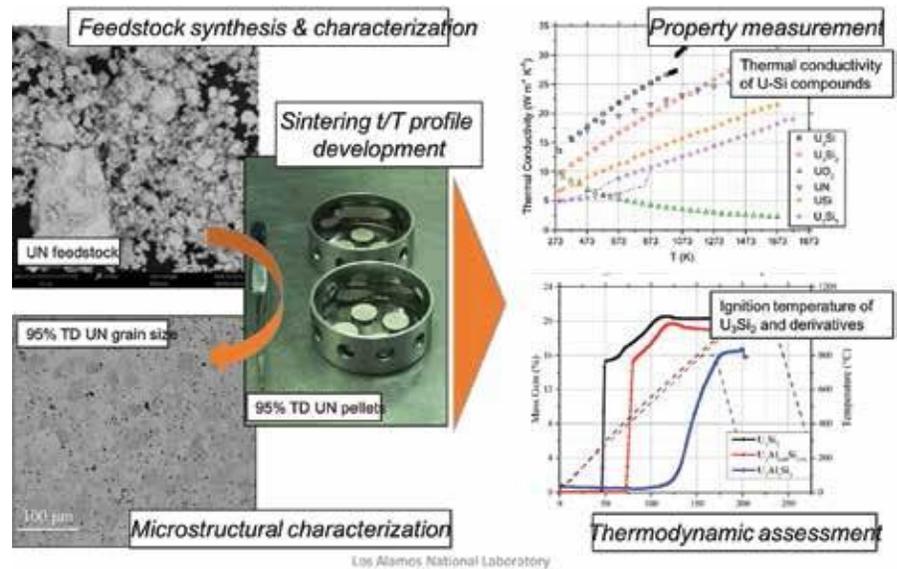
by a dedicated radiological arc melter contained within a high purity glovebox. The arc melting capabilities facilitate production of uranium alloys and compounds using metal source constituents. Arc melting has been used under AFC to produce U-Si compounds of interest as ATF fuels and constituents.

Expertise of FRL staff has also expanded beyond AFC fuels work to include exploration of the behavior of ATF cladding materials. Assessment of the effects of temperature and thermodynamic stimuli (e.g. steam, hydrogen) on cladding materials requires expertise in a number of areas parallel to those needed for fuel material synthesis and characterization including atmosphere control and high resolution electron microscopy.

The FRL continues to expand its capabilities to meet the myriad challenges of ATF qualification. Graduate students and postdoctoral staff constitute a large fraction of FRL staff, and are dedicated to executing world-class academic research to advance our understanding of nontraditional nuclear fuels.



Enriched UN / U<sub>3</sub>Si<sub>2</sub> composite fuel pellets produced at FRL for ATF-1



#### Fuels Research Laboratory Capabilities

#### Current capabilities (annual throughput ~50 kg of U-238, ~5 kg of U-235),

- Inert atmosphere glovebox three-stinger arc melter use for synthesis of uranium alloys
- Powder feedstock conditioning and characterization capabilities (mills, particle size analyzers surface area analyzers)
- Manual and automated pellet presses
- Inert atmosphere metallography preparation
- Controlled atmosphere X-Ray diffraction and scanning electron microscopy
- Metal element sintering furnaces capable of 2200C under inert, vacuum, or reducing atmospheres
- Shielded element furnaces capable of 1800C used for sintering under oxidizing or controlled oxygen activities

- Full suite of thermophysical property measurement equipment include laser flash analyzers, dilatometers, calorimeters)
- Thermogravimetric analyzers capable of measuring relevant reaction kinetics of fuel or cladding materials under steam, hydrogen, oxygen, or other environments up to 2400C
- High load and low load nanoindentation (static and continuous force capabilities) used to study basic deformation behavior of novel fuel materials
- Resonant ultrasound spectroscopy used to provide basic elastic properties of novel fuel materials



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# ADVANCED LWR FUEL SYSTEMS

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- 2.1 Accident Tolerant Fuels
- 2.2 High-Performance LWR Fuel Development
- 2.3 Analysis
- 2.4 ATF Cladding and Coatings
- 2.5 Irradiation Testing and PIE Techniques
- 2.6 Transient Testing

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## 2.1 ACCIDENT TOLERANT FUELS

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### Evaluation of ATF Concepts

*Principal Investigator: Shannon Bragg-Sitton*

*Collaborators: Jon Carmack*

The overall goal of ATF development is to identify alternative fuel system technologies to further enhance the safety, competitiveness, and economics of commercial nuclear power. The complex multi-physics behavior of LWR nuclear fuel in the integrated reactor system makes defining specific material or design improvements difficult as such, establishing desirable performance attributes is critical in guiding the design and development of fuels and cladding with enhanced accident tolerance.

The proposed technical evaluation approach and associated metrics were compiled and released in 2014 in the “Light Water Reactor Accident Tolerant Fuel Performance Metrics” report [1]. A summary of the ATF metrics was published in a technical journal article in FY16 [2], including addition of proposed weighting factors for each performance regime and fuel system attribute. These weighting factors were developed via coordination with the ATF Industry Advisory Committee (see xxx for further details) and were reviewed by the Independent Technical Review Committee that was convened in FY16.

The proposed technical evaluation methodology is intended aid in the optimization and prioritization of candidate ATF designs. Detailed evaluation of each concept will gauge its ability to meet performance and safety goals relative to the current UO<sub>2</sub> - zirconium alloy system and relative to one another. This ranked evaluation will enable the continued development of the most promising ATF design options given budget and time constraints, with a goal of inserting one (or possibly two) concepts as an lead fuel rod or assembly in a commercial LWR by 2022.

The Technical Review Committee (TRC) was organized to provide an independent assessment of the technology feasibility for near term research and development of candidate ATF design concepts and prioritization of those concepts [3]. Established in late 2015, the TRC was comprised of technology experts selected based on their knowledge of the technologies under review, reactor operations, and fuel fabrication plant operations. The cross-section of experts includes experience in

the areas of materials (metals and ceramics), neutronics, thermal-hydraulics, and severe accidents to enable assessment of the technology feasibility for near-term development of the ATF design concepts. TRC members include Carter “Buzz” Savage, Thomas Galioto, John Guerci, Dick Hobbins, Jim Lemons, Regis Matzie, Larry Ott, and Steve Zinkle. The review of ATF concepts proposed by industry and national laboratories was held January 2016 in Washington D.C. The TRC was tasked with independent assessment of technology feasibility for near term research and development of candidate ATF design concepts and prioritization of those concepts but will also provide input to prioritization of concepts requiring longer-term development.

Input from the TRC was provided to the Department of Energy to provide input to selection of industry teams and concepts for Phase II research and development work.

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## ATF Industry Advisory Committee

*Committee Chair: Bill Gassmann, Exelon*

*Collaborators: Shannon Bragg-Sitton and Jon Carmack*

The Advanced Light Water Reactor Fuel Industry Advisory Committee (IAC) was established in 2012 to advise AFC's National Technical Director (NTD) on the development and execution of a program focused on advanced fuels for light water reactors. The IAC is comprised of leaders from the commercial light water reactor industry. They represent the major suppliers of nuclear steam supply systems, owners / operators of U.S. nuclear power plants, fuel vendors, and the Electric Power Research Institute. Members are selected on the basis of their technical knowledge of nuclear plant and fuel performance issues as well as their decision-making positions in their respective companies.

The IAC meets monthly via teleconference and in a face-to-face meeting once a year. The IAC met in November 2015 in Washington D.C. at the Tennessee Valley Authority (TVA) offices. Progress toward the Technical

Review Committee prioritization of successful concepts was discussed at the November 2015 meeting. In particular, the IAC provided input to the weighting factors recommended for use in applying the technical performance metrics to evaluation of ATF concepts.

Following the TRC prioritization in January 2016, Committee work focused on more detailed discussions of technical aspects and funding associated with the concepts to be pursued going forward. Specifically, future needs for Phase 2 test reactor irradiation were considered, and utility input was sought on the funding challenges which are expected to arise following the TRC prioritization of concepts and selection of Phase 2 industry awards by DOE. Contacts were established with the Nuclear Regulatory Commission (NRC) and Institute of Nuclear Power Operations (INPO) regarding the IAC charter and planned near

term activities. Both organizations are standing by to become more involved with the IAC at the appropriate time. In addition, new utility members joined the IAC in FY16 following entry into Phase 2. In general, utility leaders have begun to take on a greater level of interest in ATF as commercialization discussions become more detailed.

The IAC is currently chaired by William Gassmann of Exelon Corporation. Additional members represent BWXT, Westinghouse Electric Company, AREVA, Global Nuclear Fuels, EPRI, Dominion, Duke Energy, and Southern Nuclear.

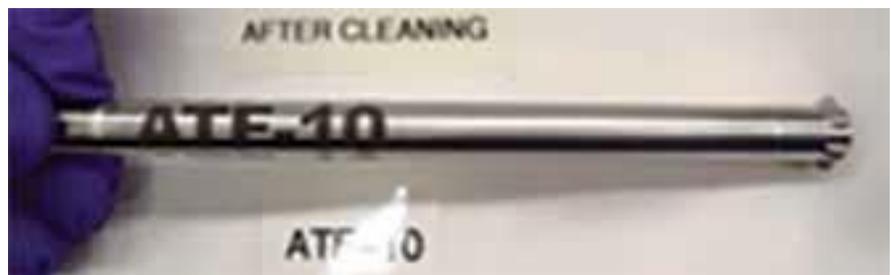
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## ATF INDUSTRY TEAMS – Westinghouse Electric Company

Principal Investigator: Edward J. Lahoda

Collaborators: Westinghouse Electric Company LLC, General Atomics, Massachusetts Institute of Technology, Idaho National Laboratory, Los Alamos National Laboratory, Southern Nuclear Operating Company and Exelon Nuclear, Argonne National Laboratory and Ceramic Tubular Products, Pennsylvania State University, University of Wisconsin, Argonne National Nuclear Laboratory (United Kingdom), Paul Scherrer Institute (Switzerland), Institute for Energy Technology (Norway), United Technologies Research Center and Ceramic Tubular Products

Figure 1. U<sub>3</sub>Si<sub>2</sub> Fueled Rodlet Now In ATR



The overall objective of this program is to introduce accident tolerant fuel (ATF) lead test rods and assemblies (LTR/LTA) for SiC and coated zirconium cladding with U<sub>3</sub>Si<sub>2</sub> fuel into a commercial reactor by 2022. The objective of the current Phase 1b work is to design, test and build using commercially scalable technologies test articles for up to 6 year-long exposure at PWR conditions of prototypical ATF fuel rodlets. The data from this 6-year test reactor exposure and test evaluation will be used as the basis to license and load LTRs into commercial reactors in 2019 and LTAs in 2022.

### Project Description:

The technical objectives of this program are:

- Fabrication of representative test fuel rodlets consisting of SiC and coated zirconium cladding with U<sub>3</sub>Si<sub>2</sub> fuel
- Development of an ATF LTR/LTA project plan
- Generation of data supporting commercial feasibility
- Determine the expected behavior of the ATF options in LOCA/station blackout scenarios
- Determine the feasibility of licensing a commercially feasible ATF

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### Accomplishments:

A coated cladding has been successfully tested in the autoclaves at the Westinghouse Churchill site and in the Massachusetts Institute of Technology reactor and are currently undergoing final testing. SiC cladding development has continued. Autoclave testing of open-ended SiC tube samples has completed and testing of closed-ended tube samples will begin in November in order to finalize the design before fabricating rodlets for exposure in the Advanced Test Reactor (ATR) and Halden reactor (Institute for Energy Technology) in 2017 and 2018 respectively. The coated cladding is cold sprayed Cr on zirconium alloy tubes produced by the University of Wisconsin. The SiC cladding is a composite SiC with an outer monolith produced by General Atomics..

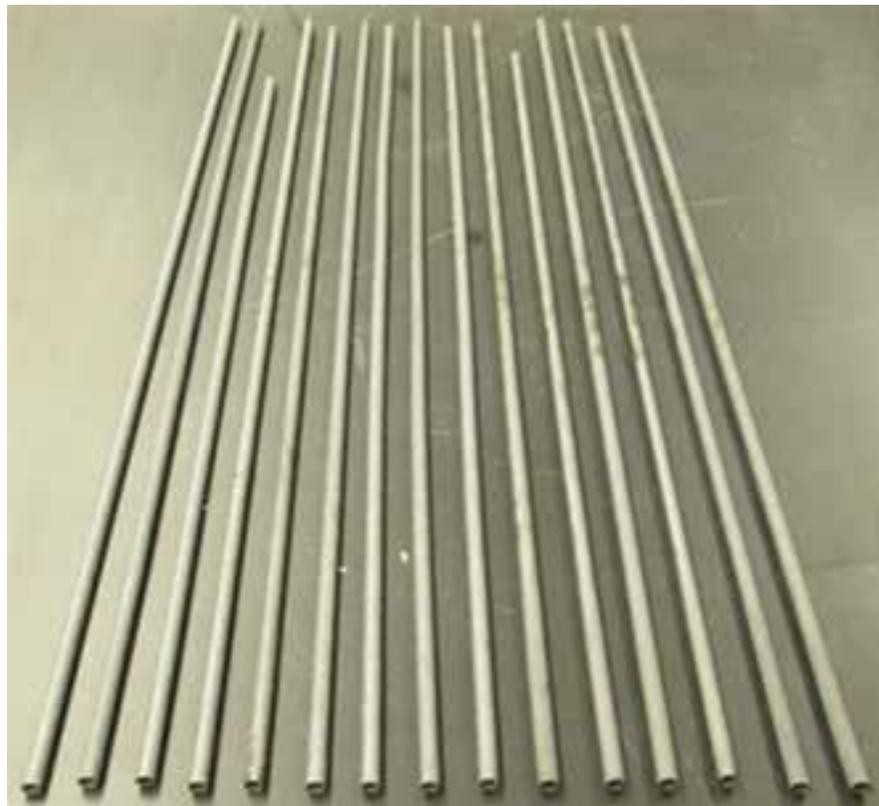
Ceramic Tubular products has developed a continuous process for doing chemical vapor infiltration (CVI) of



*Figure 2. Cr Coated Zirconium Alloy Tube Using Cold Spray*

SiC within the SiC composite windings and chemical vapor deposition of SiC on the surface of the composite. Argonne National Laboratory (ANL) has been working on Atomic Layer Deposition approaches for depositing SiC to replace CVI in order to improve the crystallinity of the final SiC composite product. ANL has also explored the deposition of zircon ( $ZrSiO_4$ ) as a corrosion resistant coating for both SiC composites and zirconium alloys. United Technologies

*Figure 3. One Meter Long SiC Tubes*



Research Center has been exploring methods for making composites utilizing SiC particles and SiC pre-polymer compounds to replace the CVI process.

The effect of irradiation on the carbon interface layer and its effect on the thermal conductivity of SiC composite has been studied by the Paul Scherrer Institute in Switzerland. Their work points out the need for alternate interface layer compounds that are not as reactive with neutron irradiation.

The Idaho National Laboratory has produced U<sub>3</sub>Si<sub>2</sub> pellets that were loaded into six, 6-inch fuel rodlets which were then loaded into the ATR to initiate testing of this new fuel form. This next generation fuel provides a 17% increase in uranium 235 density without exceeding the current 5% U<sub>235</sub> enrichment limit for commercial nuclear fuel. This increase in U<sub>235</sub> provides significant economic benefits for ATF as well as enabling the



*Figure 4. UN/U<sub>3</sub>Si<sub>2</sub> Pellets  
Manufactured by LANL*

adoption of advanced ATF cladding due to its very high thermal conductivity (~5x more than UO<sub>2</sub>). The Los Alamos National Laboratory produced composite U<sub>3</sub>Si<sub>2</sub>/UN fuel pellets which have also been put into the ATR in the form of six, 6 inch rodlets in zirconium alloy cladding. These high density pellets are a next-next generation fuel with an even higher U<sub>235</sub> content (~25% increase over UO<sub>2</sub>) with an even higher thermal conductivity.

The National Nuclear Laboratory in the United Kingdom has been experimenting with various approaches to manufacture U<sub>3</sub>Si<sub>2</sub> from either UF<sub>6</sub> or UF<sub>4</sub> as an alternative to the current route which starts with U and Si metals. Development of a route from UF<sub>6</sub> or UF<sub>4</sub> would significantly reduce the cost of U<sub>3</sub>Si<sub>2</sub> manufacture and minimize the investment required for the reconversion area of the current nuclear fuels plants.

Southern Nuclear Operating Company and Exelon Nuclear have been evaluating the economic gains from using ATF in current nuclear plants. Potentially significant gains have been identified and are being quantified using probability risk assessment techniques by Westinghouse and accident scenario modeling by Fauske and Associates.

## ATF INDUSTRY TEAMS – AREVA

Principal Investigator: Kiran Nimishakavi

Collaborators: University of Wisconsin, University of Florida, Savannah River National Laboratory (SRNL), Electric Power Research Institute (EPRI) and utility members from Duke Energy, Dominion and the Tennessee Valley Authority (TVA)

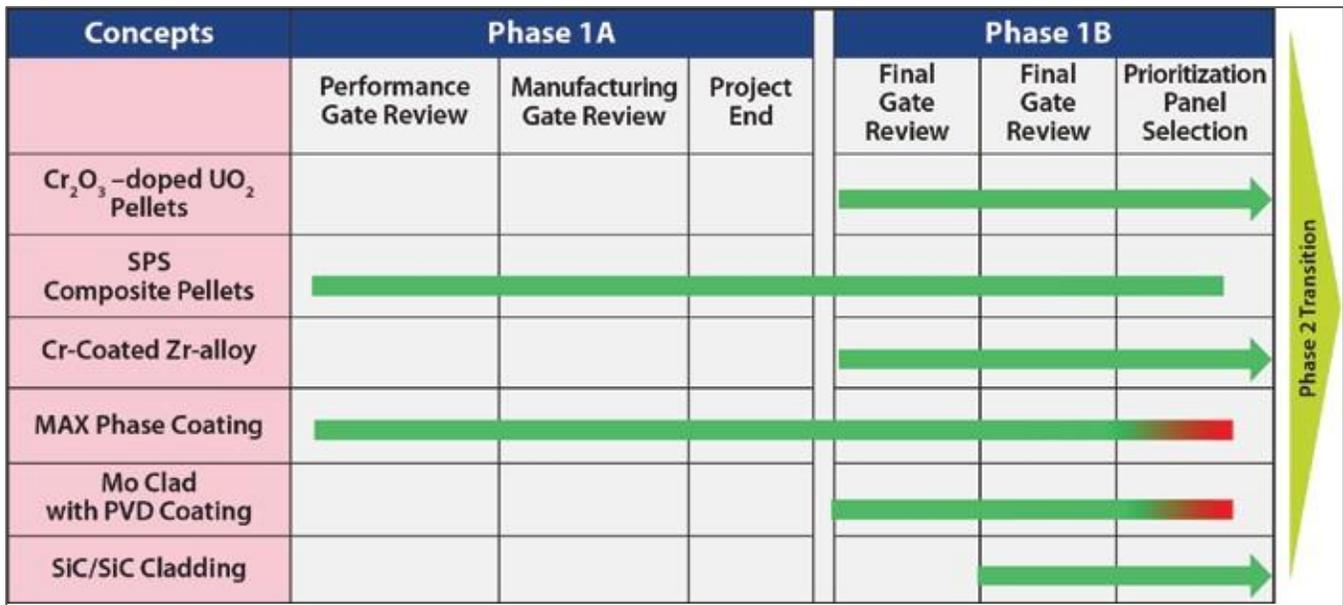


Figure 1. AREVA- EATF gate review summary

In response to the Department of Energy (DOE) funded initiative to develop and deploy lead test assemblies (LTAs) of Enhanced Accident Tolerant Fuel (EATF) into a US reactor within 10 years, AREVA has put together a focused team to develop promising technologies for improved fuel performance. Composite fuel pellets designed for higher thermal conductivity were fabricated by University of Florida (UF) using Spark Plasma Sintering process. Several types of protective metallic and ceramic coatings were developed and tested by University

of Wisconsin (UW) and Savannah River National Laboratory (SRNL). Molybdenum cladding with an outer coating was evaluated by Electric Power Research Institute (EPRI). In the 4th quarter of 2016, AREVA integrated its global EATF concepts into the DOE Program. These include chromia doped pellets, chromium coated cladding and silicon carbide sandwich cladding (SiC-SiC).

### Project Description:

One key aspect of developing a credible EATF concept is to perform extensive testing to ensure that the candidate materials meet the desired structural integrity and reliability



requirements for service as LWR fuel. During the Government Fiscal Year (GFY) 2016, AREVA team continued the development and testing of the EATFMax Phase clad coatings and pellet additives (SiC and nano-diamond) with a primary focus on understanding their behavior during normal operation and accident conditions. The AREVA Team used the gate review process to evaluate the concepts against the criteria developed by a broad group of industry experts. This process allowed for a thorough independent multi-discipline review of each concept, centered and aimed at meeting the DOE' s goal of Lead Test Rods (LTRs)/LTAs in a commercial reactor by 2022. As a result of the AREVA gate review process and the subsequent DOE technical review

committee recommendation, AREVA will pursue further development and testing of two fuel systems. Figure-1, show the results of gate reviews and down-selection process.

The ultimate goal of DOE EATF program is to develop an improved and more robust nuclear fuel design that will mitigate the consequences of reactor accidents maintaining or improving on the current fuel operational and economic performance. As part of Phase 1 program, the AREVA team' s has investigated unique fuel concepts and developed a comprehensive test program which will provide the performance results necessary to support DOE' s goal of EATFLTR/LTA insertion by 2022.

*Figure 2. Cr<sub>2</sub>O<sub>3</sub>-Gd<sub>2</sub>O<sub>3</sub>  
– Chromia Doped Pellets  
Manufactured in Erlangen.*



*Figure 3. 19.5 inch Long Zr-4 Tubes coated with chromium*

#### **Accomplishments:**

In GFY 2016, AREVA has demonstrated the fabrication of chromia-doped UO<sub>2</sub> and UO<sub>2</sub>-Gd<sub>2</sub>O<sub>3</sub> pellets in AREVA's Material Test Laboratory (See Figure-2). These pellets are currently being irradiated in lead test assemblies in a U.S. commercial reactor. AREVA is currently implementing full-scale production capability at our Horn Rapids Road (HRR) fuel manufacturing facility Richland, WA.

The composite fuel pellet development (UO<sub>2</sub>-SiC and UO<sub>2</sub>-nano-diamond) continued with UF during Phase 1B. The pellet production process parameters were refined during the optimization efforts resulting in improved surface quality, theoretical density, and dimensional stability. Six test rodlets including composite pellets were placed into the ATF-1 test apparatus in INL's Advanced

Test Reactor (ATR). The first set of three rodlets was removed in the 1st quarter of GFY 2016, at a burnup of ~8 GWd/mtU. The radiographs of the irradiated rodlets revealed that the SPS pellets were in good dimensional condition. Detailed PIE will focus on evaluating the irradiation effect on thermal conductivity of the pellets.

Extensive mechanical and corrosion tests were performed on the chromium coated cladding. Encouraging results indicate that this cladding concept could potentially be implemented with minimal changes to the current fuel rod designs. Figure-3 shows a photograph of chromium coated Zircaloy tube. The next phase of the AREVA development plan is to develop equipment and procedures capable of coating full-size fuel rods.

SRNL and UW continued to explore physical vapor deposition (PVD) coating methods with the objective of



**Bare Zr Tube**



**Zr-Si-C coated Zr tube**

producing protective MAX phase coatings on the Zircaloy-4. High-quality coatings were achieved through Physical Vapor Deposition - Sputtering (PVD-S) process. Over 17 coatings/methods were evaluated including Ti and Zr, Al-C based MAX phase coatings and Zr and Nb, Si-C based MAX phase coatings. In an attempt to create a thin layer crystalline structure on the coating, a laser “ordering” process was performed on the top surface of the PVD coating (see Figure-4). However, further research on the addition laser ordering process is needed to prevent cracking of coating during high-temperature steam testing.

Much of the work with molybdenum cladding has focused on improving the diametral properties of the cladding with optimized heat treatments. The molybdenum response to heat treatment has shown variation in mechanical properties from lot to

lot. Cathodic Arc PVD process was evaluated to apply FeCrAl coatings. However, desired final coating chemistries were not achieved with the coating process. The next phase work focused on the Hot Isostatic Press (HIP) process using concentric tubes of FeCrAl or zirconium with molybdenum. Co-extrusion of a composite tube was also considered.

Extensive out-of-pile testing has been performed on SiC/SiC composite cladding. The key finding of this work highlights an excellent retention of the mechanical behavior after exposure to LWR water. The low sensitivity of SiC/SiC composites under the high-temperature steam conditions was also confirmed. The preliminary results are positive warranting continued research to develop LTAs.

*Figure 4. Laser surface treatment of bare Zr-4 tube and Zr<sub>2</sub>SiC Coating*

## ATF INDUSTRY TEAMS – General Electric

General Electric: Raul Rebak

Contributors: Kurt A. Terrani (ORNL), Kevin Ledford (GNF), Russell E. Stachowski (GNF), Russ M. Fawcett (GNF), Jonas Gynnerstedt (Sandvik), William P. Gassmann (Exelon), John Williams (Southern Nuclear).

Figure 1. Sandvik Materials Technology made APMT tubes 9.5 mm OD and 0.8 mm wall thickness.



The General Electric (GE) accident tolerant fuel (ATF) design concept utilizes a FeCrAl alloy material as fuel rod cladding in combination with uranium dioxide (UO<sub>2</sub>) fuel pellets currently in use, resulting in a fuel assembly that leverages the performance of existing/current LWR fuel assembly designs with improved accident tolerance. The use of FeCrAl cladding is a direct near term path to improve the safety of operating commercial light water reactors.

### Project Description:

The overall goal of the U.S. Department of Energy Accident Tolerant Fuel Program for LWRs is to identify alternative fuel system technologies to further enhance the safety, competitiveness and economics of commercial nuclear power. Fuel designed for use in the current fleet of commercial LWRs reactors with enhanced accident tolerance would endure loss of cooling in the reactor core for a considerably longer period of time than the current fuel systems while maintaining or improving performance during normal operations.

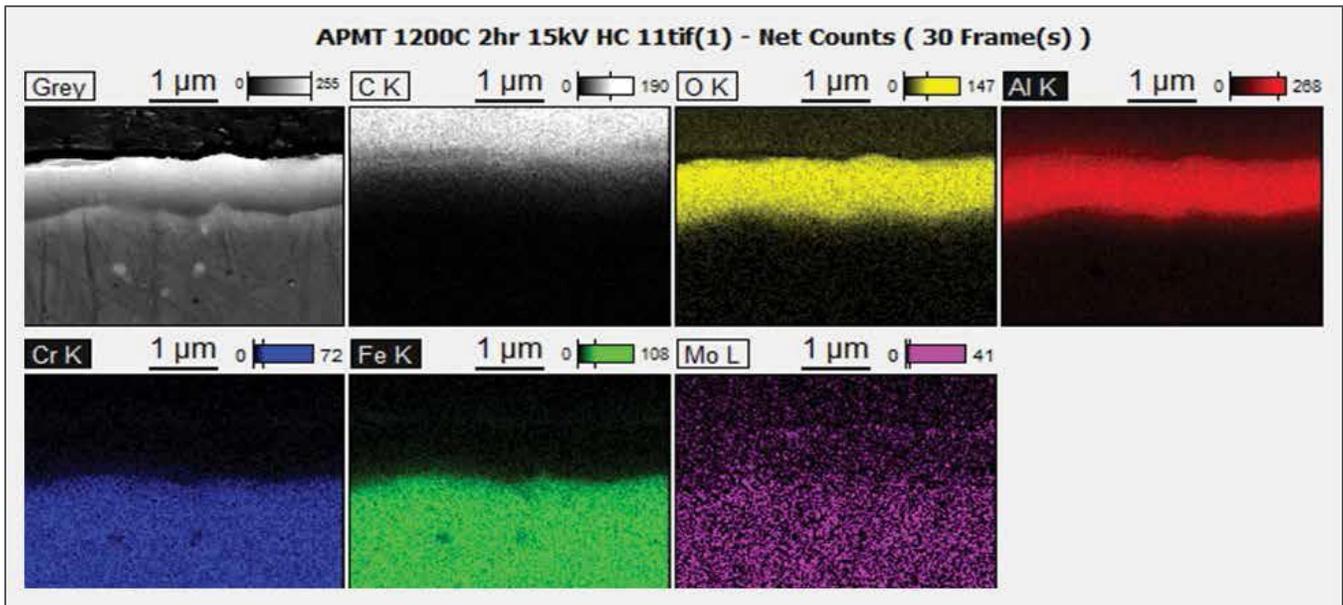
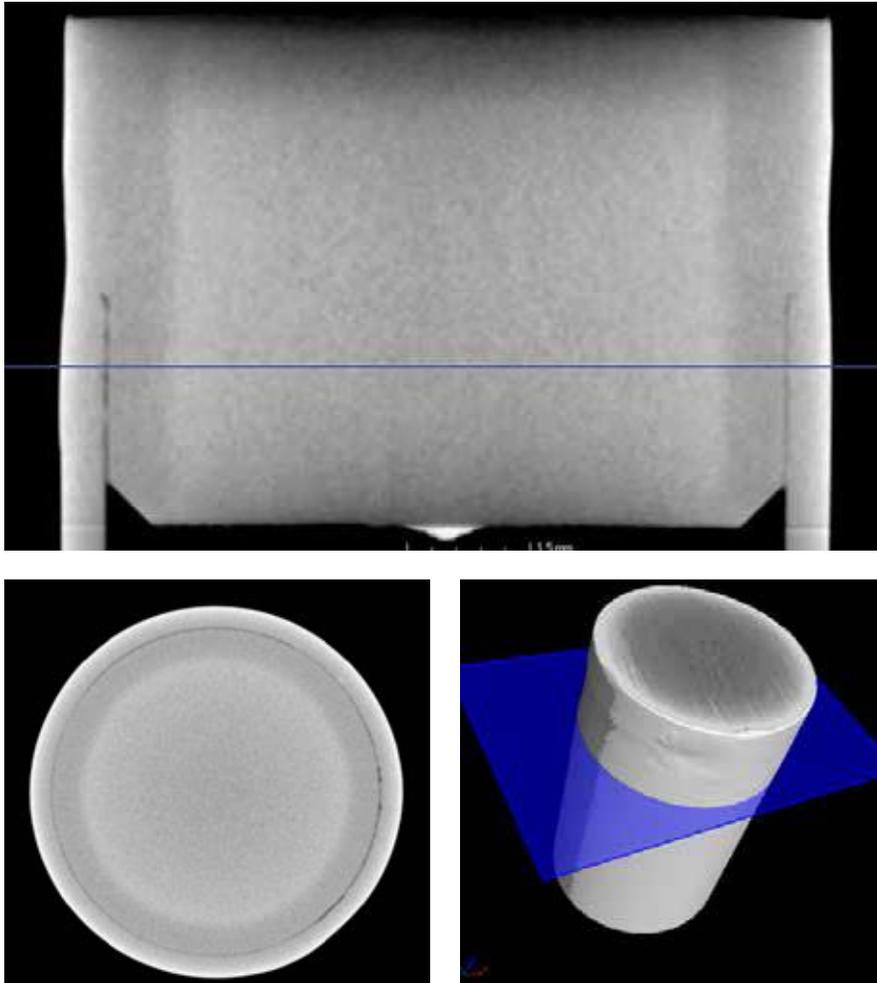


Figure 2. Heat treating APMT for 2 h at 1200°C in steam develops a 1 µm thick surface layer of alpha alumina which is a barrier for hydrogen permeation across the thickness of the cladding wall.

Under normal operation conditions in LWRs, the FeCrAl alloys offer outstanding corrosion resistance similar to current well known materials such as type 304SS or nickel alloys. Under accident conditions, FeCrAl alloys are orders of magnitude more resistant to reaction with superheated steam than zirconium, therefore generating negligible combustible hydrogen and heat of reaction. FeCrAl alloys would keep their coolable geometry for longer time allowing for quenching measures during the coping time. On the less favorable side, the FeCrAl alloys are less trans-

parent to neutrons than zirconium alloys which impacts fuel cycle cost and electricity generation costs, and they may release more tritium to the coolant. Both of the adverse attributes can be minimized or eliminated by design, fabrication and regulatory modifications.

FeCrAl clad fuel rods with UO<sub>2</sub> fuel have reasonably demonstrated capabilities to meet or exceed current fuel design technical requirements while providing increased safety



*Figure 3. Micro CT scanning of APMT cap GTAW welded on APMT tubing showing a final weld free of distortion, porosity or cracks. Several methods can be used to weld FeCrAl alloys.*

benefit during design basis events and severe accident conditions. The use of a FeCrAl alloy fuel cladding is the simplest, most cost effective and expeditious way to implement an ATF fuel design that combines the high performance of current fuel with significant accident tolerance, which will increase the safety of the current commercial reactors.

**Accomplishments:**

Fabrication is not identified as a key technical challenge since scoping studies indicated that traditional metal processing (such as pilgering, drawing, extruding, and welding) are applicable to FeCrAl materials. Although regulatory changes will be required to implement FeCrAl within a commercial product, the scope of potential changes is relatively minor and such changes will, in general, provide additional margin for safe nuclear fuel operations. FeCrAl alloys such as APMT can be produced in long thin walled tubes. Sandvik Materials Technology was able to produce APMT tubes which are 9.5 mm OD and 0.8 mm wall thickness. FeCrAl alloys can be welded by several methods, including gas tungsten arc welding, pressure resistance welding and laser welding. The use of FeCrAl cladding is compatible with large-scale production needs (material availability, fabrication techniques, waste, etc.). Iron, chromium and aluminum are common inexpensive elements. The use of FeCrAl tubes is compatible with quality and uniformity standards.

***The simple proposal from General Electric and its partners is to substitute one cladding material for another, use FeCrAl to replace the current zirconium alloy materials. The entire coolable geometry within the reactor will remain the same. The use of FeCrAl as cladding will increase the coping time in case of severe accidents and will decrease the cost of plant operation during normal conditions.***

Data is not currently available to assess the mechanical strength of FeCrAl at the end of life irradiated conditions (i.e., for an irradiation level of 15 dpa). However, it is expected that the mechanical properties, such as strength will increase and ductility will decrease, similarly to 304SS and other metallic alloys. Due to the ferritic or bcc nature of FeCrAl, it is expected that they will be more resistant to irradiation damage than some of the austenitic materials currently in use in commercial reactors for several decades. Such changes in properties can be anticipated and beginning-of-life properties will conservatively bound LFA/LRA applications until characterization is performed in future FeCrAl development tasks. Proton irradiation studies performed at the U. of Michigan showed that FeCrAl materials are resistant to proton irradiation induced cracking providing additional confirmation of the potential acceptability of FeCrAl materials for fuel rod cladding.

Another issue investigated during FY2016 was tritium release into the coolant. FeCrAl alloys do not react with hydrogen in the manner as zirconium alloys do forming hydrides. Therefore, hydrogen (tritium) may diffuse across the wall thickness of the cladding from the fuel side to the coolant side. It was found at GE GRC that a heat treatment at 1200C for 2 h will develop a one micro meter thick alpha alumina layer, which will effectively minimize hydrogen diffusion across the cladding wall.

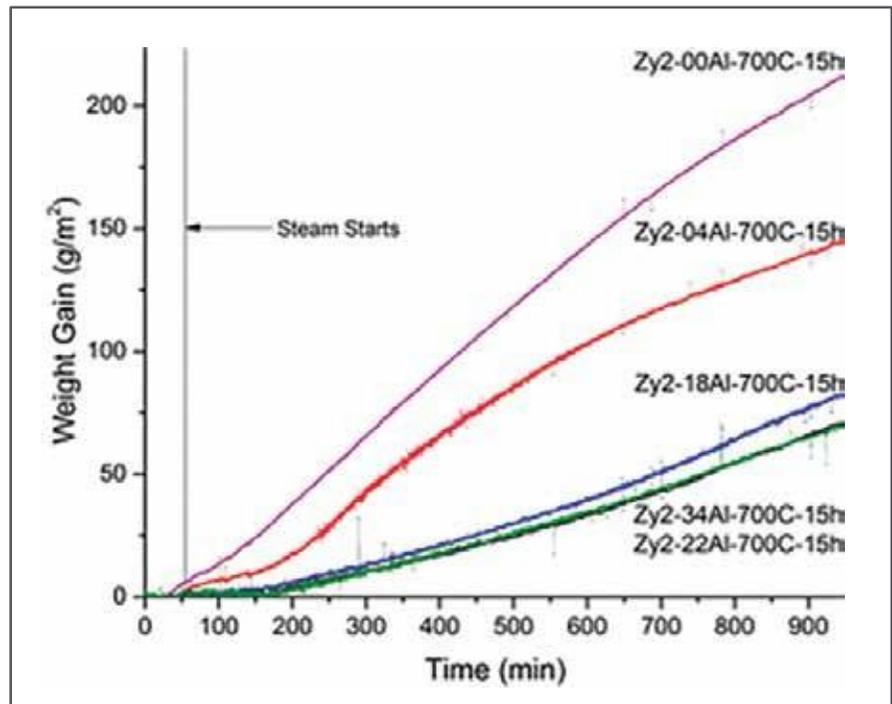
## UNIVERSITY-LED TEAMS – UNIVERSITY OF ILLINOIS

### DOE NEUP IRP 12-4728 Engineered Zircaloy Cladding Modifications for Improved Accident Tolerance of LWR Fuel

Principal Investigator: Brent J. Heuser

Collaborators: Tomasz Kozłowski, Rizwan Uddin, James F. Stubbins, Dallas R. Trinkle, Robert S. Averbach, Thomas J. Downar, Gary S. Was Yong Yang, Simon R. Phillpot, Piyush Sabharwall, Michael V. Glazoff, Jason D. Hales

Figure 1. Normalized 700C steam weight gain of various FeCrAl coatings deposited on Zircaloy-2. Increasing the aluminum concentration is seen to reduce the kinetics.



The zirconium alloys found in current US reactors exhibit extremely poor resistance to high temperature steam present in LOCA conditions. The rapid oxidation of these metals degrades their structural properties, introduces large amounts of exothermic heat, and produces a significant quantity of hydrogen gas. Modifying an alloy by applying an oxidation resistant coating is a technique that has been successfully used

in many industries to mitigate this type of adverse reaction. Applying this to Zircaloy will allow for the inheritance of its well tested bulk material properties, while enhancing the environment facing surface properties. One candidate material for a protective outer coating is alumina forming FeCrAl alloys, which are well known for their oxidation resistance. This research focuses on the development and testing of a FeCrAl coating on Zircaloy.

### **Project Description:**

The efficacy of FeCrAl coatings relies on several factors. The main focus is the identification of a range FeCrAl compositions which exhibit optimum oxidation resistance when deposited to thicknesses on the order of microns on Zircaloy. This work encapsulated the coating growth, adherence, and performance in high temperature steam. Extensive characterization of the coating, and coating-clad interactions during each step of this processes through the use of XRD, SEM, TEM, and AES garnered in-depth understanding of the physical mechanisms present in each stage. With this knowledge, a single composition was selected for exposure to normal operating conditions as well as proton and heavy ion irradiation to confirm the coatings reliability during standard operation. Modeling the effects of the coating was coupled with experimental results to understand the thermal and neutronic effects of this new layer on the reactor. The neutronics analysis ties strongly into the selection of a composition by informing how the neutron population, and therefore fuel lifetime is effected by the atomic ratios, as well as coating thickness. The experimental and computational results regarding this new parameter space provides key

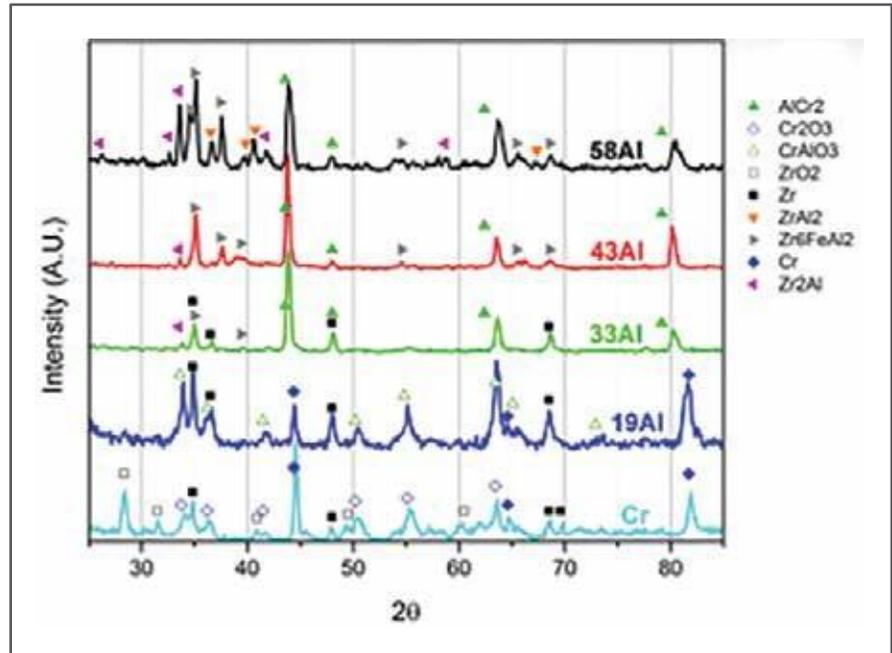
information needed for an economically informed selection of a composition from the identified safe ranges. Additional experimental research into the high temperature steam oxidation of binary alloys was conducted to further enhance our knowledge of the behavior our current alloys in accident scenarios. Alternative CrAl and Cr coatings have also been investigated.

### **Accomplishments:**

At the University of Illinois Urbana/Champaign (UIUC) Chromium-Aluminum and Chromium coatings on Zircaloy-2 with thicknesses of about 1um were exposed to 700C steam for up to 20 hours. No Zirconium oxide formed on the CrAl coated Zircaloy whereas ZrO<sub>2</sub> with the thickness of over 100um is expected to form on the uncoated Zircaloy and some was observed on the Cr coated Zircaloy-2. For CrAl coatings, (Cr,Al)<sub>2</sub>O<sub>3</sub> formed on the CrAl film with Al composition of 19 atomic percentage. Higher Al compositions did not grow thick enough oxides for resolvable XRD peaks, however STEM confirmed a 65nm thick Al<sub>2</sub>O<sub>3</sub> layer. In addition, an intermetallic layer was observed between the CrAl coating and Zircaloy and was indexed as Zr<sub>6</sub>FeAl<sub>2</sub> by XRD. As neither the exposed environment nor coatings

*CrAl coating with the thickness of 1um inhibited the oxidation of Zircaloy substrate for over 20 hours at 700C steam environment, while over 100um ZrO<sub>2</sub> is expected to form on the uncoated Zircaloy.*

Figure 2. X-ray diffraction patterns of CrAl coatings on Zircaloy-2 after exposure in 700C steam environment for 20 hours. The numbers on each pattern show the atomic percentage of the Aluminum of the as-grown CrAl coatings. ZrO<sub>2</sub> did not form on the 1 $\mu$ m CrAl coated Zircaloy-2.



materials contained Iron, it is believed these elements come from the second phase particles in the Zircaloy-2. Porosity was observed within the CrAl film beneath the alumina layer. The pores did not cause delamination and is believed that diffusion of Aluminum into the Zircaloy-2 substrate, forming the intermetallic phase, improves adhesion.

Coated and uncoated Zircaloy were exposed to normal operating water chemistry at University of Michigan (U of M) for 10 and 20 days. Additionally, one of the 10 day exposed samples was oxidized at 700C for 3.5 hours in steam at UIUC. Characterization by SEM/EDS, XRD, and TEM found that the FeCrAl coated promoted the growth of an iron-nickel spinel from ions leached from

the autoclave system. This surface feature did not affect the protective nature of the base FeCrAl coating. No change in kinetics was seen as compared to as grown samples. Post oxidation characterization revealed that although the spinel persisted through the exposure, normal alpha-alumina formation occurred beneath it. The porosity within the FeCrAl layer is thought to be caused by the clustering of the counter flow of vacancies in response to the outward diffusion of Al ions. As grown FeCrAl coatings were irradiated with protons and iron at U of M. TEM analysis of the irradiated samples is ongoing at UIUC with no substantial detrimental effects observed so far. Analysis of subsequent autoclave exposed irradiated samples is occurring at U of M. Diffraction studies of high tempera-

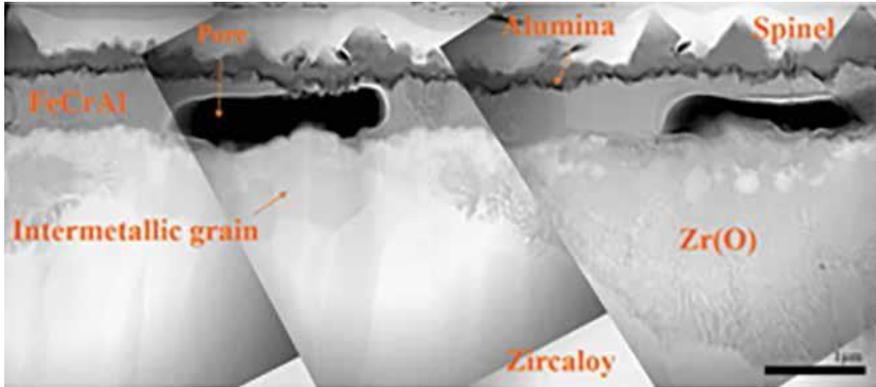
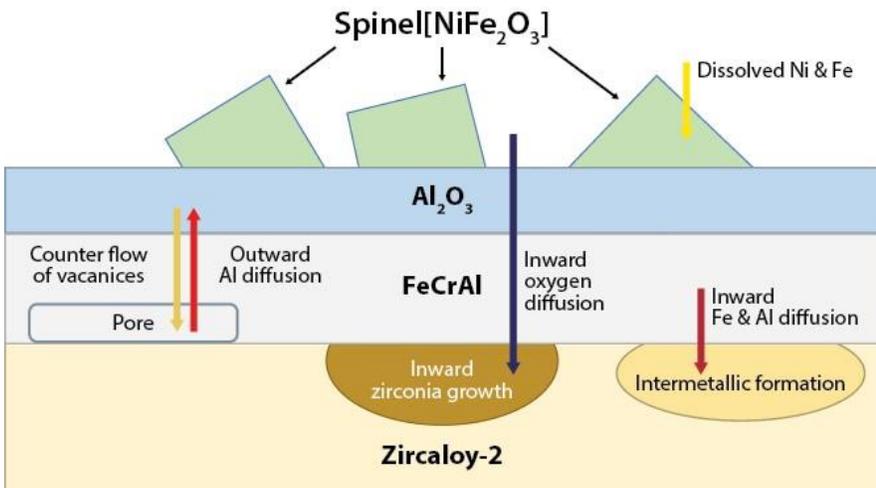


Figure 3. Cross-sectional image and schematic showing the final structure of FeCrAl on Zircaloy after 10 day normal water chemistry autoclave and 3.5 hours 700C steam exposure.



ture steam exposed FeCrAl coated Zircaloy as well as Zr-Y binary alloys were carried out in APS at Argonne. Analysis of this data is ongoing.

At UIUC to accurately capture the temperature distributions in the accident tolerant nuclear fuel, the temperature-dependent thermal conductivity of fuel including the burnup effects (from 0 ~ 80 MWd/kg-UO<sub>2</sub>) was considered using the Kirchhoff transformation to determine the temperature distribution in the

Fuel-gap- cladding-coating region.

To study the impact of the ATF at the system level, the Main Steam Line Break (MSLB) accident was analyzed using Relap5 with a coating thickness of 0.1mm and different coating materials, including FeCrAl, Fe, Cr and Al. In general, no significant difference was found in temperature distributions for different coating materials (primarily due to the thin coating).

## UNIVERSITY-LED TEAMS – UNIVERSITY OF TENNESSEE

### Ceramic Coatings for Clad (The C3 Project): Advanced Accident-Tolerant Ceramic Coatings for Zr-alloy Cladding

Principle Investigator: Kurt Sickafus

Collaborators: U. Tennessee: Maulik Patel, Larry Miller; Penn State U.: Doug Wolfe, Arthur Motta; UC California at Berkeley: Max Fratoni; U. Colorado U.: Rishi Raj; U. Michigan: Gary Was; Los Alamos Natl. Lab.: Kendall Hollis, Andy Nelson; Westinghouse: Jonna Partezana

Figure 1. Principal Investigator Kurt Sickafus discusses the microstructure of a multilayer TiN/TiAlN coating on a zirconium alloy substrate with prospective undergraduate students at The University of Tennessee. Kurt is describing both an image obtained using a scanning electron microscope (SEM) and the associated chemical analysis obtained using energy dispersive X-ray spectroscopy.



The goal of this NEUP-IRP project is to develop a fuel concept based on an advanced ceramic coating for zirconium (Zr) alloy cladding. The coated cladding must exhibit demonstrably improved performance compared to conventional Zr-alloy clad in the following respects:

1. During normal service, the ceramic coating should decrease cladding oxidation and hydrogen pickup (the latter leads to hydriding and embrittlement).
2. During a reactor transient (e.g., a loss of coolant accident), the ceramic coating must minimize or at least significantly delay oxidation of the

Zr-alloy cladding, thus reducing the amount of hydrogen generated and the oxygen ingress into the cladding.

#### Project Description:

The objective of our project is to produce durable ceramic coatings on zirconium alloy cladding. If successful, this research will have a very substantial impact on novel materials development, especially on the development of advanced materials for applications in extreme environments. If we can demonstrate that a ceramic coating resists oxidation in high temperature, aqueous water environments, as well as in the presence of energetic radiation, this will be a major advance in the development of corrosion-resistant materials for nuclear applications.

Coatings on nuclear fuel cladding may make reactors much safer, especially if coatings serve to arrest or slow cladding oxidation during a loss of coolant incident.

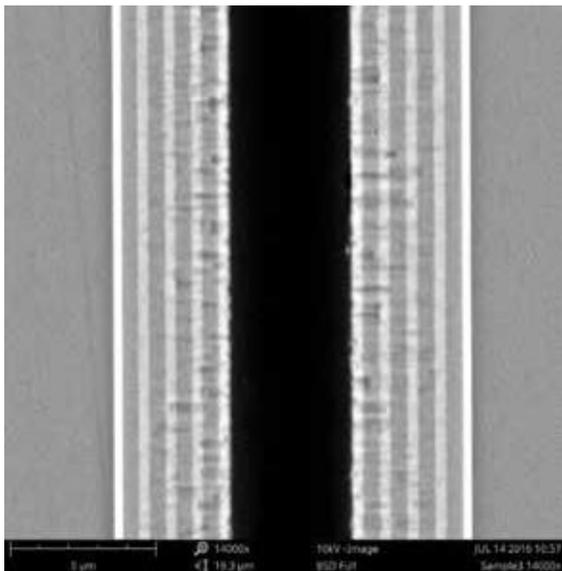


Figure 2. A cross-sectional SEM micrograph of the structure of an 8-layer TiN/TiAlN coating produced by co-principal investigator Doug Wolfe (PSU) using magnetron sputtering. The image (obtained by Sickafus and co-workers at U. Tennessee) shows two identical ceramic coatings mounted face to face (the black region between the coatings is a layer of epoxy). The outermost layer of the multilayer coating is a pure titanium nitride (TiN) layer (the SEM contrast from this layer is relatively bright). Beneath the outermost TiN layer, the coating architecture consists of alternating titanium aluminum nitride (TiAlN) and TiN layers (the TiAlN layers are darker in contrast compared to TiN). The brightest layer at the bottom of the “stack” is a pure metallic Ti layer. This layer is used as a buffer layer between the ceramic coating and the cladding substrate (it improves coating adhesion to the substrate). The substrate in this particular image is silicon rather than zirconium alloy clad. The silicon is used as a control in coating deposition experiments to ensure that the layers are deposited uniformly and with the desired compositions.

### Accomplishments:

In an attempt to develop an accident-tolerant fuel (ATF) with a cladding that can delay the deleterious consequences of loss-of-coolant accidents (LOCA), multilayer ceramic coatings were deposited onto a ZIRLO® fuel cladding by cathodic arc physical vapor deposition (CA-PVD) and magnetron sputtering (Doug Wolfe, PSU). Various coating architectures composed of alternating TiN and Ti<sub>1-x</sub>Al<sub>x</sub>N (2-layer, 4-layer, 8-layer and 16-layer) were deposited in order to investigate the minimum TiN top coating thickness necessary to avoid aluminum hydroxide phase formation during corrosion and optimum

coating architecture for good corrosion resistance and oxidation resistance. One type of 2-layer architecture consisted of a 1 μm TiN top thickness (~1/10 of the total coating thickness), while other coatings were composed of layers with approximately equal thickness. Corrosion tests were performed in static pure water at 360°C and saturation pressure (18.7 MPa) up to 90 days (Jonna Partezana, Westinghouse). Coatings having no spallation or delamination survived the autoclave test exposure with a maximum 6 mg/dm<sup>2</sup> weight gain, which is 6 times smaller than that of the uncoated ZIRLO™ sample which had a weight gain of 40.2 mg/

dm<sup>2</sup>. Post-corrosion exposure analytical characterization (PSU and U. Tennessee) showed that depositing ~1 μm TiN as a top layer prevented aluminum hydroxide formation and TiN/TiAlN 8-layer architecture provided best corrosion performance due to no hydroxide phase formation, approximately linear weight gain data without any delamination/spallation and advanced oxygen ingress prevention. Multilayer TiN and Ti<sub>1-x</sub>Al<sub>x</sub>N coatings have shown significant improvements in the high temperature corrosion resistance of zirconium alloy cladding.

## 2.2 HIGH-PERFORMANCE LWR FUEL DEVELOPMENT

### In Situ Synchrotron characterization of the Field Assisted Sintering of UO<sub>2</sub> at the National Synchrotron Light Source -II

Principal Investigator: Doug Sprouster – may move to cross cutting area

Collaborators: A.L. Hanson, E. Dooryhee, L.E. Ecker, R. Pokharel, H.M. Reiche, A.M. Raftery, D.D. Byler, and K.J. McClellan

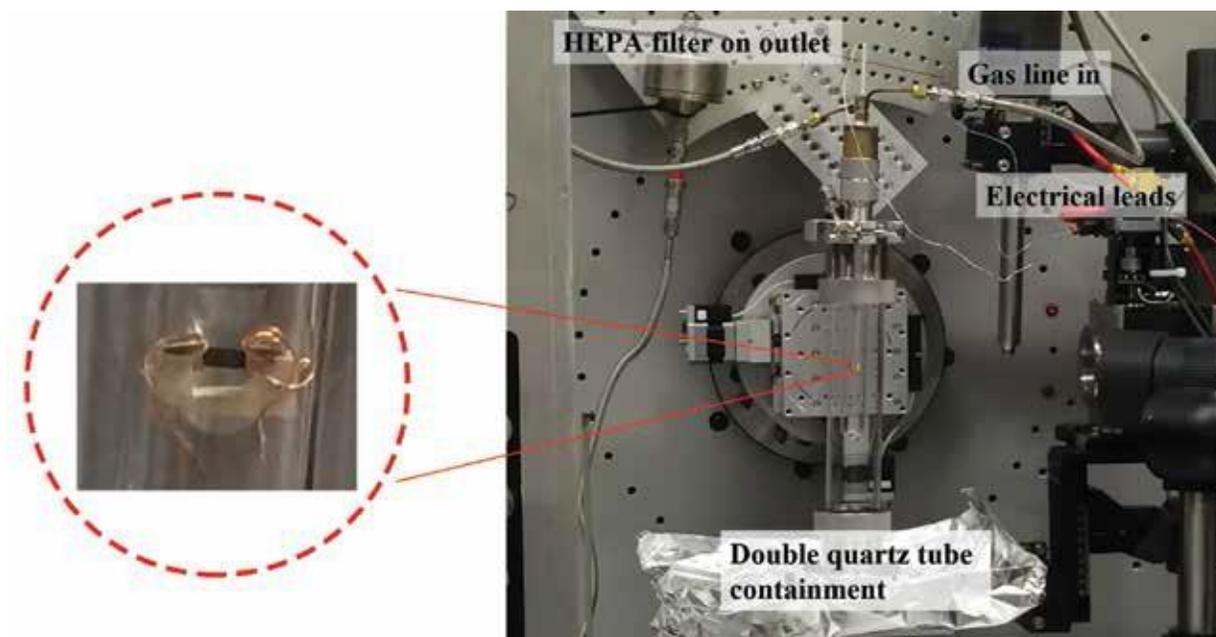


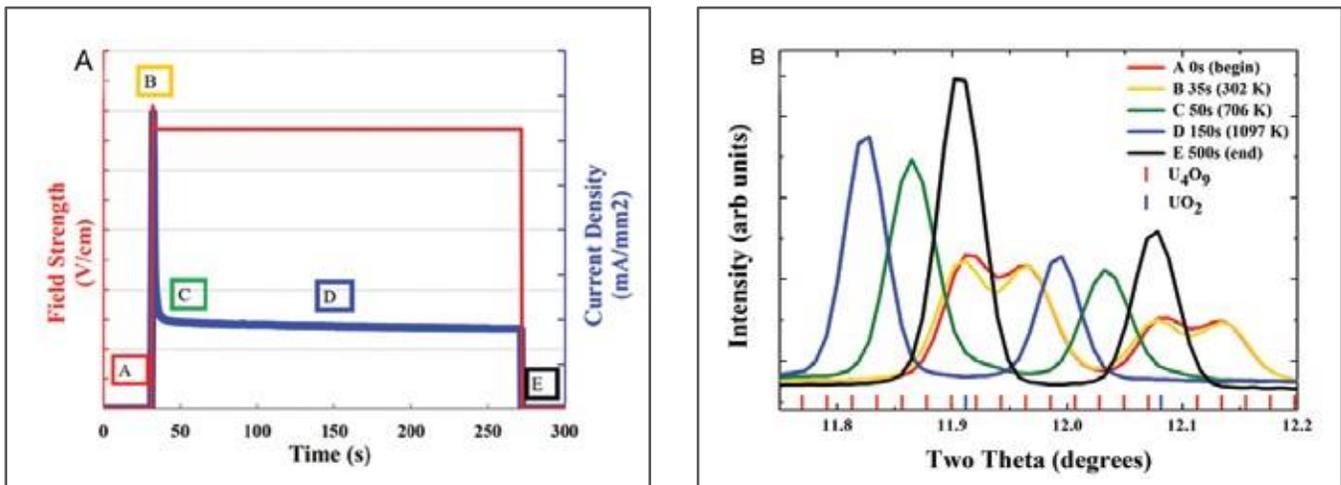
Figure 1. Experimental set up at the XPD beamline. The different components and an enlarged image of the sample in the holder is shown for reference.

During Field Assisted Sintering (FAS) it is important to understand the densification mechanisms that are activated by the applied electric field. FAS techniques employ electrical fields in combination with heat and pressure, to reduce sintering times and temperatures. FAS-induced densification is difficult to control and the rate controlling mechanisms are not yet understood. In situ techniques such as X-ray diffraction were used to identify the phase changes, microstructure and defects present in UO<sub>2+x</sub> during the application of

electric field at room temperature. The data generated by the in situ experiments will provide information for theoretical models. The ultimate goal is to develop a more controlled, optimized densification process for UO<sub>2+x</sub>.

#### Project Description:

Advanced fuels that are being studied for enhanced accident tolerance and for actinide transmutation often have unique characteristics which make conventional sintering route undesirable (or incapable) of yielding the required fuel pellet characteristics. The experiments described here explore the physical mechanisms operative



during the early intermediate and late stages of FAS. The ultimate objective is to develop optimized and reproducible processing conditions leading to a controlled densification in oxide fuels.

The specific technical objective of the experiments was to examine the crystallographic evolution of various UO<sub>2+x</sub> samples as a function of time, chemistry and applied field. By quantifying the crystallographic structure in situ, the key variables and defects that control the onset of the sintering process were revealed.

The work performed here is in direct alignment with the AFC's "goal-oriented science-based approach", where we have utilized state of the art x-ray capabilities at the National Synchrotron Light Source-II (the newest and brightest x-ray source in

the world), to perform fundamental experiments to develop understanding of the Field assisted sintering fabrication method. Through the course of the experiments performed here, we have developed a new in situ capability to examine the crystallographic and microstructure evolution in candidate fuels during processing. This structural information can be directly incorporated into modeling efforts to enable more efficient fuel synthesis route reducing processing time and/or temperature. An additional objective of this work was to develop a capability that leverages existing investments in the high-throughput experimental and analysis capabilities that would benefit similar future structural studies of advanced fuels and claddings.

Figure 2. Applied field and location on the FAS curve where XRD patterns in b were collected. The UO<sub>2</sub> and U<sub>4</sub>O<sub>9</sub> phases are overlaid for reference with the temperature based on the thermal expansion of the UO<sub>2</sub> lattice. Note the crystallographic phase after these FAS conditions is different to the initial 50/50 mixture, and the loss of U<sub>4</sub>O<sub>9</sub>.

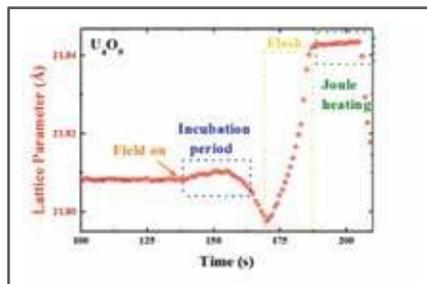
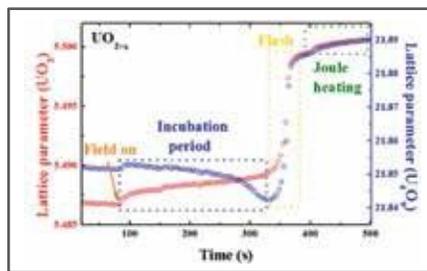
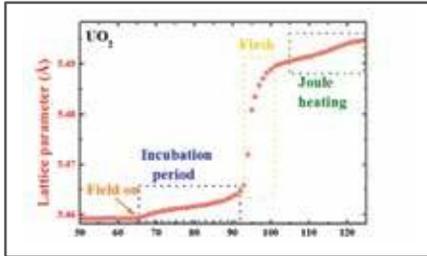


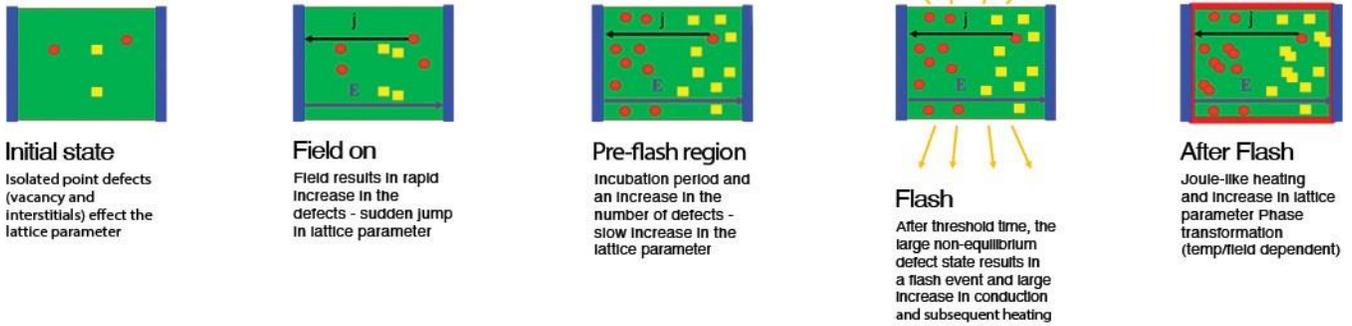
Figure 3. Lattice parameter as a function of time for three different  $UO_{2+x}$  samples. The behavior in the different regions are described in the text, and proposed model is shown below in Figure 4.

### Accomplishments:

Two sets of in-situ and ex-situ experiments were performed at the X-ray Powder Diffraction (XPD) beamline at the National Synchrotron Light Source-II (NSLS-II). High energy (66 keV) x-rays were used to probe  $UO_{2+x}$  samples as a function of stoichiometry and applied voltage. The experimental set up included a spring loaded sample holder encapsulated in a double quartz cell, fitted with gas feed lines, electrical leads and thermocouple (as shown in Figure 1). Patterns were collected in both static-mode, to spatially determine the crystallographic phase as a function of position between the contacts, and in dynamic mode where the sample was fixed and patterns were collected as a function of time through the FAS process. We also performed dynamic-mode tests while scanning the beam between the contacts during the FAS process. The collection of the diffraction patterns (over 100,000 patterns in total) was performed in transmission mode with 1.0s acquisition time with a Perkin Elmer flat panel detector. The Rietveld analysis of the diffraction patterns and extraction of the lattice parameters was performed using dedicated high-throughput python scripts that interface with TOPAS (Bruker).

The lattice parameters were quantitatively determined for the many various samples measured. A typical FAS curve is shown in Figure 2 with corresponding XRD peaks collecting over the FAS cycle shown in Figure 2 b. The changes in peak position, and lattice dynamics were quantified through Rietveld analysis. An example of the lattice dynamics is shown in Figure 3 for three different  $UO_{2+x}$  sample compositions ( $UO_2$ ,  $U_4O_9$  and a ~50/50 mixture of  $UO_2$  and  $U_4O_9$ ) during the flash experiments. The time when the field was applied is shown for reference. The in situ behavior of the samples show that there is an incubation period prior to the flash event. The incubation period is characterized by an initial increase in the lattice parameter. The samples are not highly conductive during this time and the length of the incubation period is related to the applied voltage. The incubation period was observed in all samples, with minimal (to no) increase in the temperature. Such structural changes are indicative that the number of lattice defects increases prior to the onset of the flash event and may be composition independent. Figure 3 also shows that following the incubation period there is a very-rapid increase in the lattice expansion during the flash event for all compositions. This expansion coincides with

Figure 4. Proposed mechanism based on the in-situ experimental results



an increase in conductivity and an increase in the measured temperature. After this rapid expansion there is another region where there is steady increase in the lattice parameter due to joule heating.

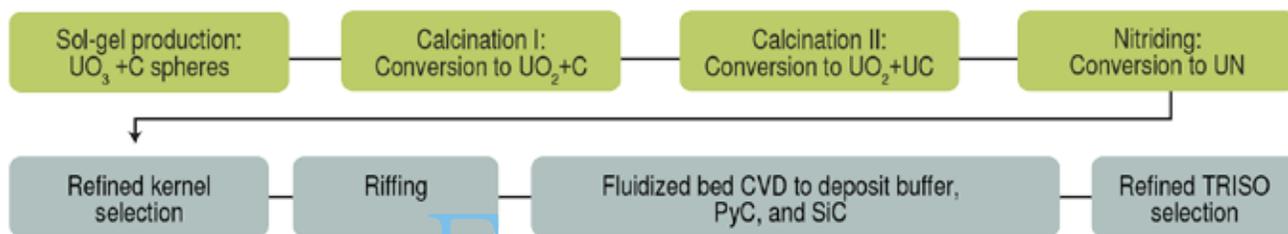
These experiments readily show that significant changes in the atomic structure ensue during field assisted sintering. A proposed model is shown in Figure 4 and depicts the different stages that are operative before, during and after the “flash” event. The next step in this work is to compare the experimental results directly to theoretically predicted behavior to determine if there is a preferential process that is governed by defect formation.

Analysis of the lattice parameter as a function of distance between the electrical contacts and time will facilitate determining if there are any spatial changes in the samples with flash conditions and any defect migration that translates to the bulk (mass) material movement. This information could then be used to tailor the application of the voltage and current to impart specific defect transport mechanisms within the  $UO_{2+x}$  materials to produce a high-density product and to tailor microstructures for specific characteristics.

*In situ synchrotron characterization and high-throughput data analysis was used to develop a mechanistic model of densification during field assisted sintering of  $UO_{2+x}$  and will be crucial in configuration simulations of the process.*

## LEU Uranium Nitride Kernel Production

Principal Investigator: Jake McMurray



particle process.  
Figure 1. Flow chart of the integral FCM TRISO

Fully ceramic microencapsulated (FCM) fuel consists of tri-structural isotropic (TRISO) particles embedded in a pellet shaped dense SiC matrix. FCM can be used as an accident tolerant alternative to conventional uranium dioxide in Light Water Reactors (LWR). Accident tolerance is achieved by a synergistic effect combining the established effectiveness of TRISO particle fuel at retaining radionuclides along with the high temperature steam corrosion resistance and added barrier to fission product release from the SiC matrix.

### Project Description

To aid qualification of FCM, irradiation testing is planned for integral FCM fuel pellet compacts. This requires ~150 grams of low enriched uranium (LEU) bearing UN to be used as kernels for the TRISO particles; therefore, a large batch of  $797 \pm 1.35$   $\mu$  m diameter UN microspheres were produced. Fabrication was carried

out using carbothermic reduction and nitriding of a sol-gel feedstock bearing tailored amounts LEU oxide and carbon with processing parameters determined from previous experimental work. These microspheres were characterized and upgraded for coating with the appropriate TRISO layers using chemical vapor deposition part of an overall process developed at Oak Ridge National Laboratory (ORNL) shown in Fig. 1. The TRISO particles are subsequently made into FCM compacts for planned irradiation testing.

### Accomplishments

The internal gelation process was used to produce 1400 - 2000-  $\mu$  m-diameter feedstock of hydrated UO<sub>3</sub> with homogeneously-embedded carbon powder. The starting material consisted of uranium oxides, with a <sup>235</sup>U enrichment of 7.35 at. % U, and Cabot Mogul L carbon black. A dispersing agent, Tamol SN, was added

to the chilled basic hexamethylene-tetramine (HMTA)/urea solution and sonicated for 5 min with a Hielscher UP200S ultrasonic probe. Since sonication heats the solution, it was subsequently rechilled. To this, a chilled acid deficient uranyl nitrate solution was added to form the broth which was then added as droplets in a controlled fashion into a flowing stream of hot (~62 °C) immiscible silicone oil to facilitate the gelation reaction. The gelled spheres were then washed and dried resulting in a product with a C/U ratio of 2.65 with ~2 moles of adsorbed H<sub>2</sub>O /mole U and traces of NH<sub>3</sub>.

A carbothermic reduction and nitriding process given in Fig. 2, resulted in a final product of phase-pure NaCl structure UN with approximately 13.5 molar % dissolved C determined from X-ray diffraction. The UN microspheres weighed 197.4 g, had an average diameter of  $797 \pm 1.35 \mu\text{m}$  and a composite theoretical density (TD) of  $89.9 \pm 0.5\%$  for a solid solution of UC and UN with the same atomic ratio. The overall conversion process is given in Fig. 1. The upgraded UN kernels will be made into FCM compact for planned irradiation testing.

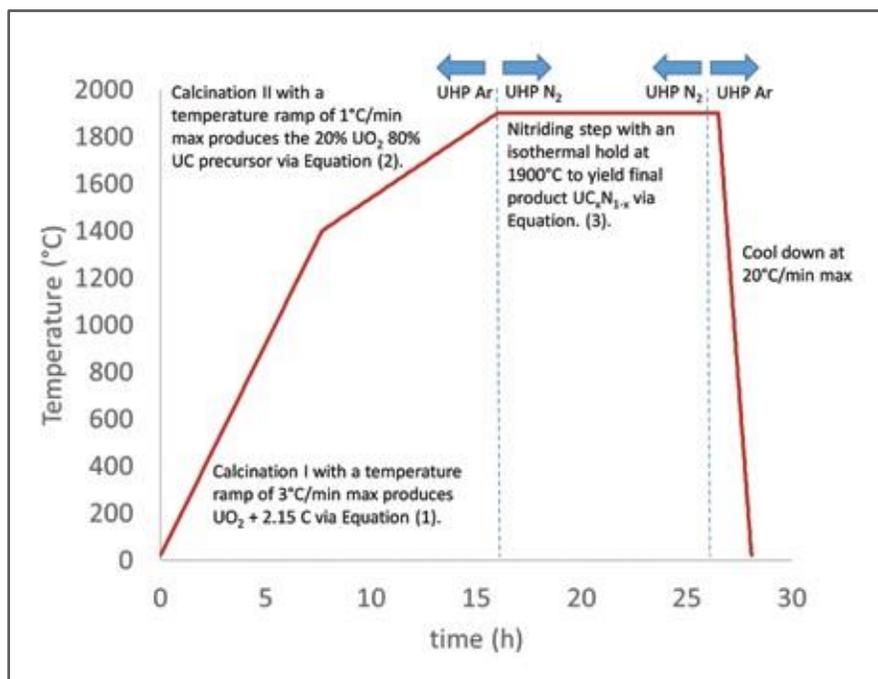


Figure 2. Process used to convert the sol-gel feedstock to ~800 $\mu\text{m}$  and 89.9% TD phase-pure UC<sub>x</sub>N<sub>1-x</sub> microspheres with 13.5 at. % dissolved C.

**A 197.4-g batch of LEU-bearing, ~800- $\mu\text{m}$ -diameter 89.9% theoretical density UN microspheres was produced for FCM irradiation testing.**

## Uranium Nitride TRISO Particle Production

Principal Investigator: Brian Jolly

Collaborators: Grant Helmreich, John Dyer, Kurt Terrani

Figure 1. Fluidized bed chemical vapor deposition system at ORNL used to produce the UN TRISO particles.



**A first of a kind uranium nitride TRISO particle was produced using 800 $\mu$ m low enriched UN kernels.**

**T**In support of fully ceramic microencapsulated (FCM) fuel development, coating development work was carried out at the Oak Ridge National Laboratory (ORNL) to produce tri-structural isotropic (TRISO) coated fuel particles using uranium mono-nitride kernels. The UN kernels are used to increase fuel density in these SiC-matrix fuel pellets. Similar to the AGR-1 program, a fluidized bed chemical vapor deposition (FBCVD) technique was used to fabricate the UNTRISO particles. However, in order to maintain acceptable coating properties,

significant process development was required due to the physical property and dimensional differences between the new 800  $\mu$  m diameter UN kernels and the previous 350  $\mu$  m UCO kernels used during AGR-1.

### Project Description

The overall goal for the UN coating development work performed this FY was to produce TRISO particles using low enriched UN kernels. Since these are a first of a kind TRISO particle, a large portion of the project involved developmental coating experiments to generate a “recipe” for producing UNTRISO particles which meet the coating property specifications. The coating specifications were based upon

the AGR-1 specifications as the AGR-1 TRISO particles have demonstrated good irradiation performance. These UN TRISO particles will be used for fabrication of FCM compacts which are planned for irradiation testing in the ATR at INL. The ability to fabricate UN TRISO particles is a critical step in the evaluation of the FCM fuel concept as an accident tolerant fuel form for light water power reactors.

### Accomplishments

In order to reach the goal of producing low enriched UNTRISO particles, first the FBCVD coating system (Figure 1) at ORNL had to be “refurbished”. Prior to the UN coating work, this coating system had not produced full TRISO particles since the end of the AGR-1 coating program (~2006). An initial batch of TRISO particles was produced, and although not fully optimized, it confirmed that the coater was fully operable.

With the FBCVD coater “refurbished”, development work began to optimize the processing conditions and the properties of each coating layer. Properties for each layer of the TRISO system are dependent upon the properties of the preceding layer. So, coating development was performed in a stepwise fashion with processing conditions for the buffer layer being optimized before proceeding to the inner pyrocarbon layer (IPyC), and processing conditions for the IPyC layer being optimized before beginning SiC depositions etc. Following

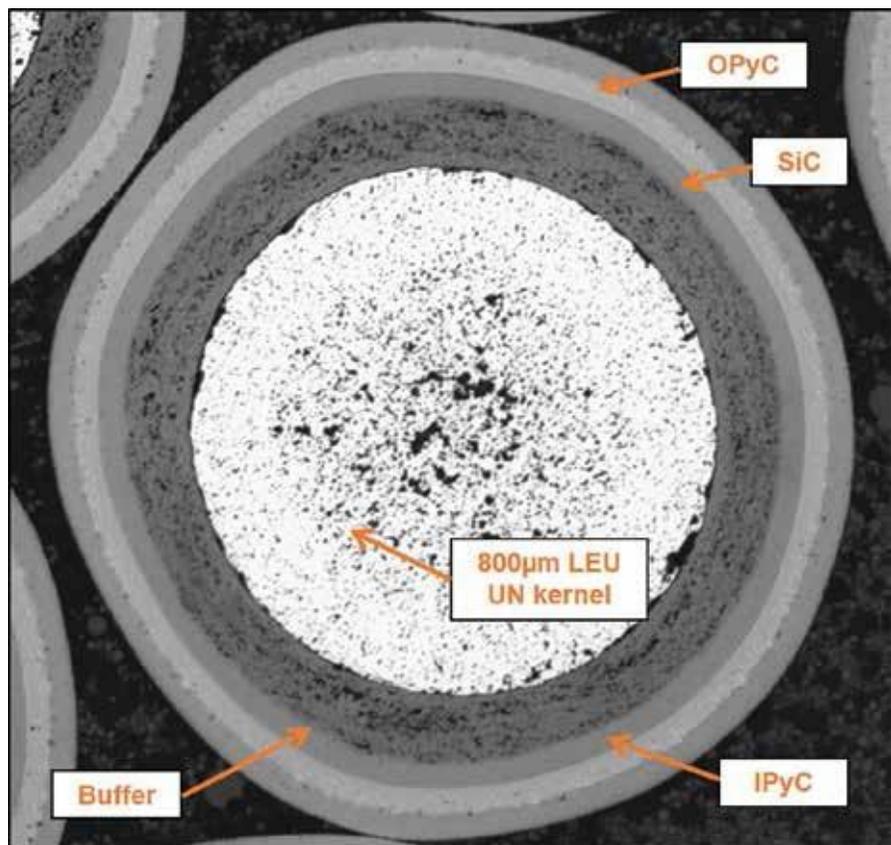


Figure 2. Optical cross section of a typical low enriched UN TRISO particle

this progressive strategy, depleted UN kernels were used to “lock down” coating conditions prior to switching to the low enriched material.

Using the “recipe” developed during the depleted UN kernel work, a first batch of TRISO particles was fabricated utilizing low enriched UN kernels. This batch met all measured

coating property specifications. An optical cross sectional micrograph showing a typical  $800\ \mu\text{m}$  diameter UN kernel and the 4 coating layers is shown in Figure 2.

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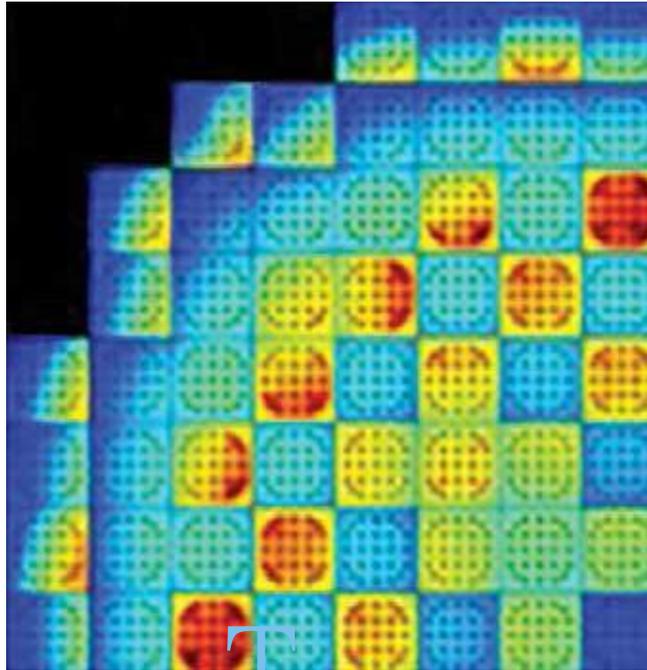
## 23 ANALYSIS

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### Development of Accident Tolerant Fuel Options for Near Term Applications

Principal Investigator: Jacopo Buongiorno

Collaborators: K. Smith, K. Shirvan, K. Sridharan, L. Shao, M. Tonks, P. Broda, W. Liu, J. Hales



ANATECH and AREVA

T

computational models

Figure 1. The IRP Team - MIT, INL, UW, TAMU, PSU,

his work focuses on development and evaluation of

under the NEAMS framework to estimate time to failure for near term accident tolerant fuel concepts. The IRP builds upon strong university capabilities at the Massachusetts Institute of Technology (MIT) with its experience in fuel design and safety analysis, the University of Wisconsin (UW) with its experience in severe accident modeling and

development of cladding coatings for the ATF industrial campaign, Texas A&M University (TAMU) with its material ion irradiation capability, and Pennsylvania State University (PSU) with its meso-scale fuel performance modeling experience. INL is a member of the team to allow for efficient implementation and integration of the models. The IRP also benefits from close collaboration with two industrial partners: ANATECH, engineering firm with state-of-art experience in fuel modeling under accident conditions and AREVA, one of the main nuclear fuel suppliers in the US.

#### Project Description

Our work is focused on current operating light water reactors world-wide, and we aim to improve their safety

performance with use of innovative accident tolerant fuels. The objective of this IRP is to develop computational tools to evaluate such ATF options for near term applications. The computational tools will be predominantly developed under the framework of the NEAMS such as MOOSE/BISON (engineering scale fuel performance). For proper validation, existing experimental data will be utilized and new low-cost experimental data will be generated within this IRP. For verification, industry standard and licensing



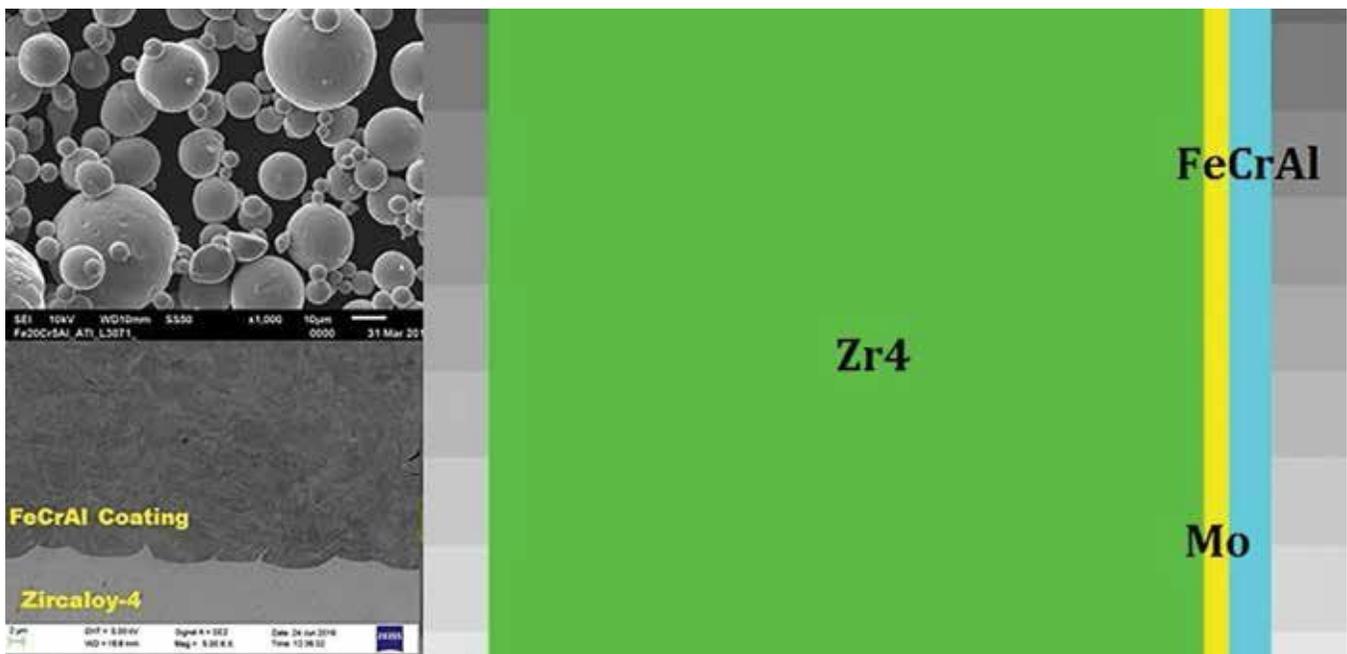


Figure 2. SEM images of the higher alloy FeCrAl powder (top) and deposited and annealed at 550oC (below) along with simulation of Zr4 cladding with 20 um Molybdenum and 30 um FeCrAl coatings in BISON.

tools for neutronics, thermal hydraulics and fuel performance will be used for model development of conventional fuel and to provide cross-code checking of ATF performance. The newly developed models will be evaluated at the core level under both steady state and transient conditions, including severe accidents, where time to failure of the fuel and core components is estimated. The project findings will advance the state-of-the-art tools for simulating the accident

tolerance of current and innovative nuclear fuels. The project will play a strong role in the successful execution of DOE's accident tolerant fuel program, which was started in response to the Fukushima accident. It will also improve selected safety analysis tools used by the nuclear regulatory commission (NRC) by extending their ability to predict fuel failure during severe accident conditions.

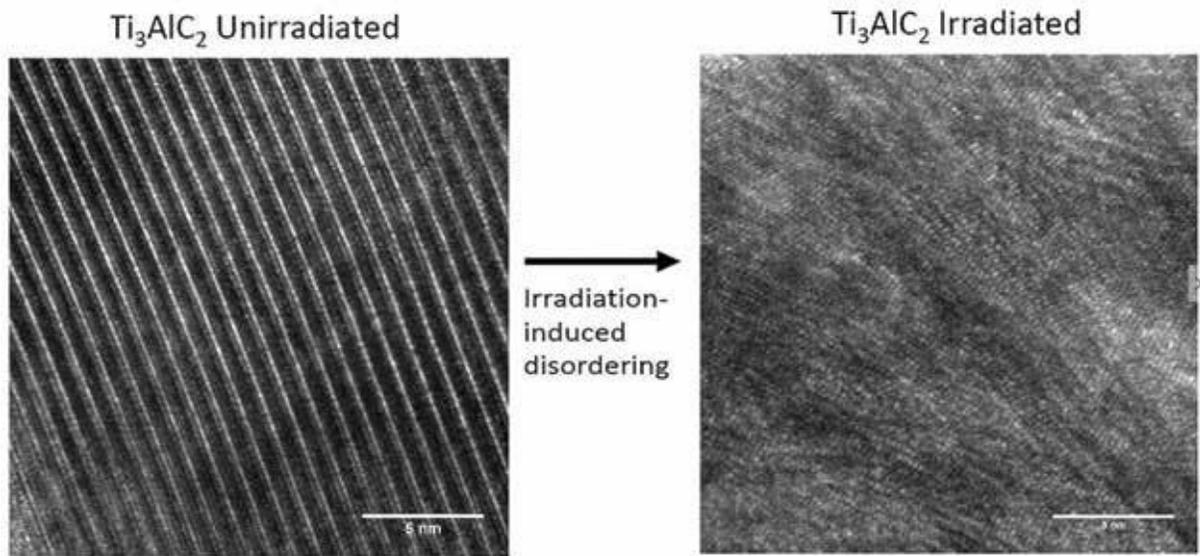


Figure 3. Ti-Al-C MAX phase coating: Before and After Ion Irradiation.

### Accomplishments

AREVA Fuels has completed the initial machining and shipment of Zircalloy-4 samples to University partners. AREVA has also obtained and installed the MOOSE/BISON fuel performance code along with the CUBIT meshing package on their machines for future use in the project.

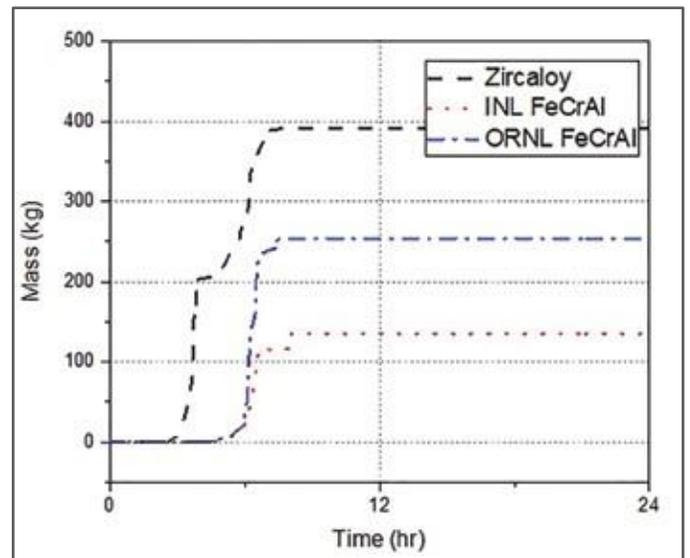
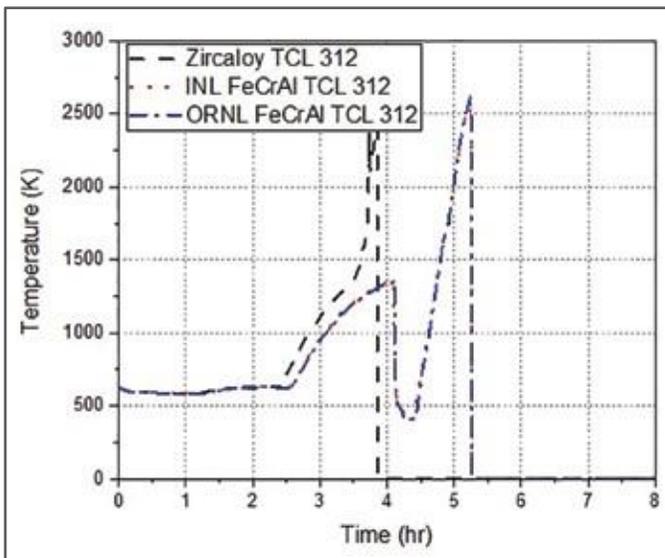
The cold spray coating technology has been used by UW to deposit Ti-Al-C MAX phase and FeCrAl alloy coatings on Zr4 fl t and tubular geometries. Inter-diffusion between FeCrAl alloy and Zr4 at high temperatures were observed. UW will investigate use of Molybdenum to prevent such inter-diffusion.

TAMU systematically studied the phase formation and diffusion kinetics at interfaces of TiAlC coated Zircalloy-4 after prolonged annealing, and ion radiation responses and subsequent mechanical property changes of each interface phase. Similar study for FeCrAl coating is ongoing.

MIT has performed the calibration of their existing experimental facilities to perform low temperature and high temperature steam oxidation in addition to PWR prototypical corrosion/CRUD deposition and 4-point bend and pressurized tube mechanical tests. MIT has also fabricated Zr4 samples to perform such tests from Zr4 tubes, solid rods and plates obtained from AREVA. In the next year, critical heat fl and quench studies will also be performed.

MIT obtained TRACE model of a 3-Loop PWR from BNL while UW obtained a similar model (Surry plant) for MELCOR. The models have been exercised for different accident scenarios including large break loss-of-coolant accident and short term station black out. The code-to-code comparison of TRACE and MELCOR to the point of fuel failure is currently ongoing. In MELCOR, UW confirmed that FeCrAl as the cladding material increases the response time allowed with initial hydrogen generation as the fi e of merit.

*“The project develops state-of-the-art simulation tools for new fuel cladding materials that will tolerate accident conditions without generating massive amounts of fission hydrogen, thus greatly mitigating the consequences of Fukushima-like events at nuclear power plants worldwide”.*



At PSU, the MARMOT mesoscale fuel performance tool has been used to investigate the impact of high thermal conductivity additives BeO and SiC on UO<sub>2</sub> thermal conductivity using 2D and 3D simulations. The impact of additives on fission gas release will also be investigated.

MIT has performed a detailed steady state fuel performance modeling of a Zr<sub>4</sub> cladding with FeCrAl, Cr, Mo and Nb coatings including combinations of each and monolithic FeCrAl using the BISON fuel performance framework. An in-house fuel performance tool

along with a finite element analysis tool, ABAQUS, were used to verify BISON calculations. It was found that plasticity models are typically required under steady state, especially during pellet-to-clad mechanical interaction due to thermal expansion and mechanical properties mismatch of each coating layer with Zr<sub>4</sub> and each other. During upcoming year, data to validate selected coating properties for BISON modeling will be produced. Also, fuel performance of such coated cladding under accident conditions will be investigated.

*Figure 4. Preliminary Initial Comparison of Zr<sub>4</sub> and FeCrAl with different Oxidation Models in MELCOR during a Short Term Station Blackout.*

## Fundamental Materials Modeling for ATF Concepts

Principal Investigators: Dr. Kurt A. Terrani and Dr. Brian D. Wirth  
 Collaborators: Gyan Singh, Brian Wirth, Ryan Sweet

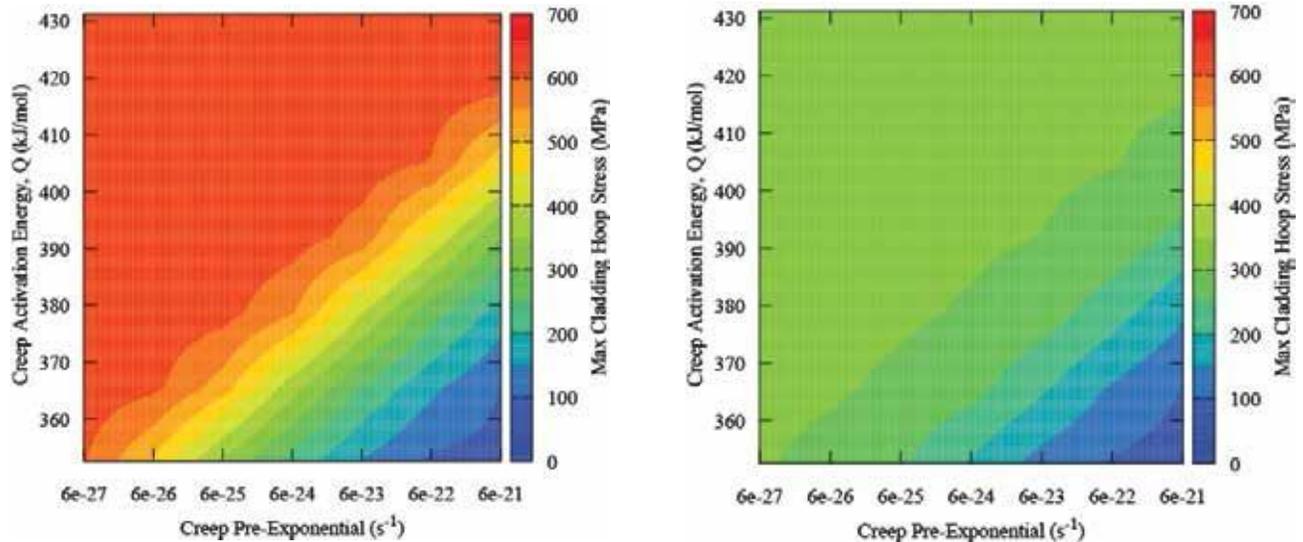


Figure. BISON model results that parametrically evaluate the thermal creep behavior of FeCrAl alloys and the corresponding effect on the maximum clad hoop stress developed during FeCrAl ATF operation. This study identified cladding thermal creep as an important material design property to improve fuel performance.

In order to improve the accident tolerance of light water reactor (LWR) fuel, alternative cladding materials have been proposed to replace zirconium (Zr)-based alloys. We have utilized computational modeling, from both a fundamental as well as engineering scale perspective to evaluate the performance of several accident tolerant fuel (ATF) clad concepts, including SiC and FeCrAl alloys. We have used the BISON fuel performance code to evaluate SiC/SiC cladding, particularly focusing on temperature dependent creep and dimensional changes that can influence the overall fuel rod dimensional stability. We have also used BISON to evaluate FeCrAl clad,

where thinner clad tubes surround larger fuel pellets. These modeling analysis have highlighted several key material properties and behavior which require further study.

### Project Description:

The Fukushima Daiichi nuclear power plant accident in 2011 highlighted the susceptibility of the light water reactor (LWR) cores to severe degradation under beyond design basis accident conditions. Efforts to enhance the safety of nuclear power plants are underway and involve extensive research and development work on enhancing the accident tolerance of fuel-cladding system. A number of advanced fuel-cladding systems are under consideration with the primary focus on enhanced steam oxidation resistance. The primary focus of

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this project is to use computational models, primarily focused on using the thermal-mechanical engineering scale fuel performance modeling capability of BISON to assess the performance of SiC/SiC composites and FeCrAl alloys as alternative ATF fuel clad. In order to compare the predicted in-reactor performance of ATF cladding with Zircaloy and assess effects from irradiation creep, thermal creep, and swelling, several geometric and material models were implemented in to the BISON fuel performance code to model either FeCrAl or SiC composites. These modeling assessments will help prioritize the research and development required to optimize an ATF concept for lead test assembly testing within the next five years, which helps meet the DOE objective of safe and reliable nuclear power in the United States.

**Accomplishments:**

Initial analysis has been performed to assess FeCrAl alloys (namely Alkrothal 720 and APMT) as a suitable fuel cladding replacement for Zr-alloys, using the MOOSE-based, finite-element fuel performance code BISON and the best available thermal-mechanical and irradiation-induced constitutive properties. Likewise, BISON simulations have been performed for SiC/SiC composites that provide insights about the stress distribution and variation

with time in the cladding as well as the interaction of fuel pellet with the cladding under different conditions of initial fuel pellet-cladding gap and steady state power levels. The FeCrAl ATF simulations identify the effects of the mechanical-stress and irradiation response of FeCrAl and provide a comparison with Zr-alloys. The results from the FeCrAl analysis showed the development of high tensile stresses in the cladding across several different BWR operational scenarios. However, it is important to note that large changes in the results can arise from small shifts in data and are due to the complex, coupled sensitivities inherent in integral fuel performance modeling. While these simulations show very large stresses (>600 MPa) forming in the FeCrAl cladding (much larger than the yield strength at these temperatures), they do provide information on important parameters for alloy design and certain modeling techniques. Future efforts will focus on continuing the systematic evaluation of FeCrAl and UO<sub>2</sub> thermal-mechanical constitutive properties and irradiation behavior models, including against experimental data being developed within the FCRD program, and initiating the modeling investigation of transient fuel clad behavior.

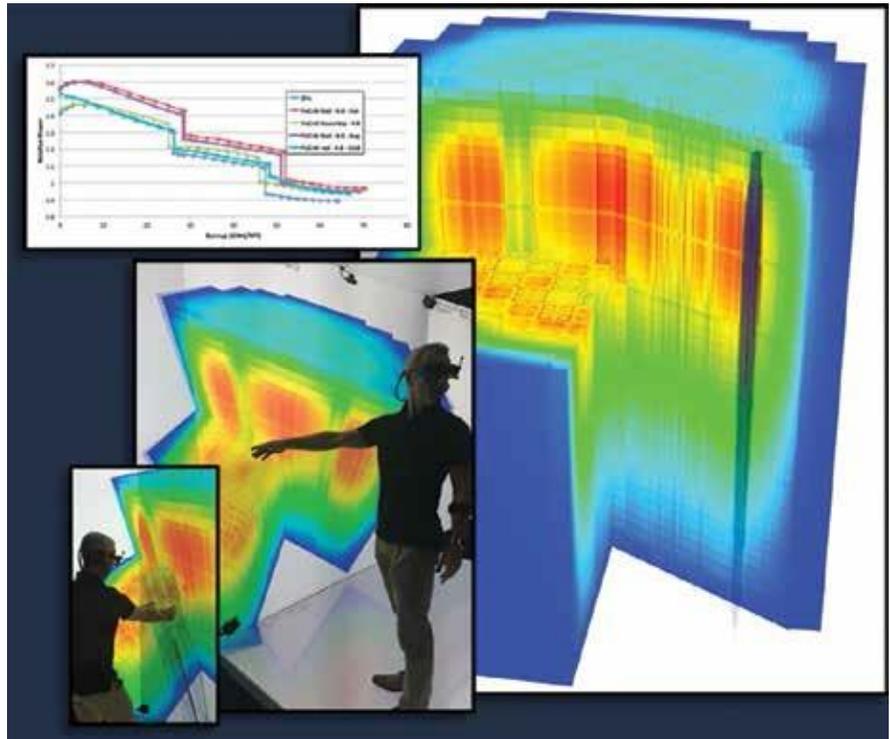
*This research provides critical assessments of candidate ATF clad materials, which can directly identify the most promising candidates, as well as prioritize needed ATF clad research to ensure timely completion of a lead test rod and assembly testing of the most promising concept.*

## ATF Analysis in Support of Metrics Development

Principal Investigator: Andrew Worrall

Collaborators: Nick Brown, Henrik Liljenfeldt, Jake McMurray, Josh Peterson, Jeff Powers  
Kevin Robb, Kurt Terrani, Troy Eckleberry

Figure 1. Detailed whole core analysis has been completed using industry, as well as advanced codes (VERA, CASL) to evaluate the local peaking factor impact of loading ATF. The high order solutions available in CASL's tools provide an additional level of detailed resolution and interrogation, and provide additional confidence in the results.



The primary focus is on the development of metrics for the assessment of ATF concepts regarding their impact on reactor performance and safety characteristics of the reactor. These include evaluation of additional margins (e.g. coping time, reduced cooling) upon utilization of ATF fuels and materials in LWRs. A secondary activity around engineering scale neutronics impact of these ATF concepts is also within the scope. Finally, these activities pave the way for informed interaction

with industry and NRC to support LTR/LTA timeline and support down-select and licensing.

### Project Description

The ORNL work has 2 major high-level objectives: (1) to provide appropriate analysis to support the prioritization and potential down-selecting of ATF concepts (primarily neutronics, fuel performance, and severe accident analysis), (2) to provide appropriate data and analysis to industry and NRC to support the future LTR/LTA irradiation programs.

The ability to demonstrate enhanced accident tolerance can be measured by a number of different physical properties that can increase coping time. One of the objectives of the work this year was to generate new experimental data on melt temperatures of FeCrAl claddings, and evaluate the impact on coping time for ATF. This would be generating new experimental data, and the ability to combine this with severe accident analysis using the new data in MELCOR is world-leading.

For any new fuel to be able to be loaded into a commercial reactor, the operator and regulator must be confident in the performance of that fuel, and the ability to predict its performance under a range of operating conditions (e.g., normal and off-normal operations). For standard UO<sub>2</sub>-Zr fuels, a large database of validation data exists, including neutronics, fuel performance etc. But this is not the case for ATF concepts. Therefore, in the absence of experimental data at this time, the objective of this year's work was to establish an analysis benchmark progression problem set to (a) evaluate the neutronics codes for application to ATF concepts, (b) identify potential gaps/needs in the methods and data, and (c) use this to inform on the future LTR and LTA irradiation

programs. This includes using both industry, and state-of-the-art tools for modeling LWRs, such as those in CASL, and will assist in shortening the development and deployment timescales for ATF.

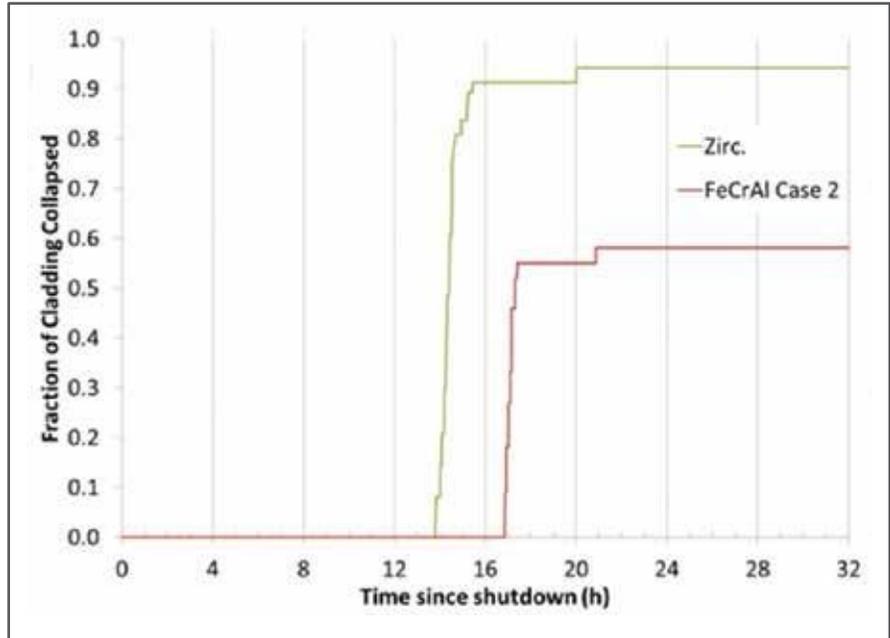
#### **Accomplishments**

*K R. Robb, J. W. McMurray,  
K A. Terrani, ORNL*

There is a need to quantify the potential gains of Accident Tolerant Fuel (ATF) concepts under active development by DOE and industry. ORNL/TM-2016/237 (M2FT-16OR020205042) aimed to quantify the potential gains provided by the Iron-Chromium-Aluminum alloy (FeCrAl) ATF concept. Refined values for the melting point of FeCrAl metal alloys and the oxide formed by FeCrAl were pursued. These values were then used to update predictions of the severe accident performance of a BWR plant utilizing FeCrAl cladding and channel boxes. Seven different accident scenarios were analyzed. Overall, when compared to the traditional Zircaloy-based system, the FeCrAl concept provided a few extra hours of time for operators to take mitigating actions and/or for evacuations to take place. A coolable core geometry

*The applied analysis is important in that it informs not only on the performance of ATF concepts, but also assisting in the licensing of the concepts, supporting vendors and regulators as well as DOE.*

Figure 2. Compared to previous work the gains afforded by the FeCrAl ATF concept over the existing Zircaloy system currently in use were predicted to be less. However, FeCrAl provided gains over Zircaloy in most metrics for all scenarios analyzed with respect to timing and flammable gas generation. An example of the extent of the benefit is shown above.



was retained longer, enhancing the ability to stabilize an accident. Finally, due to the slower oxidation kinetics, less hydrogen was generated, and the timing of generation was delayed. This decreased the amount of non-condensable gases in containment and would lower the potential for deflagrations to inhibit the accident response. The figure below depicts the fraction of cladding that collapses during a mitigated station blackout (SBO) where water injection is unavailable during the 8-16 h timeframe.

*Nick Brown, Henrik Liljenfeldt, Josh Peterson, Jeff Powers, Kevin Robb, Kurt Terrani, Andrew Worrall*

There have been a number of domestic and international neutronic assessments of the various ATF concepts over recent years. However, there has been little if any work on determining the suitability (accuracy in methods, nuclear data available or evaluated appropriately etc) of these tools, either at the whole core, or the assembly level. In order for an ATF concept (LTR or LTA, or full reload) to be irradiated in a commercial reactor, the accuracy of these tools for reactivity/cycle

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length, pin powers, reactivity coefficient shutdown margin etc, has to be assured for ATF. One may argue that the existing industry codes are indeed capable of accurately predicting the performance of ATF, but without code comparisons and/or experimental validation, that may not be the case; evidence is required to make that judgment. High order solutions such as those from CASL can provide additional confidence in the industry and other methods.

To this end, and based on an existing benchmark established for CASL, a series of progression problems has been produced and written in the form of a benchmark that can be used with the ATF program, as part of the broader DOE programs (e.g., NEUP), with the industry partners, or with international partners. The specification has been completed and a number of sample solutions produced using a variety of DOE-used tools (TRITON, KENO, VERA/CASL), as well as industry tools (CASMO-SIMULATE) and NRC (POLARIS). Conclusions from the full set of results will be drawn next FY once all of the analyses have been completed.

Related to the benchmark, and a challenge-problem facing the ATF LTR/LTA is the ability to accurately predict

pin-power histories, and their associated achieved burnups. This information is used in the thermo-mechanical fuel performance assessments, and is imperative to be able to predict with confidence as it affects general design criteria such as center-line (melt) temperature, rod internal pressure etc. In particular, the initial loading of a ATF into an existing core of UO<sub>2</sub>-Zr fuel will cause heterogeneity in the assembly and/or core, and as such is a greater analytical problem for the tools. Analysis has been completed in which a single ATF pin, or single assembly, loaded into the core to evaluate the impact it would have on power histories and peaking factors. This is a key stage in establishing the licensing basis for test irradiations, as well as designing the fuel pellet and fuel rod designs, including the location and duration of the irradiation. The analysis has shown how the ATF LTR/LTA has a notable impact on local peaking factors, even when reducing enrichment. This results in a high burnup for the ATF fuel, likely to be much higher than viable for the first irradiation trials.

## 2.4 ATF CLADDING AND COATINGS

### High Temperature Oxidation Testing Of Cladding Materials

Principal Investigator: B. A. Pint

*This project is testing candidate accident tolerant cladding materials at up to 1700°C steam to assist in materials development efforts and support the development of performance models*

Compared to the current UO<sub>2</sub>/Zr-based alloy fuel system, alternative cladding materials need to offer slower oxidation kinetics and a smaller enthalpy of oxidation in order to significantly reduce the rate of heat and hydrogen generation in the core during a coolant-limited beyond design basis accident. The steam oxidation behavior of candidate materials is a key metric in the evaluation of ATF concepts and also an important input into models. The Severe Accident Test Station at Oak Ridge National Laboratory (ORNL) is able to measure steam oxidation behavior at up to 1700°C.

#### Project Description:

The project is examining the steam oxidation behavior of candidate materials suggested by the research community (i.e. community testing) and supporting ORNL work on the development of FeCrAl and SiC. The steam oxidation data being collected at 1200°-1700°C is unique as no prior work has considered steam oxidation of alloys at such high temperatures, particularly near the solidus temperature of FeCrAl at ~1500°C.

A key objective of the program is to develop reliable performance models. However, prior modeling work of FeCrAl cladding used incomplete information on the physical properties of FeCrAl. Therefore, this project is developing integral data on the melting points of oxides and alloys as well as the structural integrity of FeCrAl compared

to Zr-based cladding at up to 1700°C in steam. In addition, the steam oxidation behavior of FeCrAl after 1-yr exposures to LWR conditions was evaluated in ramp (i.e. heating at 5°C/min) testing up to 1500°C.

#### Accomplishments:

To support community testing, steam oxidation testing of the MIT Fe-12Cr-3Si was conducted, showing good steam oxidation up to 1000°C but more rapid attack at 1100°C where Fe-rich oxides formed at the specimen edges. Based on the model Fe-Cr alloy work at ORNL, it is possible that a combination of Mo, Mn and Si additions could reduce the required Cr content in a Fe-Cr alloy to lower than 20%. However, recent characterization work of a model Fe-Cr alloy showed that a protective SiO<sub>2</sub> layer did not form at the Cr<sub>2</sub>O<sub>3</sub>-metal interface after exposure in steam at 1200°C like it did at lower temperatures. Therefore, it is questionable if Fe-Cr alloys could form a protective oxide at temperatures above 1200°-1300°C and be a viable alternative to FeCrAl alloys, which form a more stable alpha-Al<sub>2</sub>O<sub>3</sub> scale and remain protective to ~1500°C in steam. High (>12%) Cr content alloys will be susceptible to alpha embrittlement under LWR-relevant irradiation temperature and dose regimes.

Another area that was investigated this year was the behavior of model FeCrAl alloys after exposure to LWR service conditions for 1 year. Ramp testing was performed in 1 bar steam

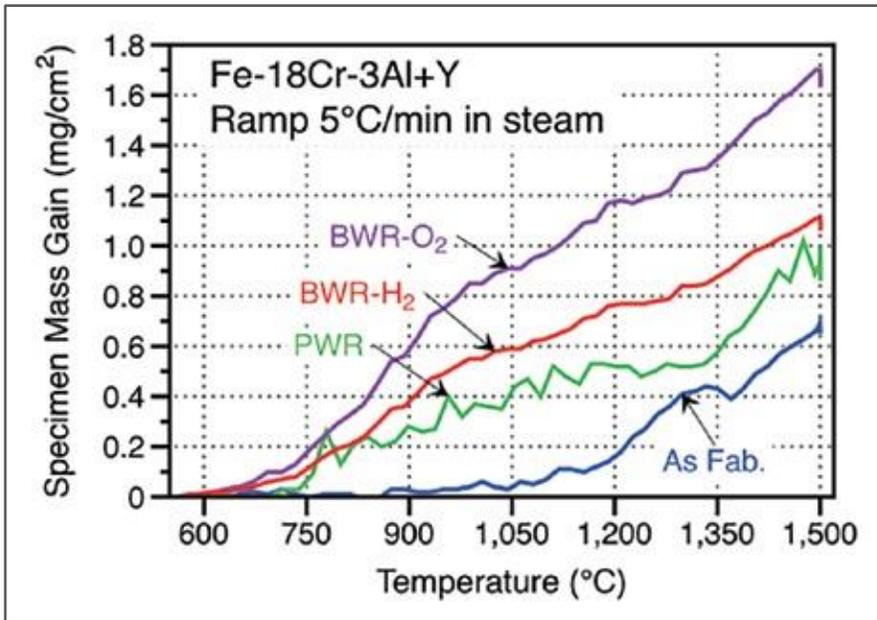


Figure 1. Specimen mass gain as a function of temperature during ramp testing (5°C/min heating) in 1 bar steam to 1500°C for Fe-18Cr-3Al+Y specimens with and without 1-year exposure to three different LWR operating conditions.

in the thermogravimetric analysis (TGA) on specimens with and without LWR exposures at General Electric under three different LWR environments. Figure 1 shows an example of the specimen mass gain data during ramp (5°C/min heating rate) testing in 1 bar steam in the TGA. The model Fe-18Cr-3Al+Y specimens all showed low mass gains to 1500°C and metallographic cross-sections of the specimens confirmed a thin protective oxide formed. Currently, transmission electron microscopy is being used to further characterize these oxides.

To support the development of FeCrAl performance models, experiments were conducted to measure the melting point of both oxides and alloys. To assess the mechanical integrity of the cladding after complete oxidation, assessments were made in steam to 1700°C. Tube specimens were ramped to temperature in ~90 min with an Ar purge to 600°C when the steam was introduced. After reaching temperature, the specimen was held for ~1 min before cooling to room temperature. Comparison experiments were conducted in dry air to examine the role of steam at these high

temperatures. A baseline was created for Zircalloy-4 with exposures from 1400°-1700°C. Similar experiments were conducted on commercial Kanthal alloy APM to 1600°C. As expected, the APM tube specimens survived testing to 1400° and 1500°C in steam and air with the formation of a relatively thin surface oxide. However, the APM specimen exposed in steam at 1600°C did begin to slump after exposure. Additional exposures up to 1700°C are in progress and new low-Cr FeCrAl tubing will be tested in the next quarter.

## Neutron Irradiation Effects in FeCrAl Alloys

Principal Investigator: Kevin Field

Collaborators: Samuel Briggs, Phillip D. Edmondson, Richard Howard, Yukinori Yamamoto, Kurt A. Terrani, Christian M. Petrie, Tarik A. Saleh, O. Anderoglu

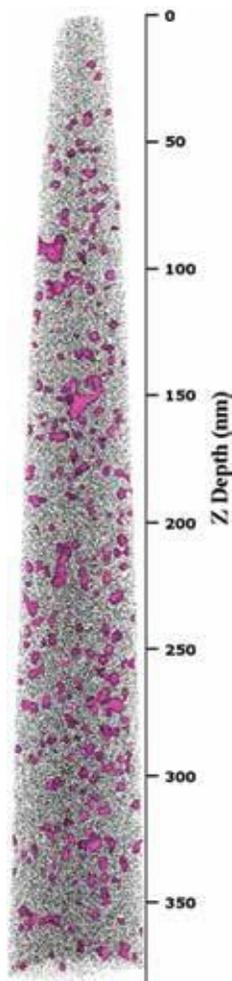


Figure 1: Atom probe reconstruction illustrating precipitate microstructure in the Fe-15Cr-7.7Al alloy, irradiated to 7 dpa at 320 °C. Precipitates are displayed using 34 at.% Cr concentration isosurfaces (purple) with 2% of total matrix Fe atoms shown (black).

The development and understanding of the radiation effects of neutron irradiated FeCrAl alloys is becoming a critical need as these alloys continue to be developed for accident tolerant fuel (ATF) applications. The use of materials test reactors, such as the Advanced Test Reactor (ATR) and the High Flux Isotope Reactor (HFIR), provides an avenue for accelerated testing and acquisition of data directly applicable to the radiation effects in these FeCrAl alloys. Post irradiation examination (PIE) including mechanical and microstructural evaluations of these irradiated specimens from materials test reactors provides the science-based understanding on the performance of FeCrAl alloys at prototypical light water reactor (LWR) operating environments.

### Project Description:

The primary objective of research into the neutron-induced radiation effects in FeCrAl alloys is to establish a materials database that includes both mechanical properties such as strength and ductility as well as microstructural features contributing

to the observed mechanical properties. Establishing a robust database, across several different irradiation conditions such as irradiation temperature, neutron dose (or flux) and alloy chemistry allows for trends associated with these variables to be established. Use of these trends, and distinct correlations between microstructural and mechanical properties, can be used to inform alloy development efforts in FeCrAl alloys. In particular, the chemistry and microstructure of FeCrAl alloys can be optimized for use within LWRs thereby providing a nuclear-grade FeCrAl alloy that can be deployed as an accident tolerant fuel cladding within the current commercial LWR fleet as well future nuclear reactors. Commercialization of an ATF cladding would result in greater safety margins during an accident scenario thereby meeting the DOE objectives of safe and reliable nuclear power production within the United States.

### Accomplishments:

Significant accomplishments include the completion of a materials database that includes 12 different FeCrAl alloys neutron irradiated in over 10 different unique irradiation conditions that spans 0-13.8 dpa at temperatures between 200

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and 550°C using three different materials test reactors (HFIR, ATR, and Halden). Spallation Neutron Irradiated material from the STIPV irradiation were also tested spanning 6-16 dpa at irradiation temperatures of 150-480°C. FeCrAl alloys studied to-date include Generation I FeCrAl alloys and Generation II FeCrAl alloys developed by ORNL in both tube and sheet product form as well as commercial alloys studied in sheet product form. A large portion of the irradiations were completed in a passive manner, i.e. no online measurements were made, but several active monitoring irradiations have been completed including an irradiation creep and swelling study. PIE has included mechanical testing, fractography, transmission electron microscopy (TEM), atom probe tomography (APT), small angle neutron scattering (SANS), and scanning transmission electron microscopy coupled with energy dispersive spectroscopy (STEM-EDS).

The established database derived from the PIE activities has enabled the determination of distinct trends in the microstructural formation and progression under irradiation and the

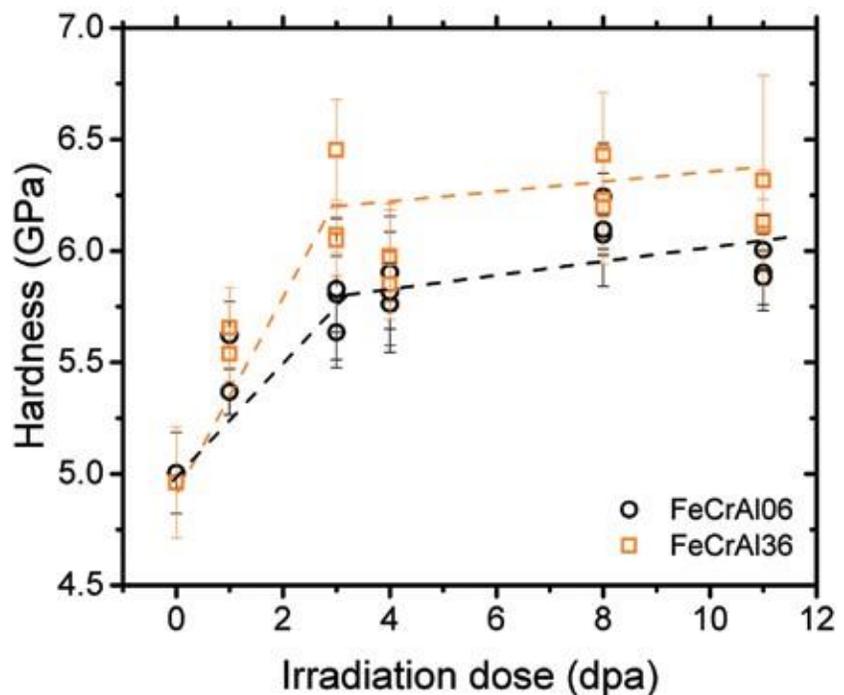
corresponding mechanical response. Primary microstructural features were found to include Cr-rich  $\alpha'$ , dislocation loops, black dot damage, and line dislocations. The magnitude, e.g. size and number density, of these microstructural features strongly depended on irradiation temperature, irradiation dose, the alloy of interest. Use of structure-property correlations revealed that the formation of the Cr-rich  $\alpha'$ , shown in the attached Figure, is a strong contributor to increases in strength and reduction in ductility when observed in significant number densities. The trends and fundamental understanding that were developed as a result of these studies were used to determine near-optimized compositions for FeCrAl alloys in terms of radiation tolerance and therefore continued to assist in efforts of commercializing FeCrAl alloys as an ATF cladding for LWRs.

## Ion Irradiation Effects in FeCrAl Alloys

Principal Investigator: Osman Anderoglu

Collaborators: Stu Maloy

Figure 1. Irradiation hardening as a function of irradiation dose in 5 MeV Fe irradiated FeCrAl at 300C.



High Dose irradiation data is needed for development of enhanced accident tolerant fuels for light water reactors. One of the leading alloys are FeCrAl alloys. Because of the long time it takes to obtain data in reactor (2-3 years), initial data is being obtained through performing irradiations with ions at Los Alamos National Laboratory.

### Project Description:

Ion irradiations have been attracted much attention recently to study irradiation damage in materials due to inexpensive operation, possibility of obtaining high doses orders of magnitude faster compared to neutrons and little or no activation issue due to irradiation with energetic ions. The objective of this study is to investigate irradiation damage at high doses (up to >10 dpa) and

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understand mechanical property changes using heavy ion irradiations in newly developed nuclear grade iron-chromium-aluminum (FeCrAl) alloys. Different alloy compositions have been produced for optimized manufacturability, neutron economy and manufacturability. Due to much higher displacement damage compared to neutrons, self-ions are proposed to investigate irradiation hardening. However, due to very limited irradiation depth (~2  $\mu\text{m}$ ), nanoindentation is proposed to probe the hardening from very shallow depths. The alloys are shown to form stable alumina scale when exposed to high temperature steam and outperform Zircaloy significantly. Therefore, the alloys were chosen as a more accident tolerant cladding option than the Zircaloy. However, investigation of higher dose performance of the alloys would take years in a test reactor. On the other hand, ion irradiations can be performed in a matter of hours or days depending on the ions species. Since the alloys are Fe based, Fe was chosen to irradiate the materials >10 dpa. Nanoindentation technique is proposed to test irradiation hardening.

#### **Accomplishments:**

5 MeV Fe<sup>++</sup> irradiations were successfully performed on the selected FeCrAl alloys. The total irradiation doses of 1, 3, 4, 8, and 11 dpa were obtained at 300C. Then Nanoindentation technique was used to measure hardness on reference as well as irradiated alloys at different doses. The average hardness was carefully calculated at around 100-200nm depth below the surface. The results show that both alloys quickly harden due to irradiation within 1-3 dpa as seen in Figure 1. Then hardening saturation was reached by ~8 dpa. No significant dependence on the composition was observed. As a result, both irradiation hardening, hardening saturation and the dependence on the composition were investigated successfully using self-ion irradiations and nanoindentation. Hardening saturation is observed in neutron irradiated ferritic alloys usually by 10 dpa. Therefore the results are consistent with neutron irradiations.

*Irradiation hardening and saturation of irradiation hardening were observed using heavy ion irradiations in nuclear grade FeCrAl alloys.*

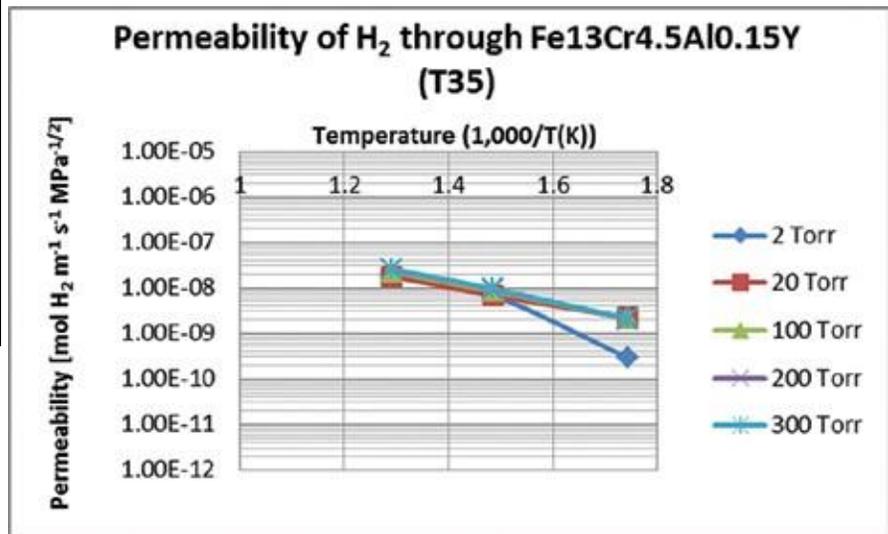
*- Osman Anderoglu*

## Tritium/Deuterium Diffusion Studies on ATF Cladding Materials

Principal Investigator: Stuart Maloy  
Collaborators: Joe Wörmer and Eric Tegtmeier

New measurements of deuterium diffusion were performed on FeCrAl tubes to investigate the permeability of tritium through these tubes under reactor operation.

Figure 1. Graph showing permeability of deuterium through a Gen I FeCrAl alloy (T35)



FeCrAl alloys are leading candidate materials for cladding for enhanced accident tolerant fuels. Significant data is required to qualify these materials as LWR fuels. One important aspect of the cladding is that it needs to hermetically seal the fuel keeping isotopes including tritium within the cladding during irradiation.

### Project Description:

FeCrAl tubes of different Gen I FeCrAl alloys were brazed in the deuterium diffusion measurement assembly. Tubes were pressurized with deuterium to pressures up to 40 kPa at temperatures from 400 to 600C. A residual gas analyzer measured the deuterium as it permeated through the tubes into the vacuum chamber. Initial measurements were on bare tubes and future measurements will be performed on tubes with an oxide coating.

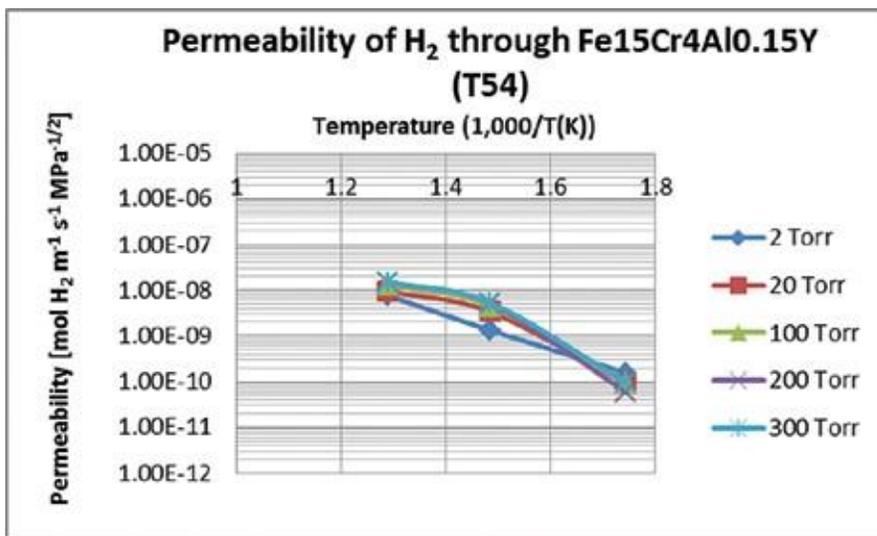


Figure 2. Graph showing permeability of deuterium through a second Gen I FeCrAl alloy (T54)

**Accomplishments:**

Deuterium diffusion measurements were completed on tubes of two Generation 1 FeCrAl alloys. Measurements were performed at different pressures up to 40 kPa and temperatures from 400 to 600C. Measurements were compared to those made on 316L showing a higher permeability in the FeCrAl alloys than an austenitic alloy like 316L. In addition, deuterium measurements

were compared to hydrogen measurements made at ORNL showing good agreement. Future measurements will be performed on tubes with a slight oxide layer on them to investigate the effects of this alumina based layer on permeability.

## FeCrAl ODS Alloy Development for Fission Platforms

Principal Investigator: Sebastien Dreyepont

Collaborators: Caleb Massey and Philip Edmonson

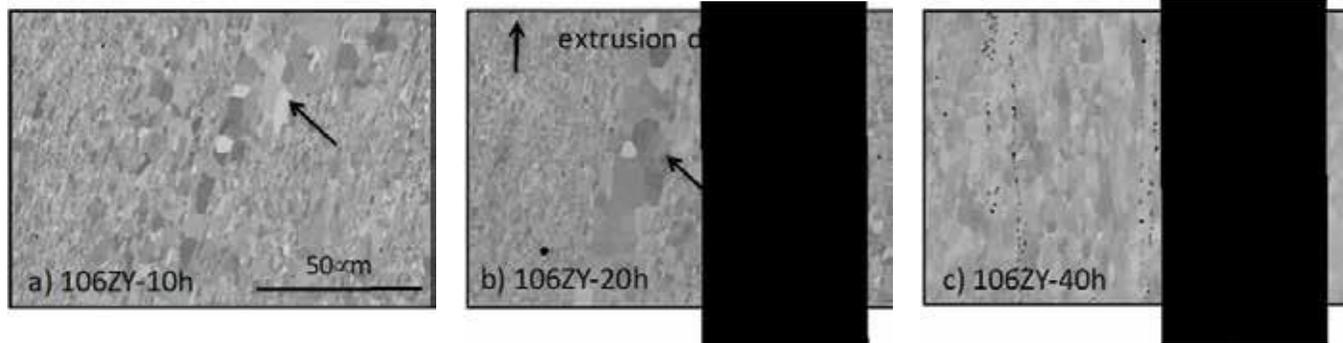


Figure 1. Back scattered scanning electron microscopy images showing the grain structure of alloy a) 106ZY-10h, b) 106ZY-20h, c) 106ZY-40h. Scale bars for a) and b) are five times larger than scale bars for c). Black arrows highlight large grain areas

After the Fukushima Daiichi accident, many research programs were initiated to develop new accident-tolerant fuel cladding materials with improved high temperature oxidation resistance. Advanced wrought and oxide dispersion strengthened (ODS) FeCrAl alloys are among the leading candidates due to their excellent oxidation behavior at temperature up to 1450°C in steam. The superior mechanical strength of these FeCrAl alloys up to ~800°C allows for the use of thinner cladding, thus limiting the neutronic penalty from the replacement of Zr-based alloys by Fe-based alloys. The high level of Cr in former commercial ODS FeCrAl alloys such as PM2000 or MA956 is, however, likely to result in alloy embrittlement during irradiation at 200-500°C because of the formation of the brittle alpha prime Cr-rich phase. ODS FeCrAl alloys typically

exhibit low ductility at  $T < 500^{\circ}\text{C}$ , which limits the alloy fabricability. New low-Cr ODS FeCrAl alloys with improved ductility need therefore to be developed.

### Project Description:

The overall project goal is to develop new low-Cr ODS FeCrAl alloys exhibiting great oxidation resistance in steam at  $T > 1400^{\circ}\text{C}$ , superior mechanical strength up to 800°C, and sufficient ductility at low temperature to allow for the production of tubes less than 500 micrometers thick. The first technical challenge is to optimize the alloy chemical composition to obtain the desired properties. Cr is known to improve the alloy oxidation performance, but Cr concentration needs to be low enough to avoid the formation under irradiation of the alpha prime phase. Minor additions such as Ti and Zr and impurity (C, N) content could also have a significant effect on the microstructure,

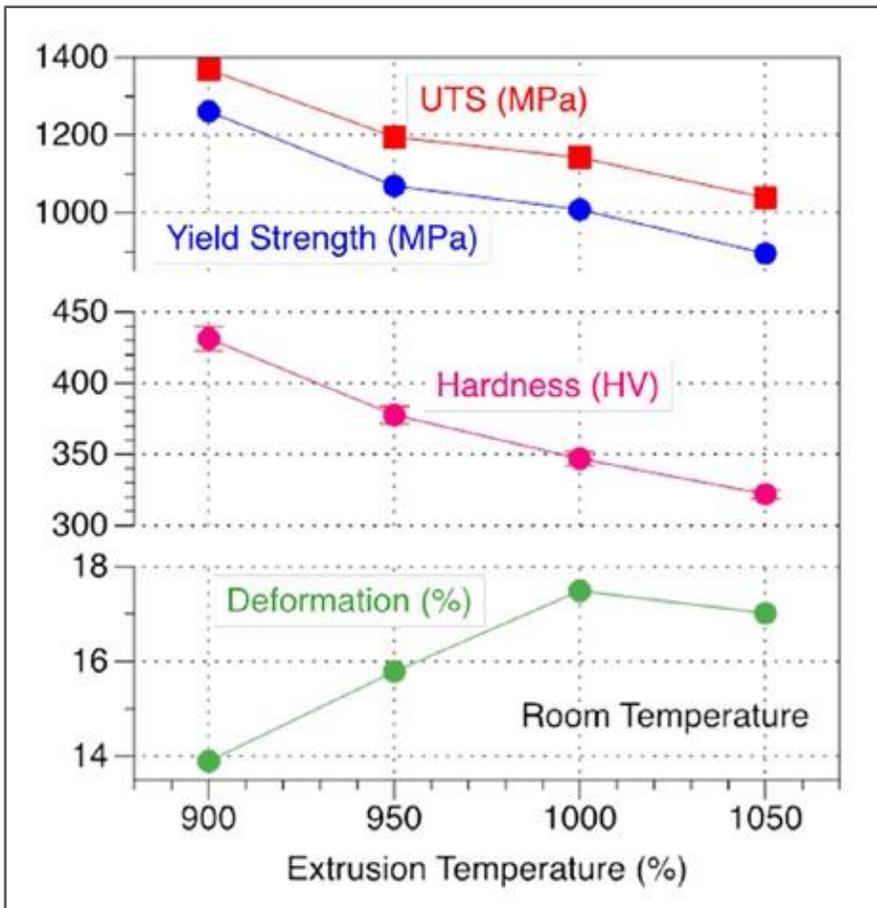


Figure 2. Evolution of the tensile properties and hardness values at room temperature for ODS Fe-10Cr-6Al-0.3Zr alloys extruded at temperature ranging from 900 to 1050°C.

mechanical properties and oxidation resistance of the alloys. The second main challenge is to understand the relationship between the fabrication route and the resulting alloy microstructure to produce thin tubes with the desired properties. Extensive work on ODS FeCr alloys revealed, indeed, that the nano precipitate size and density, as well as the alloy

grain structure, are directly related to the alloy fabrication process. Finally, the effect of irradiation on the microstructure and mechanical properties of ODS FeCrAl alloys needs to be assessed. The expected project outcome is the production of ODS FeCrAl cladding with enhanced safety margin for the current and next generation reactors.

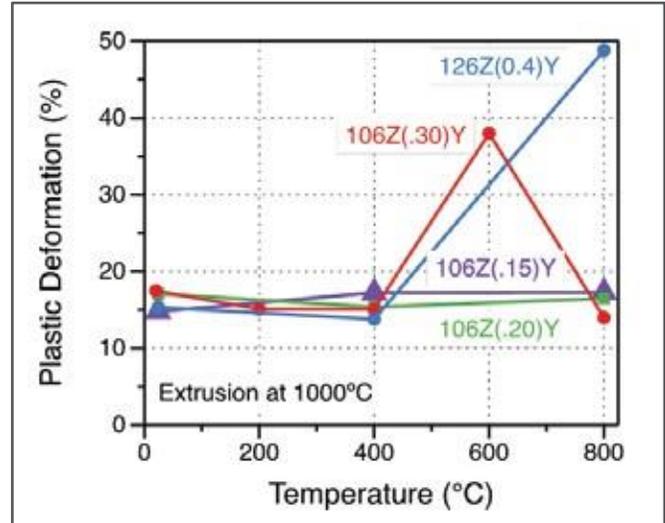
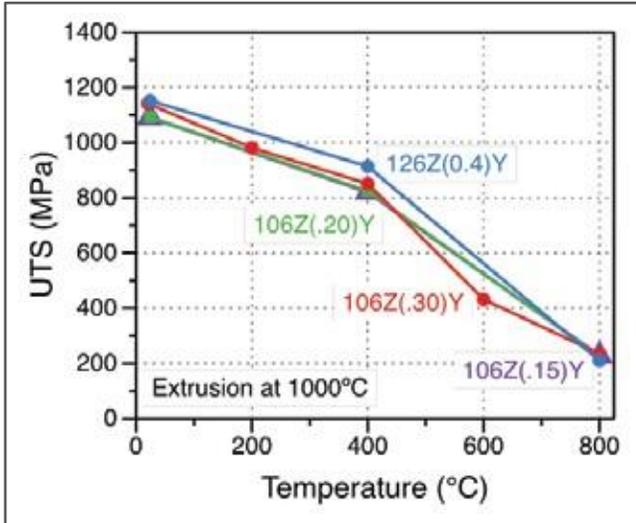


Figure 3. Tensile properties of several Fe-10-12Cr-6Al ODS FeCrAlZr alloys extruded at 1000°C with Zr concentration varying from 0.15wt% to 0.4wt%, a) UTS and b) Plastic deformation

#### Accomplishments:

Work on wrought FeCrAl alloys showed that a minimum Cr concentration of 10 to 12 wt% with an Al concentration of 5 to 6wt% was required to form a protective alumina scale at temperature > 1400°C in steam. Characterization in FY15 of several first generation ODS Fe-12Cr-5Al+Y<sub>2</sub>O<sub>3</sub> alloys suggested that Zr addition could improve the alloy strength. New Fe-10-12Cr-6Al-Zr powders were therefore ball milled with Y<sub>2</sub>O<sub>3</sub> powder to fabricate and characterize a second generation ODS FeCrAlZr alloys using different processing routes. Fe-10Cr-6Al-0.3Zr and Y<sub>2</sub>O<sub>3</sub> powders were ball

milled for 10h (106ZY10h alloy), 20 (106ZY20h) or 40h (106ZY40h) and then extruded at 950°C. Increasing the ball milling time led to an increase of the alloy strength and a decrease of the alloy ductility. As can be seen in Figure1, this ball milling effect is likely due to a decrease of grain size with increasing ball milling time, in addition to a bi-modal grain size distribution for the 106ZY10 and 106ZY20 alloys. Improvement of the alloy ductility may also be related to the decrease of the C and N concentration with shorter ball milling duration. The effect of extrusion temperature was also investigated, and Figure 2 shows idecreasing ultimate tensile strength, decreasing hardness and increasing

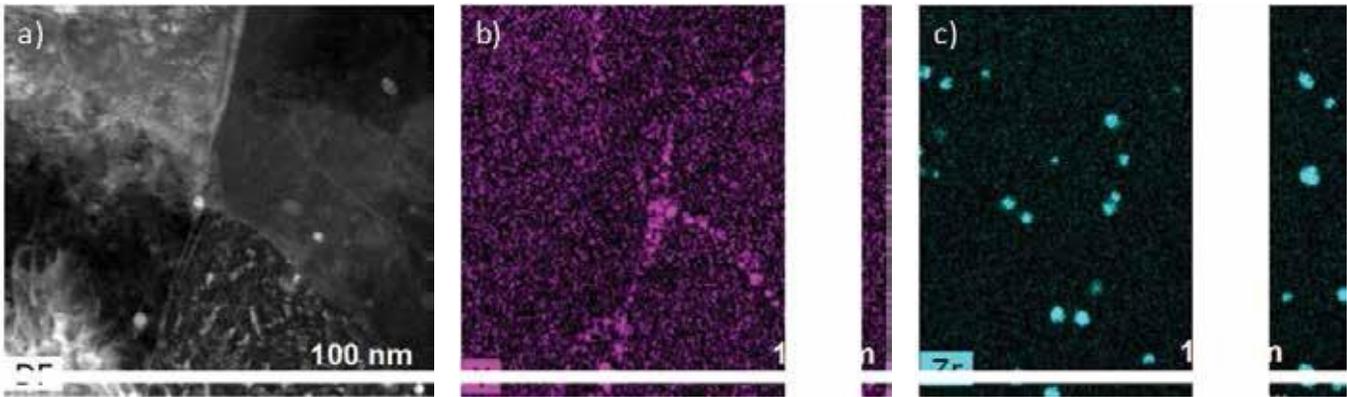


Figure 4. TEM micrographs of alloy 106ZY-40h showing segregation of Y at the alloy grain boundary and the formation of small Zr-rich carbonitrides

ductility when the temperature was raised from 900 to 1050°C. The last parameter studied was the Zr content in the Fe-10-12Cr-5-6Al alloys. Figure 3 shows that small variation of Zr concentration, from 0.15wt% to 0.5 wt% did not affect significantly the alloy tensile properties. Overall, all the ODS FeCrAlZr alloys showed good ductility at temperature ranging from room to 800°C, and initial transmission electron microscopy (TEM) characterization revealed the presence of Zr-rich carbonitrides and Y-rich precipitates at some grain boundaries (Figure 4). Further TEM work is required to confirm the presence of mainly (Y,Al)-rich oxides and Zr-rich carbonitrides, as was observed for a first generation

Fe-12Cr-5Al-0.3Zr alloy. The oxidation behavior of the second generation ODS FeCrAl was also evaluated, and many of these alloys formed a protective oxide scale after exposure for 4h at 1400°C in steam. A first generation ODS Fe-12Cr-5Al alloy showed recently lower embrittlement after irradiation at temperature above 300°C than wrought FeCrAl alloys, and irradiation of a few second generation ODS FeCrAl alloys will therefore be initiated in FY17. Finally, Fe-10Cr-6Al-0.3Zr master rods were produced to start the fabrication of thin tubes.

**New Fe-10-12Cr-6Al-Zr ODS alloys showing excellent steam oxidation resistance, great mechanical strength and good ductility were developed at ORNL for accident tolerant fuel cladding.**

## HFIR Irradiation of SiC Tubes under Prototypical Radial Heat Flux

Principal Investigator: Yutai Katoh

Collaborators: Christian M. Petrie, Takaaki Koyanagi, Gyanender Singh, Kurt A. Terrani, Christian Deck

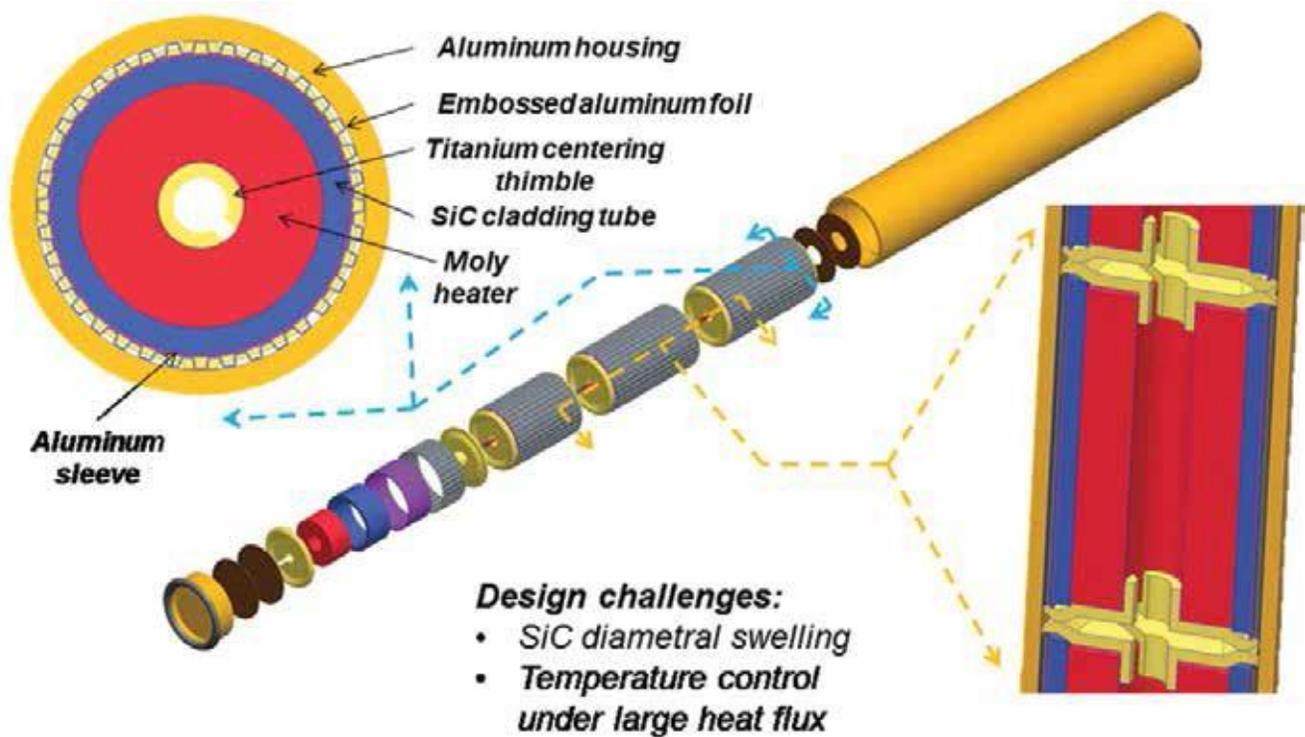


Figure 1. Illustrated design concept for innovative high heat flux “Fire Rabbit” vehicle that enabled study on the neutron irradiation – radial high heat flux synergism for development accident-tolerant fuel cladding for light water reactors.

Silicon carbide (SiC) - based fuel cladding for light water reactors is anticipated to develop a unique stress state as a combined result from the high radial heat flux and irradiation-induced swelling assisted by thermal conductivity decrease. In order to verify the multi-physics analysis of this unique stress state, a novel experiment capturing the synergism of a high radiation field and a high radial heat flux in tubular silicon carbide specimens had been designed and implemented in the High Flux Isotope Reactor.

### Project Description:

The SiC composite-based structures offer unparalleled combination of high temperature strength, irradiation tolerance, and severe accident performance among the candidate accident-tolerant fuel cladding concepts based on industrially-available engineering materials. However, during the fast neutron irradiation as the self-stabilizing radiation defects build up in SiC, accompanying the significant increase in thermal resistivity, the resultant differential swelling across the cladding thickness creates an anticipated unique secondary stress state.

This occurs in the material during the normal operation of the fuel when a high flux of neutron irradiation and a high radial heat flux are simultaneously present. Both the simplified analytical and detailed finite element modeling results indicate that magnitude of the stress will be high enough to probabilistically induce cracking on the inner surface of the cladding that experiences tensile stress. In the SiC-based cladding, the stress magnitude may result in a significant probability of cladding failure in typical operating conditions for the light water reactors. In order to verify the multi-physics finite element analysis predictions of this unique stress state, a novel experiment capturing the synergism of a high radiation field and a high radial heat flux in tubular SiC specimens had been designed and implemented in the High Flux Isotope Reactor, Oak Ridge National Laboratory. In this experiment, small diameter tubular test specimens made of monolithic or composite SiC were irradiated to a dose of  $\sim 2 \times 10^{25}$  n/m<sup>2</sup> ( $E > 0.1$  MeV) under a radial heat flux of  $\sim 0.6$  MW/m<sup>2</sup> while the outer surface temperature was maintained at a target temperature of  $\sim 300^\circ\text{C}$  achieving a steep temperature gradient through the cladding wall thickness. The initial investigation of irradiated tubular specimens indicated the temperature gradient along thickness of the tube wall as expected.

#### **Accomplishments:**

Specimens in a tubular geometry were irradiated under a high radial heat flux. The specimens were made of high purity CVD SiC, SiC/SiC composite, or a layered structure consisting of composite and monolithic CVD SiC. The composite and layered tubes were designed and fabricated by General Atomics. The irradiation is performed in the Flux Trap Rabbit facility of the High Flux Isotope Reactor (HFIR) at ORNL. Neutron fluence was chosen to be 2 dpa to ensure that the saturation is achieved for swelling and thermal conductivity change. The target temperature for the specimens at their outer surfaces were 573K. Achieving the target irradiation temperature is a significant challenge in this experiment due to the high heat flux and the uncertainty in in-situ thermal conductance across multiple contact interfaces. The radial heat flux was 0.6 MW/m<sup>2</sup> to represent the typical heat flux in LWR's while retaining the risk of tube cracking low.

An innovative "Fire Rabbit" irradiation vehicle was developed specifically for the present experiment at ORNL [1]. Bulky pieces of molybdenum were utilized as the heat sources. The biggest challenge in designing the

*This research addresses a critical technical feasibility issue for silicon carbide composite-based fuel cladding for light water reactors.*

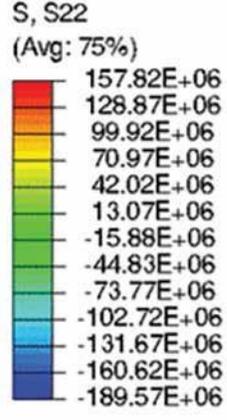
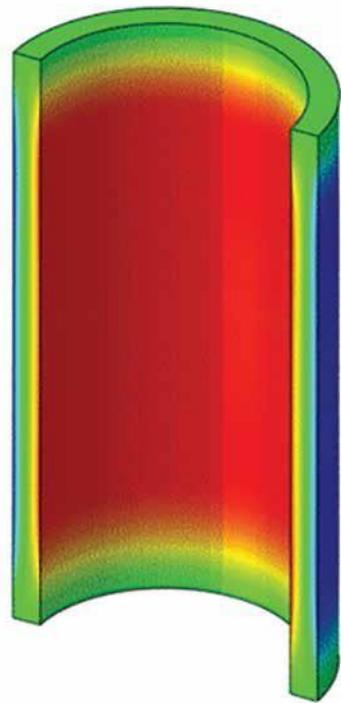
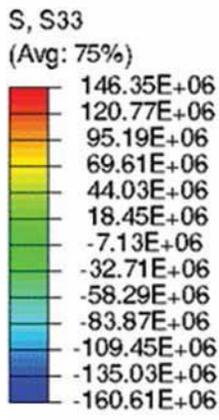
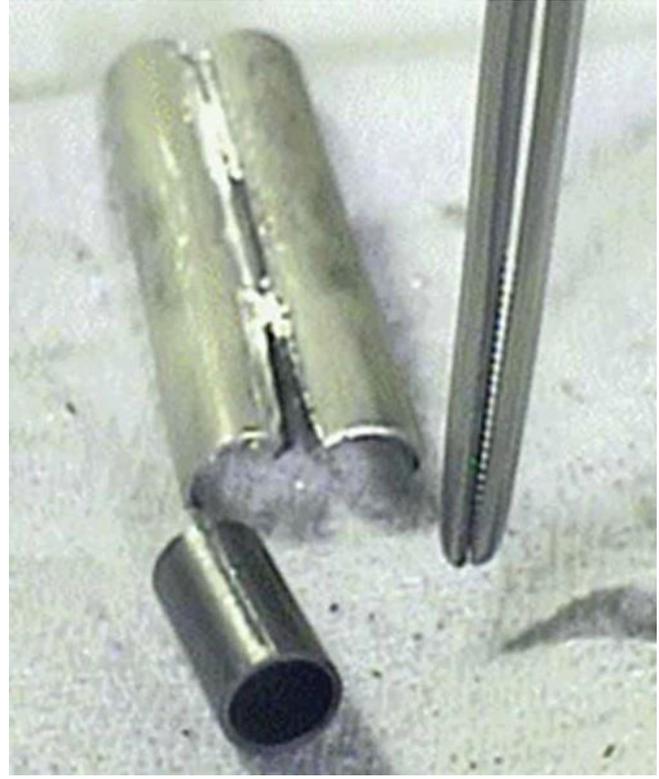


Figure 2. Stress distributions in SiC/SiC tube after cooldown to room temperature following ~2 dpa irradiation in Fire Rabbit experiment for study on the neutron irradiation – radial high heat flux synergism, estimated by a multi-physics finite element analysis.

vehicle was to develop a mechanism to maintain the irradiation temperature within an acceptable range under the rapid swelling of specimens at the beginning of irradiation. It was unable to adopt the commonly used gas gap technique to achieve the target temperature due to the extreme sensitivity of specimen temperature to the gap width. To overcome this difficulty, an embossed aluminum foil structure was engineered, minimizing the impact of tube swelling. Moreover, an extendable aluminum sleeve was developed to ensure the circumferential temperature homogeneity.

In parallel with the capsule design work, finite element modeling and analysis were performed to estimate the distributions of temperature and stress in the tubular test articles during and at the end of irradiation using the ABAQUS code [2,3]. The axial and hoop stresses during and after irradiation, and temperature distribution during irradiation were calculated. Although the present calculation is preliminary, it indicates that an axially uniform stress state is achieved for the majority of length of the specimen. Moreover, this stress analysis is useful to analyze the post-irradiation examination (PIE) results.



*Figure 3. Disassembly of Fire Rabbit vehicle in the 3025E Hot Cell Facility at Oak Ridge National Laboratory, following neutron irradiation in the High Flux Isotope Reactor. Three tubular specimens surrounded by engineered embossed aluminum foil sleeves were irradiated in single capsule (left). The SiC specimens were successfully retrieved after axially milling the capsule housing through its length.*

Two main goals of the PIE for this study are to determine radial distribution of irradiation temperature and stress state and radial distribution. To achieve these goals, the currently considered PIE items include 1) slit ring test on as-irradiated tube slices, 2) irradiation temperature mapping by micro-scale lattice strain measurement, and 3) isochronal annealing incorporating slit ring analysis,

in addition to more conventional examinations such as the macroscopic swelling and ultrasonic detection for micro-cracks [4]. Techniques that enable these PIE items including Raman spectroscopy temperature mapping and resonant ultrasonic spectroscopy were developed [5,6]. PIE is anticipated to take place in Fiscal Year 2017.

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## Thin-walled FeCrAl Tube Production

Principal Investigators: S. Maloy and Y. Yamamoto  
(Y. Yamamoto)



*The project has successfully established a commercial fabrication pathway of thin-wall seamless ATF FeCrAl alloy tubes which were traditionally unable to be produced because of poor deformability nature of FeCrAl alloys.*

The development of nuclear-grade enhanced accident tolerant fuel (ATF) cladding alloys targets a new, metal-base structural material for nuclear fuel cladding in Light Water Reactors (LWR). FeCrAl alloys were selected as candidate ATF cladding materials based on their excellent oxidation resistance in high temperature steam environments up to 1475°C (provided by the sufficient amounts of Cr and Al additions), compared

to the industry standard zirconium alloys which do not have such high temperature tolerances. This is the key for enhancing safety margins under severe accident conditions by limiting heat and hydrogen production, which occur when the fuel cladding reacts with steam during a severe accident. Thin-wall tube production of FeCrAl alloys, however, is a technical challenge since it was traditionally unable to fabricate because of poor deformability nature of FeCrAl alloys. Thus,

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it is critical for the ATF concepts to establish the production pathway of thin-wall seamless FeCrAl alloy tubes. This task targets to produce thin-wall FeCrAl tubes, by following the previous ATF FeCrAl alloy development efforts including the improvement of processability of FeCrAl alloys.

**Project Description:**

The objective of this task is to evaluate the feasibility of compressive deformation routes for thin-wall seamless FeCrAl tube production through commercially available tube fabrication processes. The end result is the determination if compressive deformation routes can be used to produce large quantities of the FeCrAl tube products through commercial manufacturers. Three different thin-wall tube fabrication approaches have been identified: cold-drawing, warm-drawing, and cold-pilgering, which are conducted by Century Tubes, Inc. (San Diego, CA), Rhenium Alloys, Inc. (North Ridgeville, OH), and Superior Tube Company, Inc. (Collegeville, PA), respectively. All process steps including melting, production of master bars, gun-drilling, and

thin-wall tube fabrication were conducted at commercial manufacturers. A part of the master bar production was made at Oak Ridge National Laboratory (ORNL) as well. The FeCrAl tube production to date including characterization results of the as-received tubes has been discussed. The final target is to establish the commercial mass production pathway of thin-wall seamless FeCrAl tubes with controlled microstructure which achieve sufficient qualities to meet with the requirements for ATF cladding concept such as the mechanical properties, size tolerance, reproducibility, weldability, as well as the oxidation resistance. .

Two different Gen. I FeCrAl alloys and three different Gen. II FeCrAl alloys were selected for tube production. The master columnar ingots with 3-4" diameter were prepared by a vacuum induction melting at Sophisticated Alloys, Inc. (Butler, PA), and the ingots were then extruded to produce master bar samples with 1" diameter at either

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ORNL or Sophisticated Alloys, Inc. The master bars were gun-drilled to prepare the master tubes with a uniform wall thickness of ~0.10" at each tube manufacturers or at Grover Gundrilling, LLC. (Oxford, ME).

**Accomplishments:**

Cold-drawing process at Century Tubes, Inc. has successfully produced thin-wall seamless tube of Gen. I and Gen. II alloys and met the dimensional requirements. Gen. II alloy tubes tend to show finer grains (~20-50  $\mu$  m) than those of Gen. I alloy tubes (~80-100  $\mu$  m) due to the additional alloying of Mo in the Gen. II alloys leading to increased resistance to grain coarsening during inter-pass annealing. Several master tubes showed cracks along the axial direction during the early stages of

the drawing process, indicating that the hoop stress introduced during the area reduction was significant. Elimination of this effect required balancing the optimized amount of the area reduction per pass (by selecting small changes in the size of the die and the mandrel as possible) and a proper inter-pass annealing. A large quantity of Gen. I alloy tube segments (B136Y3, total length: ~480 ft, shown in Fig. 1) were delivered on August 2016, and several segments were provided to collaborators inside ORNL for property evaluation.

The warm-drawing process was conducted by Rhenium Alloys, Inc. in expectation that the increased temperature would lead to better tube fabricability, since the deformation resistance of FeCrAl alloys significantly dropped above ~200-300°C. However, no significant

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advantage in the tube fabricability was observed compared to cold-drawing process; the crack formation along the axial direction was observed occasionally, similar to the cold-drawing process. After production failure of several Gen. II alloy tubes, optimized processing conditions were introduced to balance the improved fabricability and provide microstructure control. Another large quantity tube production of Gen. II alloy (C35M4, expected 250 ft. length) is currently in progress.

Cold-pilgering process of Gen. II alloys at Superior Tube Company, Inc. was originally scheduled to be completed in August 2016. However, the compressive reduction process has not been initiated within this

year because of the delay of the machined tool delivery to Superior Tube Company, Inc. Modification completion schedule of the tube reduction process is November 2016.

Although the cold-pilgering has not been completed yet, it can be summarized that the cold-drawing process is the most feasible candidate for mass production of thin-wall seamless ATF-FeCrAl tubes to date. A discussion with Century Tubes, Inc. was initiated regarding the next procurement of Gen. II FeCrAl tube production.

***The project has successfully established a commercial fabrication pathway of thin-wall seamless ATF FeCrAl alloy tubes which were traditionally unable to be produced because of poor deformability nature of FeCrAl alloys.***

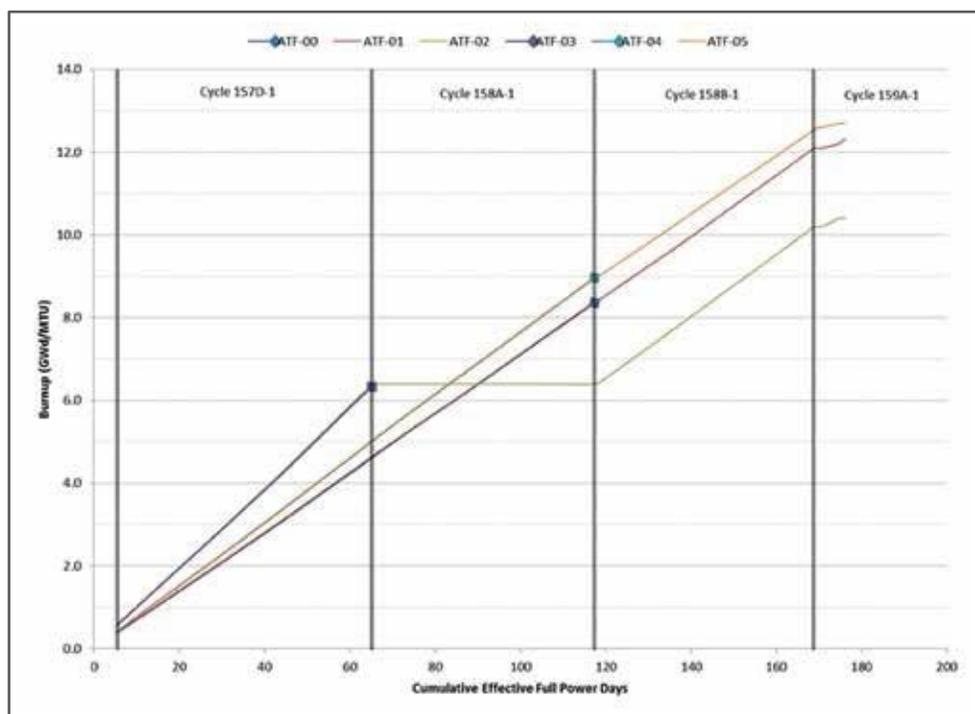
# 2.5 IRRADIATION TESTING AND PI TECHNIQUES

## ATF-1 Irradiation Testing in ATR

Principal Investigator: Gregory Core

Collaborators: Connor Woolum, Nate Oldham, Dan Chapman, Bryon Curnutt, Kelly Ellis, Steven Galbraith, Glenn Moore

Figure 1. Burnup as a function of Cumulative Effective Full Power Days of AREVA Capsules ATF-00 through ATF-05



The purpose of the Accident Tolerant Fuels (ATF) experiments is to investigate materials and fuels to replace or enhance the current uranium-oxide fueled, zirconium-based clad system used in Light Water Reactors (LWR). At the Idaho National Laboratory (INL), a diverse set of experiments is currently undergoing irradiation

at the Advanced Test Reactor (ATR). These static irradiations, known as “drop-in capsule experiments” are referred to collectively as ATF-1. Due to the fact that these ATF-1 experiments are not in direct contact with the primary coolant of the ATR, the primary goal of the ATF-1 experiments is to determine the physical, thermal, and chemical changes of the various cladding and fuel concepts after irradiation.

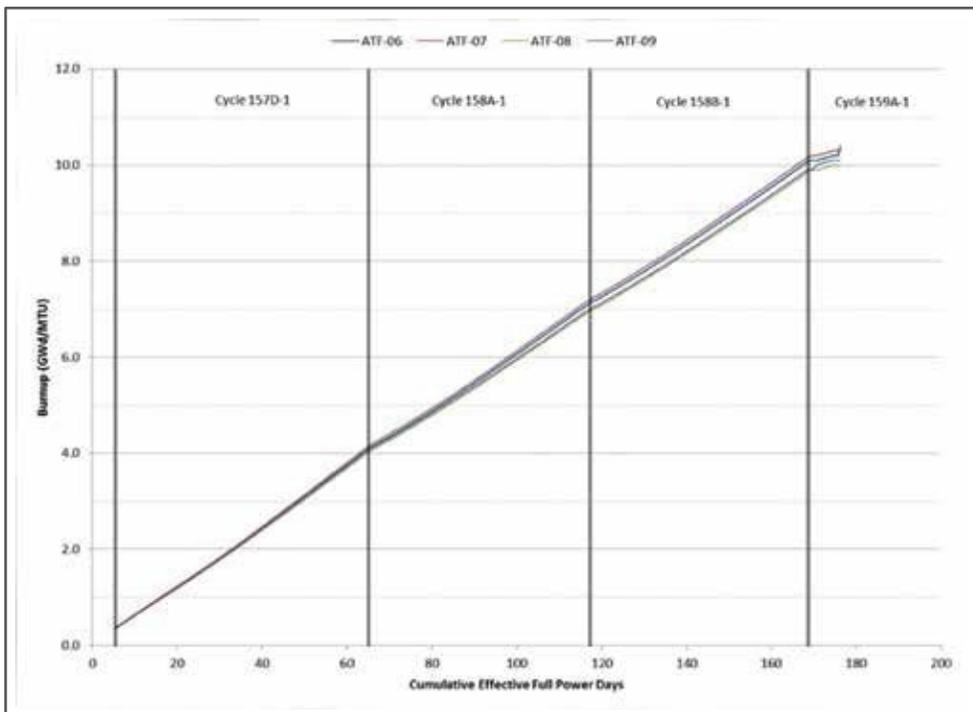


Figure 2. Burnup as a function of Cumulative Effective Full Power Days of GE Capsules ATF-06 through ATF-09

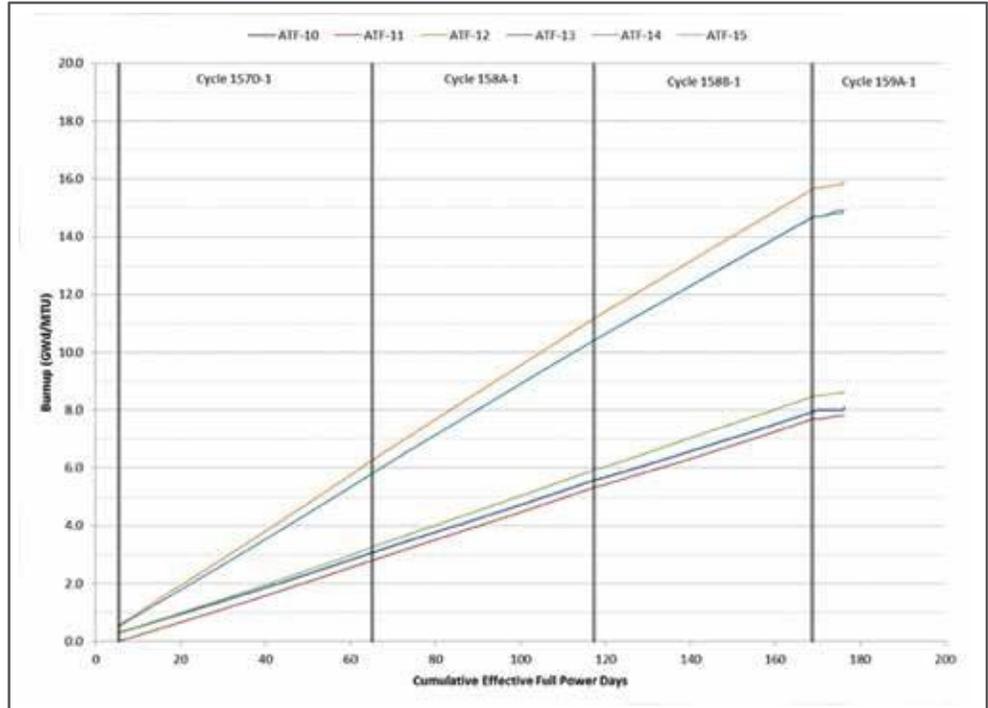
**Project Description:**

As mentioned, the ultimate goal of the ATF program is to demonstrate improved fuel and/or cladding concepts offering the potential to replace the Zircaloy-UO<sub>2</sub> system currently used throughout the LWR industry. To support this goal, the congressional appropriation language for FY 2012 included specific language for DOE NE to initiate an aggressive Research, Development,

and Demonstration (RD&D) program for LWR fuels with enhanced accident tolerance. The test data collected as part of the ATF program will support demonstration of lead test rods (LTRs) or lead test assemblies (LTAs) in a commercial LWR within 10 years (i.e., by the end of FY 2022).

As a step toward this goal, a Phase 1 (Figure 1) feasibility irradiation test series of drop-in capsule

Figure 1. Burnup as a function of Cumulative Effective Full Power Days of AREVA Capsules ATF-00 through ATF-05



experiments, denoted ATF-1, was fabricated and inserted into the Idaho National Laboratory (INL) Advanced Test Reactor (ATR) in FY 2015 to demonstrate fabricability and viability for ATF-2 (Phase 2) water loop testing (e.g., hermeticity, fuel/clad performance, structural stability). As part of feasibility testing, ATF-1 experiment fuel cladding is not exposed to the ATR primary coolant system (PCS) to ensure ATR safety in the event of breached fuel rodlets. The ATF-2 ATR water loop experiments are a continuation of the ATF-1 drop-in

capsule feasibility experiments with the primary objective of testing ATF concepts under conditions prototypic of Pressurized Water Reactors (PWR) to demonstrate concept viability, thus exposing the fuel pins (rodlets) directly to the PWR water chemistry and flow.

Upon reaching programmatically defined irradiation test objectives, ATF experiments will be discharged from ATR and shipped to the INL Materials and Fuels Complex (MFC) for post-irradiation examination (PIE) and/or Transient Testing in the Transient Reactor Test (TREAT) facility.

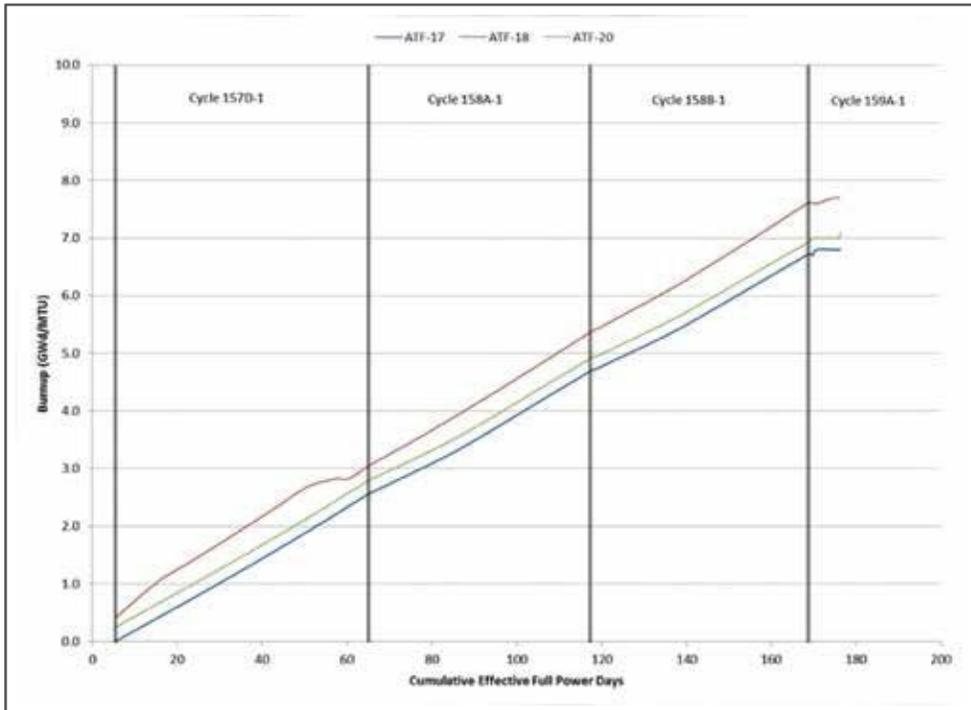


Figure 2. Burnup as a function of Cumulative Effective Full Power Days of GE Capsules ATF-06 through ATF-09

### Accomplishments:

As the purpose of the irradiation test is to provide irradiated samples to post-irradiation examination (PIE) experts and scientists, the ATF-1 experiment has completed the irradiation in the ATR of three ATF concept capsules and subsequently shipped those capsules to INL's MFC to begin PIE. This effort alone required the input of many individuals and parties including Nuclear Safety, Packaging and Transportation, ATR reactor engineering staff, Nuclear Science and Technology Staff at INL, Experiment Fabrication Engineers, and numerous crafts (e.g. machinists, radiological

technicians, etc.) Furthermore, analysts at the INL and experiment support staff have allowed the experiments to continue irradiation in the ATR by confirming safety parameters and shuffling the experiment capsules. This continual analysis allows for both the programmatic and safety requirements of the experiment to be met with precision.

Additionally, eight capsules were fabricated and inserted into the ATR for irradiation in 2016, bringing the total number of capsules irradiated to date to 27.

**The ATF-1 irradiation testing experiment provides experimental fuel and fuel cladding combinations to PIE with quality documentation allowing for accurate understanding of the effects of irradiation on new LWR fuel system concepts.**

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## ATF-1 Experiment Fabrication

Principal Investigators: Glenn Moore, Connor Woolum

Collaborators: Nate Oldham and Greg Core

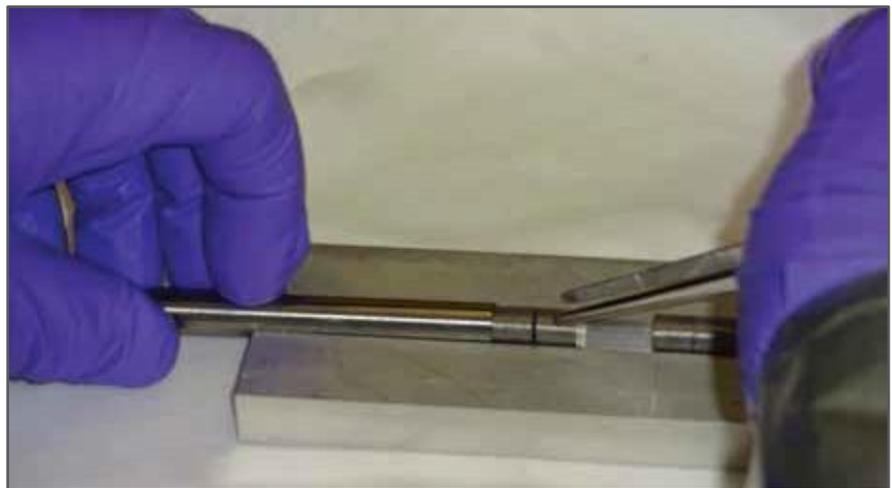


Figure 1. Loading of fuel pellets into an ATF rodlet.

The Accident Tolerant Fuels (ATF) program aims to develop a fuel system exhibiting improved performance, safety, and reliability over the current UO<sub>2</sub>-Zircaloy system. In conjunction with partners from industry and other national laboratories, candidate fuel-cladding test specimens were prepared for irradiation testing in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL).

### Project Description:

The ATF program has developed a three phase approach leading to commercialization of an accident tolerant fuel concept. Phase 1 includes drop-in irradiation experiments in the form of fueled rodlets encapsulated inside of a stainless steel pressure boundary (capsule). These drop-in assemblies fabricated in Phase 1 are intended to be a feasibility study

to aid in the prioritization of candidate fuel/cladding concepts as the transition is made to Phase 2. Phase 2 includes loop testing in ATR under prototypical pressurized water reactor (PWR) conditions. Transient testing of both fresh and previously irradiated fuel in the Transient Reactor Test Facility (TREAT) is another, later component of Phase 2. These two phases support the insertion of a Lead Fuel Rod (LFR) or Lead Fuel Assembly (LFA) in a commercial reactor in 2022.

The commercialization of an accident tolerant fuel concept will have many benefits including those beyond the increased safety margin. The new fuel system will provide reactor operators with increased coping time to respond to any catastrophic events, due to the inherently safe nature of the fuel system.



Figure 2. Micro-TIG torch setup in inert atmosphere glovebox for closure weld of weep hole on ATF rodlets.



Figure 3. From the top down: Example of ATF-1 style capsule, welded rodlet, fuel pellets, and non-welded rodlet.

The program is currently transitioning from Phase 1 to Phase 2. This past fiscal year, nine full rodlet and capsule assemblies were fabricated and qualified to begin irradiation testing in ATR this fiscal year. The fabrication of these experiments was a collaborative effort.

#### Accomplishments:

Several rodlet and capsule assemblies were fabricated and qualified at INL in FY2016. A total of eleven rodlet/capsule drop-in style assemblies were fabricated in support of the ATF-1 series of experiments. Nine of these were qualified for insertion and irradiation testing in ATR and two of these were fabricated to support development of advanced non-destructive examination (NDE) techniques at Los Alamos National Laboratory (LANL). Six of the nine assemblies fabricated for irradiation testing contained a UN-U3Si2 fuel as part of a Westinghouse led concept.

Additionally, three of the nine rodlets were a LANL led concept, assembled at INL for irradiation testing.

The fuel for the six Westinghouse assemblies was fabricated by both INL and LANL. The U3Si2 component was fabricated at INL, then shipped to LANL who fabricated the UN component of the fuel then pressed and sintered pellets. Final centerless grinding and inspection of the fuel occurred at INL, followed by rodlet assembly. These rodlets made use of an endcap design that had been modified since the initial ATF-1 assembly campaigns. The new endcap design incorporated several features deemed beneficial to assembly such as a weep hole and a new mechanism for centering the rodlet within the capsule. These six assemblies were inserted in ATR for irradiation beginning in Cycle 160A-1.

A total of five rodlets were assembled at INL utilizing U3Si5 and UN-U3Si5 fuel pellets manufactured at LANL. This LANL led concept included Kanthal-AF as the concept cladding material. The weld development work necessary to assemble the rodlets, along with final rodlet fabrication, assembly, and encapsulation was performed at INL. Two of these five capsule assemblies are to be shipped to LANL for use as mock-up specimens supporting development of advanced NDE methods. Two of the remaining three LANL assemblies were inserted in ATR, beginning irradiation testing in Cycle 160A-1. The remaining LANL capsule is slated to begin irradiation testing in December 2016 in ATR Cycle 160B-1.

## ATR ATF-2 Loop Design

Principal Investigators: Heather Chichester and Kristine Barrett

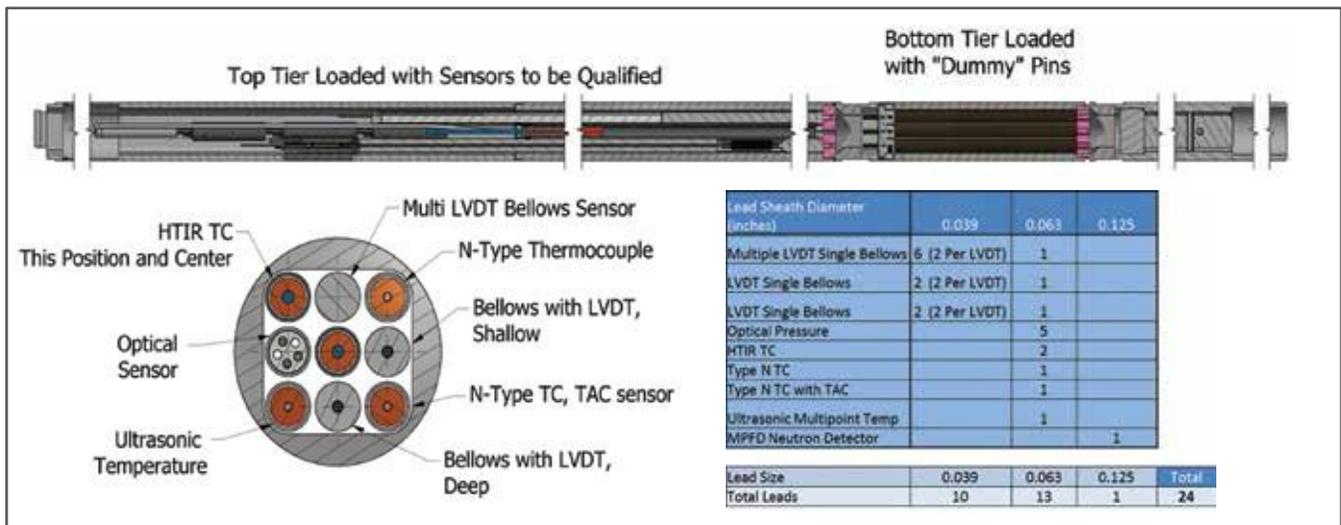


Figure 1. SQ Test Sketch and cross section identifying sensors and associated wiring leads (i.e., lead arrangement) to be included in the top tier. Lower tier will contain cladding only (i.e., "dummy") pins to test corrosion and/or other material degradation/alteration.

The Idaho National Laboratory (INL) has been tasked with the responsibility for conducting irradiation experiments in the Advanced Test Reactor (ATR) for the purpose of assessing the performance of Accident Tolerant Fuels (ATF) concepts under prototypic Pressurized Water Reactor (PWR) operating conditions in an ATR water loop with controlled and monitored water temperature, flow, and chemistry. INL is leading the planning, design, and analyses of these irradiation experiments in coordination with Industry Partners that are engaged in developing the ATF concepts. INL will perform these irradiation experiments in the ATR and will coordinate the postirradiation examination (PIE) on

the discharged fuels. The discharged fuels will be shipped from ATR to the Hot Fuel Examination Facility (HFEF) at the Materials & Fuels Complex (MFC) or the Transient Reactor Testing (TREAT) facility, as desired by the ATF concept development team for each fuel pin.

### Project Description:

The mission of the ATF Initiative is to develop the next generation of LWR fuels with improved performance, reliability, and safety characteristics during normal operations and accident conditions and with reduced waste generation. Enhancing the accident tolerance of the fuel is the focal point of the initiative. The initial RD&D efforts are to focus on applications in operating reactors or reactors with design certifications. However,

what is learned and developed during this process may be applicable to the design of the next generation of LWRs.

The ultimate goal of the Accident Tolerant Fuels (ATF) program is to demonstrate improved fuel and/or cladding concepts offering the potential to replace the Zircaloy UO<sub>2</sub> system currently used throughout the Light Water Reactor (LWR) industry. To support this goal, the congressional appropriation language for FY 2012 included specific language for DOE/NE to initiate an aggressive RD&D program for LWR fuels with enhanced accident tolerance. The test data collected as part of the ATF program will support demonstration of Lead Fuel Rods or Lead Fuel Assemblies in a commercial LWR by the end of FY 2022. The ATF-2 ATR water loop experiments are a continuation of the ATF-1 drop-in capsule feasibility experiments with the primary objective of testing ATF concepts under PWR prototypic conditions to demonstrate concept viability.

#### **Accomplishments:**

ATF-2 irradiation testing work performed in FY 2015 included: (1) design and analysis of a Sensor Qualification Test (SQT) in the ATR prior to inserting a fully loaded fueled test train (Figure 1), (2) continued conceptual design and scoping of the ATF-2 fueled test train in preparation for final design analysis, (3) fuel rod assembly development and mock-up

demonstrations using dummy fuel rod materials, (4) identification and procurement of assembly equipment such as induction brazing equipment, a laser welder, 4 axis workcell, and portable x-ray radiography equipment, (5) design and planning for an out-of-pile flowing autoclave test replicating ATR water loop conditions for the SQT (Figure 2), and (6) design and procurement of sensors to be used in the autoclave, SQT, and ATF-2 experiments. INL collaborated with the Institute for Energy Technologies (IFE) in instrumented lead assembly development, sensor design and preparation for wiring at the ATR, and preparation for the autoclave test. INL is subcontracting the autoclave test support to Westinghouse to utilize existing autoclave equipment at their Churchill, PA research facility.

A final conceptual design for the ATF-2 water loop experiment has been established in collaboration with the ATF Industry Partners Westinghouse, General Electric, and AREVA (Figures 3 and 4). The final design information for each of the Industry Partners is documented in FCRD reports. The final conceptual design will be used to perform final design safety analysis for ATR insertion, finalize design drawings, and perform fuel performance analysis for each ATF concept.



*Figure 2. Autoclave at the Westinghouse Research Laboratory to be used to examine sensor and cladding durability under prototypic PWR and ATR flow conditions prior to ATR insertion.*

## ATF-2 Fabrication Development

Principal Investigator: Connor Wbolum

Collaborators: Clint Baker, Brian Durtschi, Kristine Barrett, Kyle Kofford, Kip Richards

*The ATF-2 experiment will require fabrication of many one-of-a-kind assemblies; FY16 efforts set the stage for successful fabrication campaigns.*

Figure 1. Induction brazing equipment installed in HTTL.



The Accident Tolerant Fuels (ATF) program aims to develop a more robust and accident tolerant fuel system than the current UO<sub>2</sub>-Zircaloy system. The new fuel system must exhibit increased safety, performance, and reliability over the current system in order to be a viable

candidate. Several concepts are undergoing evaluation in order to prioritize the candidates. The ATF program is currently transitioning from drop-in style irradiation tests to a loop test in the Advanced Test Reactor (ATR) to characterize performance under prototypic pressurized water reactor conditions (PWR).

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### Project Description:

The ATF program has adopted a three phase approach leading to commercialization of accident tolerant fuel(s) in 2022. Phase 1 of this three phase approach includes drop-in irradiation experiments in the form of rodlets encapsulated in a stainless steel pressure boundary (capsule). Irradiation testing is underway at ATR, with three initial ATF-1 assemblies having already reached their burnup targets. These three assemblies are currently undergoing post irradiation examination (PIE). This first phase of ATF testing is a feasibility study to assist with prioritization of the candidate fuel/cladding systems. This leads to Phase 2 which begins with irradiation testing in the 2A loop of ATR. This test will subject the candidate fuel/cladding to prototypic PWR conditions including chemistry, pressure, and temperatures. Transient testing of both fresh and previously irradiated specimens is included as a later part of Phase 2. This is slated to occur at the Transient Test Reactor Facility (TREAT) at INL's Materials and Fuels Complex (MFC).

The loop testing to occur in Phase 2 will consist of both instrumented and non-instrumented fuel pins. The instrumented fuel pins may either be instrumented lead pins for in-situ monitoring during irradiation, or

the fuel pins may contain internal instrumentation to allow for measurements in between reactor cycles. This instrumentation will allow for fuel pin internal pressure measurement, fuel and/or cladding elongation, and temperature measurements during irradiation. These fuel pins will be the first of their kind to be assembled at INL with instrumented leads, requiring both brazing and welding as joining techniques.

### Accomplishments:

Fabrication of the ATF-2 experiment will include many complex efforts over the coming months. The ATF-2 experiment involves an autoclave test to verify performance of sensors to be used in the ATF-2 fueled test. A Sensor Qualification Test (SQT) will be performed in the same loop of ATR that the ATF-2 fueled test will be irradiated in. This will demonstrate performance of sensors to be used for measurement of fuel pin characteristics under like-for-like irradiation conditions. These efforts culminate in the ATF-2 fueled test. Each of these tests require separate fabrication efforts, although there are commonalities that will allow for knowledge transfer from one effort to another.



Figure 2. Example endplug welded as part of laser welder demonstration; material is SS316L.

*Figure 3. Sample brazes produced to demonstrate feasibility of brazing as an appropriate joining method for ATF-2 geometries.*

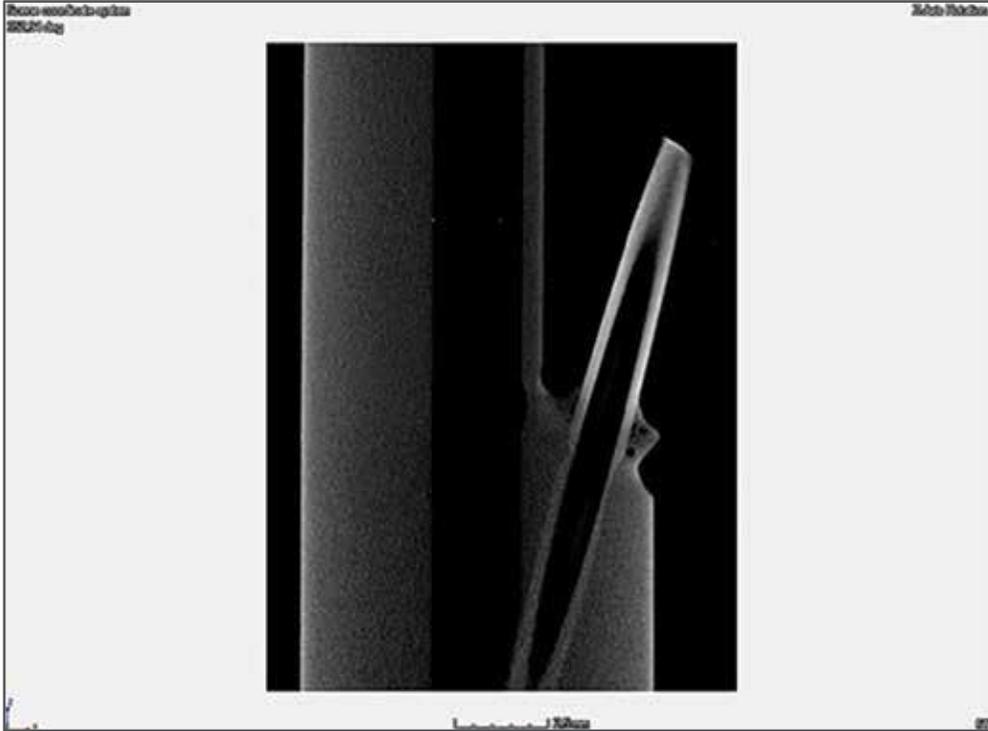


In order to support the upcoming assembly of these experiments and their components, several activities were performed in fiscal year 2016. This includes procurement and installation of induction brazing equipment, demonstration and selection of a laser welder, and selection and procurement of ancillary support equipment.

Induction brazing was chosen as the primary method for joining instruments to the endplug of experimental assemblies. Brazing provides a robust joint utilizing a well-established

technology enabling the experimental assembly to remain hermetically sealed throughout irradiation testing. Induction brazing equipment was procured and installed in the High Temperature Test Laboratory (HTTL) located within the Energy Innovation Laboratory (EIL) at INL. HTTL serves as the fabrication location of many instruments to be tested within ATF-2, thus collocation of the induction brazing equipment provides many benefits.

The ATF-2 experiments require a multitude of welds of various materials, ranging in thickness from



*Figure 4. Cross-section of a 3D CT scan of a sample braze joint.*

approximately 0.008” to 0.025” .A robust and repeatable welding method was desired, and laser welding was selected as the primary joining technique to support assembly of ATF-2 experiments. Sample components were sent to multiple vendors, who were asked to demonstrate the ability of their equipment to meet program needs. Based on these evaluations, a 700W Laser Welding 4-Axis Workcell, manufactured by IPG Photonics, was selected for procurement. An order has been placed and the welder is anticipated to arrive at INL in February 2017. Initial weld development work will be performed at the vendor’ s

facility, prior to the welder arriving at INL, in order to support necessary assembly schedules.

In addition to the brazing and laser welding efforts, partial support was provided for various equipment and capability enhancements. This includes laser etching equipment, a radiography source for digital radiography equipment, and a new portable x-ray unit. The staging of these tools will result in increased efficiency and improved results during the fabrication of ATF-2 assemblies.

## Halden 3D Fuel Experiment

Principal Investigators: Heather Chichester and Kristine Barrett

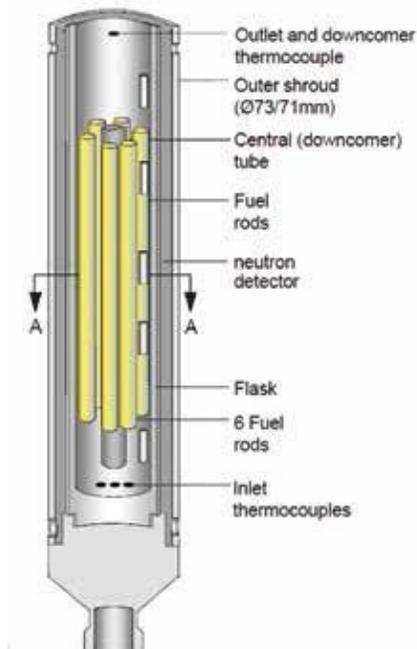


Figure 1. Typical test rig loaded with instrumented fuel rods for testing in the HBWR (provided by Halden Reactor Project [HRP]).

The Idaho National Laboratory (INL) has been tasked with the responsibility for conducting irradiation experiments in the Advanced Test Reactor (ATR) for the purpose of assessing the performance of Accident Tolerant Fuels (ATF) concepts under prototypic Pressurized Water Reactor (PWR) operating conditions in an ATR water loop with controlled and monitored water temperature, flow, and chemistry. However, the ATR is not capable of testing ATF concepts under Boiling Water Reactor (BWR) prototypic conditions. Therefore, INL is collaborating with the Institute for Energy Technology (IFE) in Halden, Norway to perform test rig design and scoping analysis of ATF concepts in the Halden Boiling Water Reactor (HBWR) to augment the data collected in the ATR.

### Project Description:

The mission of the ATF Initiative is to develop the next generation of LWR fuels with improved performance, reliability, and safety characteristics during normal operations and accident conditions and with reduced waste generation. Enhancing the accident tolerance of the fuel is the

focal point of the initiative. The initial RD&D efforts are to focus on applications in operating reactors or reactors with design certifications. However, what is learned and developed during this process may be applicable to the design of the next generation of LWRs.

The ultimate goal of the Accident Tolerant Fuels (ATF) program is to demonstrate improved fuel and/or cladding concepts offering the potential to replace the Zircaloy UO<sub>2</sub> system currently used throughout the Light Water Reactor (LWR) industry. To support this goal, the congressional appropriation language for FY 2012 included specific language for DOE/NE to initiate an aggressive RD&D program for LWR fuels with enhanced accident tolerance. The test data collected as part of the ATF program collected at both ATR and Halden test reactors will support demonstration of Lead Fuel Rods or Lead Fuel Assemblies in a commercial LWR by the end of FY 2022.

### Accomplishments:

INL established a Statement of Work (SOW) in FY 2016 to design test rigs and perform safety analysis of ATF concepts under BWR conditions. The SOW was included in an existing INL/IFE subcontract revised to serve as a bi-lateral agreement between INL and IFE for the HBWR experiments.

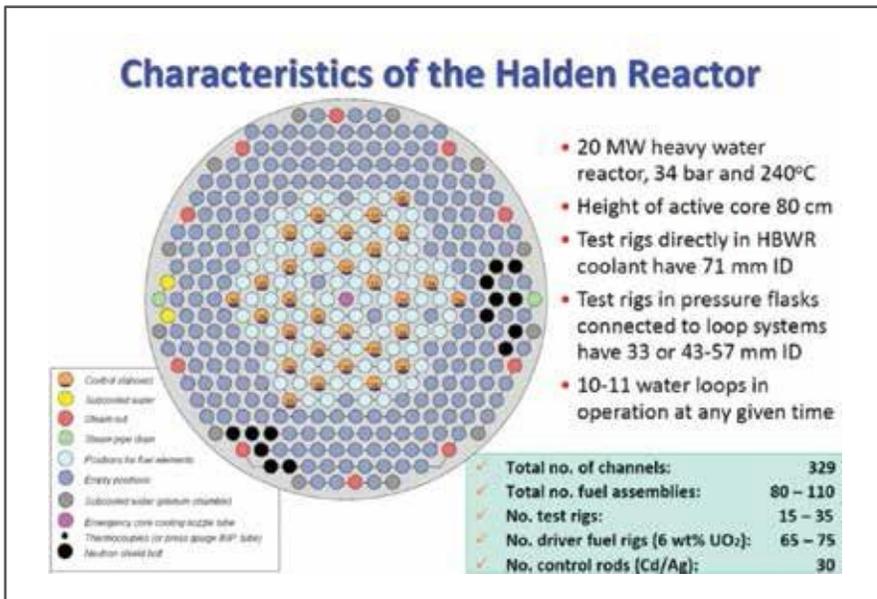


Figure 2. Cross Section and Characteristics of the Halden Boiling Water Reactor (provided by HRP)

***Irradiation testing of ATF concepts in the HWBR will augment the test results in the ATR water loop in support of commercial demonstrations by 2022 and future NRC licensing of Accident Tolerant Fuels.***

The subcontract also includes IFE support for design and fabrication of sensors to be used during testing in out-of-pile autoclave and ATR water loop testing of ATF experiments under PWR prototypic conditions.

Preliminary scoping analysis was performed by IFE in FY 2106 to determine if the HWBR is suitable for performing ATF experiments at desired power level and fuel burnup requested by the Industry Partners. The scoping analysis report identifies different booster fuel enrichments and loading positions in the HBWR core needed to reach desired power

levels and burnup for the preliminary ATF concepts proposed. The results indicate that the burnup targets require an average linear heat rate of approximately 21-23 kW/m over a 6 year duration (depending on the fuel type irradiated). The scoping analysis calculations indicate that this can be reached in several reactor positions in the HWBR. Figure 1 illustrates a typical test rig in the HWBR while Figure 2 illustrates the test positions in the HWBR and characteristics of the reactor.

## Halden ATF Test Planning and Fabrication

Principal Investigator: Kurt Terrani  
 Collaborators: Dr. Yukinori Yamamoto

Segment	CEA <sup>1</sup>	KAERI <sup>2</sup>	IFE <sup>3</sup>	ORNL	EPRI <sup>4</sup>	Reference
	Rod 1	Rod 2	Rod 3	Rod 4	Rod 5	Rod 6
Top	~50µm Cr	~50µm Cr alloy (CrAl)	CrN	FeCrAl	Mo-LCAC FeCrAl	Zry-4
Top-mid	~15µm Cr	ODS ~50µm Cr alloy (CrAl)	FeCrAl		Mo-LCAC Zry	Zry-4
Bottom-mid	~5µm Cr	~100µm Cr Cr/FeCrAl	CrN		Mo-ODS FeCrAl	Zry-4
Bottom	~15µm Cr	ODS + ~100µm Cr Cr/FeCrAl	FeCrAl		Mo-ODS Zry	Zry-4

Table 1. Test matrix of fuel segment/rod with ATF cladding in IFA-796 experiment. All designations except FeCrAl and Mo, indicated coatings on Zr-based alloys. The segments designated for AFC supplied FeCrAl cladding are shaded in green. 1CEA: Commissariat à l'énergie atomique et aux énergies alternatives, French Alternative Energies and Atomic Energy Commission

<sup>2</sup>KAERI: Korea Atomic Energy Research Institute

<sup>3</sup>IFE: Institutt for energiteknikk (manager of OECD-HRP), Norway

<sup>4</sup>EPRI: Electric Power Research Institute, Palo Alto, CA

FeCrAl alloys are the most advanced accident tolerant fuel (ATF) cladding candidates to replace Zr-based alloys in commercial light water reactors (LWRs). These alloys exhibit exceptional high-temperature steam oxidation resistance, comparable or better corrosion behavior to Zr-alloys under normal operating conditions, and at reduced thickness are able to preserve the cycle length of a typical LWR plant. Although many separate effects tests including neutron irradiation of the alloys and dry capsule irradiation with fuel have taken place to date, integral tests under prototypical conditions are essential for further elevation of technology readiness level for this class of cladding materials. Therefore, the focus of this activity is to conduct integral fuel testing using uranium pellets inside FeCrAl cladding tubes inside a PWR loop in a test reactor.

### Project Description:

The focus of this activity is to conduct an integral fuel irradiation experiment in the OECD Halden reactor using ATF cladding. Specifically, this test designated under IFA-796 ID, is a PWR test loop with a 6 fuel rod configuration with the rods placed symmetrically around a circular array. Table 1 describes the test matrix of ATF cladding in IFA-796. Note that 5 of the 6 rods consist of four segments. These segments are 16 cm in length and are meant to expand the breadth of testing within the limited space in the test matrix. Rod 4, 60 cm in length, is the only rod that is fixed and has instrumentation attached to it. The instrumentation on Rod 4 will consist of at least a cladding elongation (EC) sensor to provide information regarding the integral creep and swelling behavior of the cladding as well as to pinpoint the onset of pellet-clad mechanical interaction in this system. All the designations

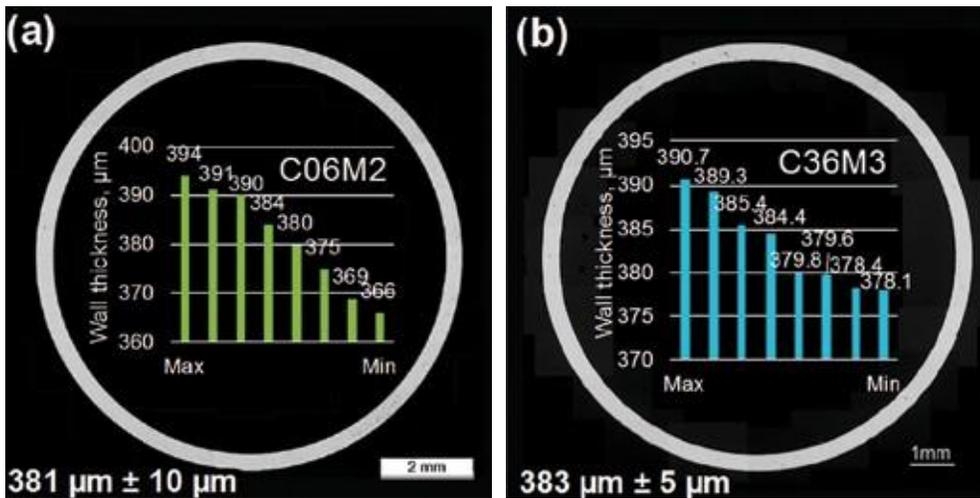


Figure. Optical micrograph of the entire cross section for a) C06M2 and b) C36M3 FeCrAl cladding alloys and the corresponding thickness across the cladding periphery.

in Table 1 refer to coatings on Zr-based alloys except FeCrAl segments in Rod 3 and Rod 4, and coated Mo-based cladding in Rod 5. Rod 6 consists of standard Zircaloy-4 cladding as reference material. The experiment was initially proposed by AFC and the FeCrAl segments were provided by ORNL under the AFC program.

**Accomplishments:**

ORNL has been developing a variety of nuclear grade FeCrAl cladding materials over the past few years under the auspices of the Advanced Fuels Campaign. These same alloys have been undergoing irradiation testing at a number of test reactors (High Flux Isotope Reactor, Advanced Test Reactor, and Halden Reactor) in the form of tensile bars, gun-drilled tubes, and flat coupons. These alloys are all of the wrought variant (instead of some of their commercial counterparts that are produced via the powder metallurgy

route, e.g. APMT), and have reduced Cr content (<13 wt% Cr). The reduction in the Cr content, from >20 wt% Cr in many of the commercial alloys, was intentional since it alleviates embrittlement issues due to Cr-rich  $\alpha'$  precipitate formation under irradiation conditions pertinent to LWR cores. ORNL, Los Alamos National Laboratory, and General Electric company (GE) have all focused some of their activities over the past two years on production of seamless tubing from nuclear-grade wrought feedstock material or commercial bulk alloys. This work has been conducted in collaboration with a number of commercial outlets, namely Century Tubing Inc., Rhenium Alloys Inc., Superior Tube, and Sandvik. Three successful seamless tube production trials were carried out at Century Tubing Inc. (San Diego, CA) and a portion of the resulting thin-walled tubing was sent to the Halden reactor for inclusion in IFA-796 tests.

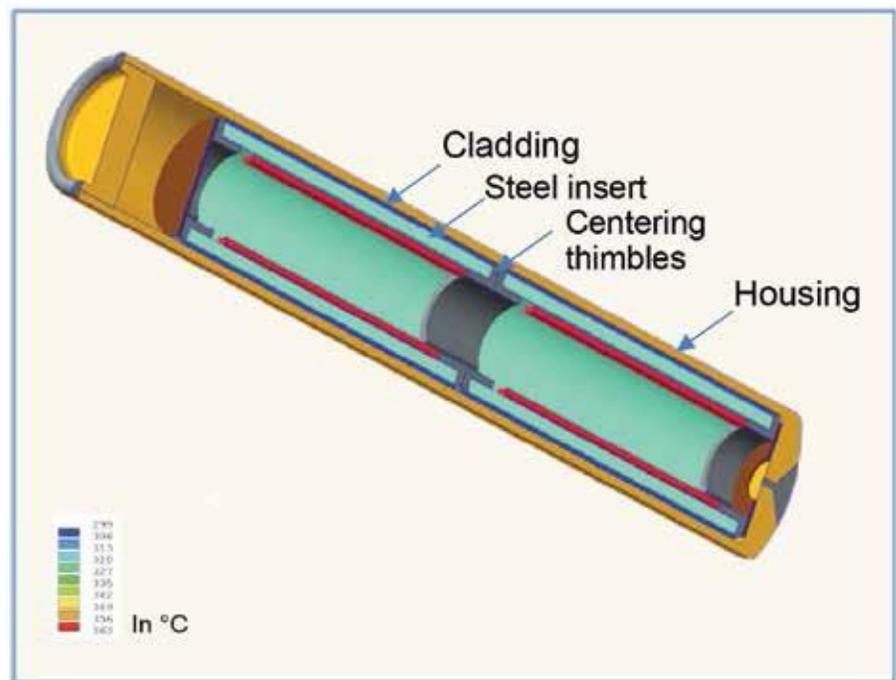
*This experiment is the first integral fuel test with any ATF cladding under prototypical PWR conditions.*

## Irradiation and PIE of ATF Concepts in HFIR

Principal Investigator: Kurt A. Terrani

Collaborators: Kevin G. Field and Christian M. Petrie

Figure 1. 3D model and corresponding thermal analysis of the flexible irradiation capsule for candidate ATF cladding to be irradiated in the HFIR at ORNL.



FeCrAl alloys are currently under study as an accident tolerant fuel (ATF) cladding in commercial light water reactors (LWRs). In this application, thinned walled (~375  $\mu\text{m}$ ) tubing is the primary component and these cladding tubes can experience neutron irradiation up to a few tens of dpa at temperatures in the range of 275-350°C. Neutron irradiation can lead to degradation of properties including radiation-hardening and embrittlement. Production of thinned walled tube can induce

greatly different microstructures compared to sheet product as well as the application leads to non-uniaxial loading. Hence, a need exists to determine the performance of FeCrAl cladding tubes within these unique environmental conditions.

### Project Description:

The primary focus of this project was to develop a flexible test bed for conducting irradiation and subsequent post-irradiation examination (PIE) of candidate ATF cladding tubes in the High Flux Isotope Reactor (HFIR)

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at Oak Ridge National Laboratory (ORNL). The test configuration needed to be flexible enough to handle several different dimensional variations of cladding tubes while still providing irradiation and temperature conditions that are prototypical to LWR radiation environments. Establishing this test bed enables rapid radiation testing of candidate ATF cladding, hence providing critical data on the performance of these cladding materials in-reactor. Furthermore, use of cladding tubes versus typical sheet product provides a better simulation of the final microstructures and stress-states expected for commercial LWR cladding. These assessments will aid in the commercialization of ATF cladding and hence meets the DOE objectives of safe and reliable nuclear power production within the United States.

**Accomplishments:**

The primary goal was to allow for irradiation of shortened cladding tubes in the HFIR while minimizing radial and axial temperature gradients within the rig. This goal is difficult to achieve, as the HFIR exhibits an axial neutron flux gradient that can contribute to significant temperature variations if not managed accordingly. This goal was achieved by developing a 3D model with a specialized configuration for centering the cladding tubes within

the irradiation capsule while placing passive SiC thermometry in direct contact with the cladding tubes. The attached Figure shows the irradiation capsule design and a cut-away model of the cladding showing the thermal gradients expected under irradiation in the HFIR. As seen in the Figure, axial gradients were on the order of 25-50°C. Plans are currently underway to deploy the proposed irradiation capsule configuration including the irradiation of Generation I and Generation II FeCrAl cladding tubes produced via a commercial manufacturer. Additionally, supporting projects have investigated advanced techniques for mechanically testing the specimen's post-irradiation, thereby providing the highest-quality data available on the radiation performance of FeCrAl cladding tubes after irradiation to prototypical LWR radiation environments.

This research developed an irradiation capsule design to directly test the radiation performance of ATF cladding tubes within the HFIR at ORNL.

*This research developed an irradiation capsule design to directly test the radiation performance of ATF cladding tubes within the HFIR at ORNL.*

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## 2.6 TRANSIENT TESTING

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### Transient Response of LWRs Containing ATF Designs

*Principal Investigator: Nicholas Brown*

*Collaborators: Aaron J. Wysocki, Maolong Liu, Kurt A. Terrani, Daniel M. Wachs*

Advanced cladding materials with potentially enhanced accident tolerance will yield different light-water reactor performance and safety characteristics than the present zirconium-based cladding alloys. These differences are due to cladding material properties, reactor physics, and thermal hydraulics characteristics. Differences in reactor physics are driven by the fundamental properties (e.g., neutron absorption cross section in iron for an iron-based cladding) and also by design modifications necessitated by the candidate cladding materials (e.g., a larger fuel pellet to compensate for parasitic absorption). The objective of this research in FY16 was to explore the potential differences due to reactor physics and thermal hydraulic characteristics of candidate ATF cladding materials.

#### **Project Description:**

In FY16, the project effort focused on the potential impact of candidate accident tolerant cladding materials on both the low temperature and high temperature phase of a super prompt reactivity initiated accident. Pellet cladding mechanical interaction (PCMI) is the responsible mechanism for failure during the low temperature

phase and a boiling crisis (departure from nucleate boiling in a PWR or dryout in a BWR) is responsible for failure in the high temperature phase.

The project utilized reactor core kinetics simulation capabilities to estimate differences in the ATF concept pulse response characteristics during hypothetical RIA events. The results of this study apply to future in-pile transient testing (e.g. TREAT testing) as well as to out-of-pile separate effects tests. Essentially these results will be used to inform future experimental studies with appropriate test conditions to study mechanical response of the cladding.

For the PCMI phase of RIA, the applicable conditions were determined using three-dimensional nodal kinetics simulations of an RIA in a representative PWR with both FeCrAl and SiC/SiC cladding materials. The study yields pulse shape boundary conditions for use in future mechanical tests of candidate materials to simulate the RIA event, specifically peak unconstrained fuel thermal expansion and unconstrained fuel thermal expansion rate during the power pulse following the rod ejection. Other example information of interest includes the pulse width and the energy deposition in the fuel.

#### **Accomplishments:**

Two major studies were conducted in FY16. The first study was focused on identifying differences in super prompt RIA response based on reactor physics characteristics. Three-dimensional nodal core models were developed to account for thermal feedback. The core models are in a three-batch equilibrium cycle with approximately 5% enrichment for all but one case with FeCrAl cladding.

As shown in Figure 1, cladding material and fuel design make a difference for RIA response, including core response and fuel rod response. This study identified distinct differences in the behavior of the cases with FeCrAl cladding from the Zircaloy and SiC/SiC cases. These simulations help to draw out distinctions between responses of different fuel and cladding designs.

The RIA simulations indicate that the predicted RIA energy deposition is similar for the Zircaloy, FeCrAl, and SiC/SiC cladding options. The total thermal expansion of the fuel is similar because it is a function of the energy deposition. However, the predicted maximum fuel diameter change rate is higher and the pulse is greater in magnitude with reduced full width at half maximum for FeCrAl

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cladding. These differences result from the greater fuel-to-moderator ratio in the FeCrAl configurations considered in this paper, as well parasitic absorption in iron and chromium. This hardened spectrum drives a shorter neutron generation time, yielding impacts on pulse width and timing, which will impact the strain rate that the cladding would experience in a PCMI event.

The second study conducted in FY16 focused on scoping parametric analyses of the impact of changes in critical heat flux (CHF). Surface conditions are important factors for CHF, primarily the wettability that is characterized by contact angle. Smaller contact angle indicates greater wettability, which increases the CHF. Surface roughness also impacts wettability. Results in the literature for pool boiling experiments indicate changes in CHF by up to 60% for several ATF cladding candidates. The study included parametric evaluation of perturbations of CHF for RIA and Anticipated Transients without SCRAM (ATWS).

Although several parametric evaluations were performed, the findings presented here are from a series of

RELAP5-3D simulations performed as a sensitivity analysis. The nucleate boiling heat transfer coefficient, critical heat flux, transition boiling heat transfer coefficient, and film boiling heat transfer coefficient were perturbed for both hot zero power and hot full power RIA transients in PWRs and BWRs. For hot zero power perturbations, the sensitivity was greatest to the critical heat flux and transition boiling heat transfer coefficient. Figure 2 shows an example set of cases that indicate the sensitivity of a hot zero power RIA in a BWR to a multiplier on CHF and also on the transition boiling heat transfer coefficient. Generally, the lower the CHF, the longer the duration of the dryout conditions for this example.

***This project directly supports the resumption of transient testing in the United States and the restart of the TREAT reactor by providing information about the impact of candidate advanced cladding materials on transient progression.***

## ATF-3 Transient Testing of Accident Tolerant Fuels

Principal Investigator: Rob O'Brien

Collaborators: N. E. Woolstenhulme, A. A. Beasley and D. M. Wachs

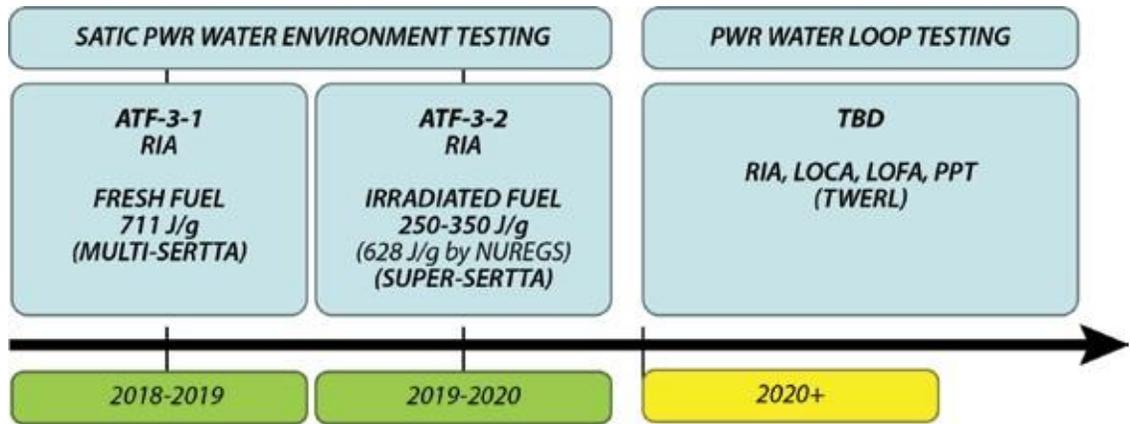


Figure 1. The ATF 3 campaign series, their objectives, and transient test environments.

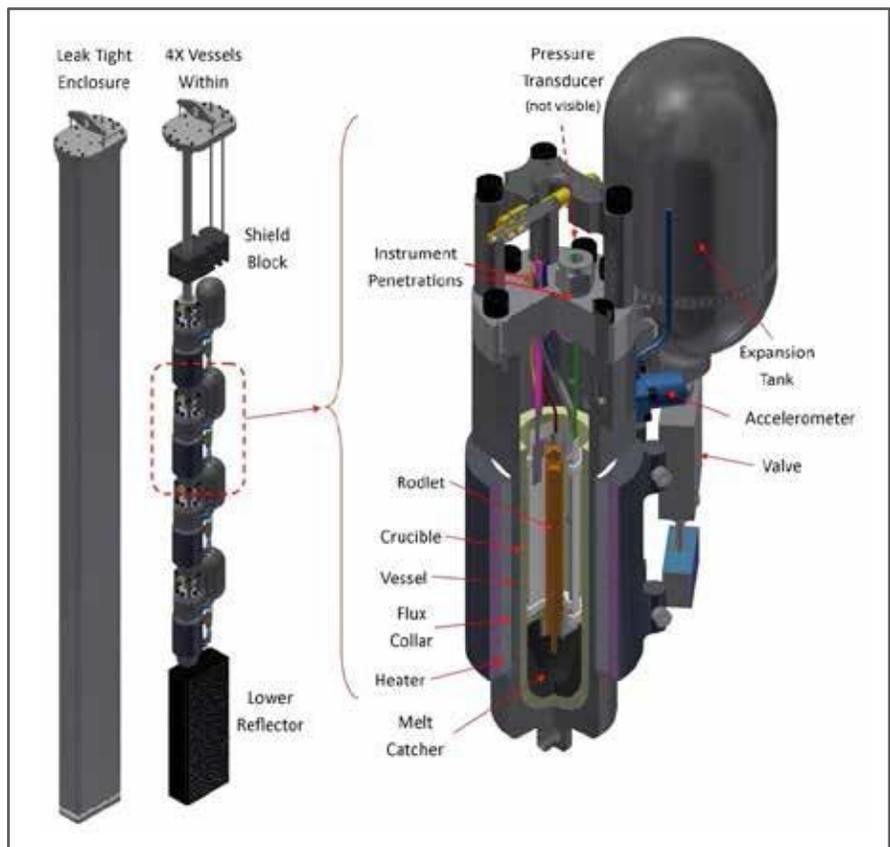
Accidents (RIA) and Loss of Coolant Accidents (LOCA). Overall, these fuel concepts aim to improve their reaction kinetics with steam in order to reduce oxidation of claddings and hydrogen production, minimize fuel Pellet to Cladding Mechanical Interactions (PCMI) and Fuel to Cladding Chemical Interactions (FCCI), and improve fission product retention under these credible accident conditions. Following the ATF-1 and ATF 2 campaigns, the ATF program will perform transient testing of fuel concepts in the Transient Reactor Test (TREAT) reactor at the Idaho national laboratory under the ATF 3 campaign. The ATF 3 campaign will primarily focus on the determination of concept fuel performance during exposure to short duration critical heat transients such as those experienced in RIA events under prototypical environmental conditions for Pressurized Water Reactors (PWR).

### Project Description:

The purpose of ATF 3 is to evaluate the performance of fuel concepts under transient nuclear heating and accident environments. Specifically, ATF 3 will initially perform prototypical RIA transient testing in the ATF-3-1 and ATF-3-2 series in the TREAT reactor using a static environment irradiation vehicle, but other advanced accident environmental testing may be evaluated and may be executed such as LOCA and testing under flowing water conditions. Both fresh and irradiated concept fuel specimens from the ATF 1 and ATF 2 campaigns will be tested under the ATF 3 campaign. The total energy deposited (TED) within candidate fuel specimens will be varied as a function of their level of burnup due to prior irradiation as required by the U.S. Nuclear Regulatory Commission NUREG codes. In order to perform the testing under ATF 3, several self-contained multi purpose test vehicles are currently being developed for TREAT to accommodate the fuel

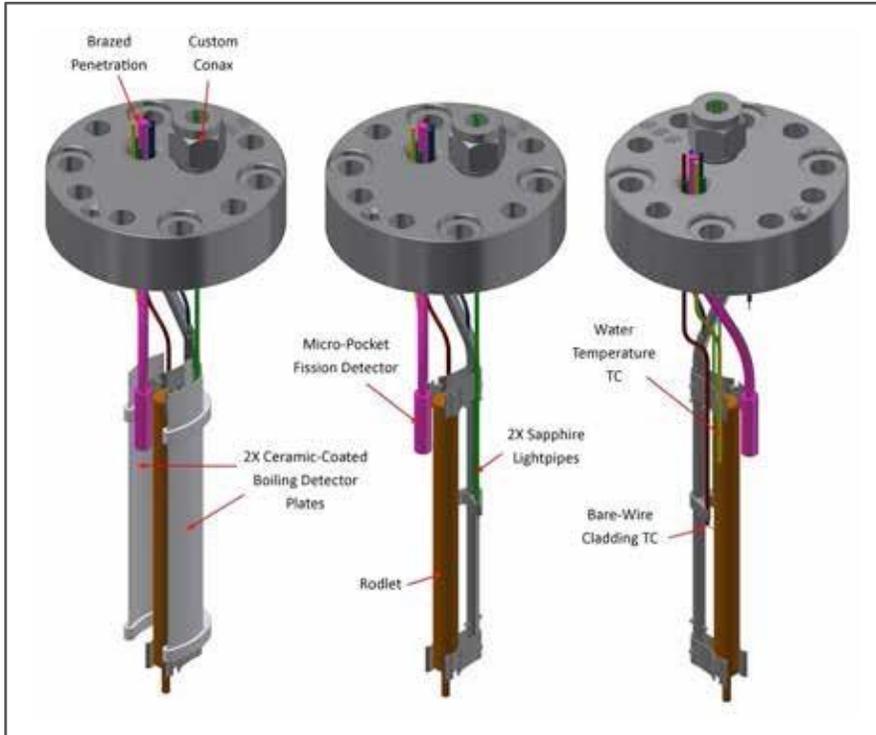
specimens in static wet (PWR H<sub>2</sub>O) or steam environments. The ATF 3 campaign will be divided into 2 initial series with the specific test objectives of testing both fresh and irradiated fuel specimens under transients that are characteristic of RIAs. These initial series will focus on providing a PWR Hot Zero Power (HZP) water environment. Each series will be initiated by a number of calibration transient tests performed in instrumented calibration test vehicles that will provide neutronically similar characteristics of to be tested with ATF fuel concept specimens. These calibration transients will be executed to verify and validate the transient design and performance of the test vehicle prior to the use of prototype fuel rodlets. Of primary importance is to ensure that the correct energy deposition within the fuel specimen will be achieved within any given configuration of the test vehicle and accident condition to be emulated. The use of instrumentation such as thermocouples and fission wires will be essential in the performance of calibration testing.

A conservative approach to testing the fuel concept rodlets will be executed in order to maximize the science return with a limited number of irradiated specimens, while pushing each specimen towards the failure thresholds for traditional, state of the art LWR fuels. A comparison of the ATF concept fuels will be made with reference to current state of the art zirconium clad UO<sub>2</sub> fuels. Characteristic of this approach, calibration rodlets



of each concept will be subjected to a series of calibration transients to verify the computed Power Coupling Factors (PCF) used in experiment design and to ensure good accuracy in delivering the prescribed energy during tests with prototype specimens. Data from in situ instrumentation and radiochemistry analyses will be used to verify the computed Power Coupling Factor (PCF) and to determine Transient Correction Factors (TCF) where necessary. This verification procedure will be implemented in intervals

*Figure 2. The Multi-SERTTA Vehicle to be used to support the ATF-3-1 RIA series of tests on fresh fuel ATF specimens.*



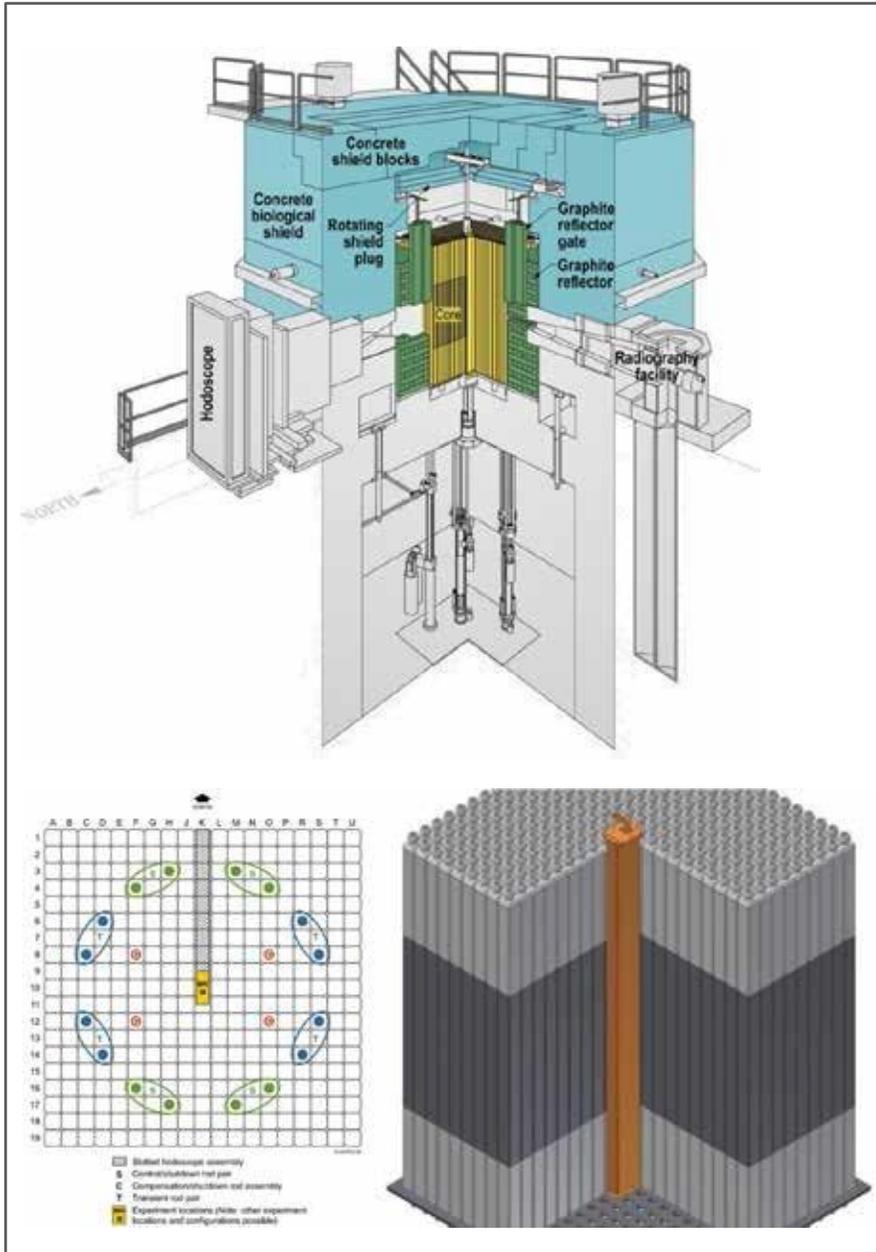
*Figure 3. The Multi-SERTTA internal instrument array that will be used to provide in-pile data to support test execution and post-test analysis.*

between progressively higher energy depositions during calibration tests. For fresh fuel specimens, typical RIA TEDs will be made at 711 J/g. Typical RIA TEDs for samples irradiated at 50-70 GWd/mt will be initiated in the range of 200 J/g to 628 J/g with the potential to continue beyond these energies to investigate thresholds for complete release of fission products from irradiated specimens. to illustrate the importance of the approach to RIA TED magnitude specification. All testing under the ATF-3-1 and ATF-3-2 series will be performed in water at prototypical PWR pressures of 15.5 MPa provided by the Multi SERTTA test vehicle.

### Accomplishments:

The ATF 3-1 and ATF-3-2 test plans and matrices are currently being developed to accommodate each concept fuel specimen. The draft test plan was completed in June of Fiscal Year (FY) 2016. This test matrix will continue to be evaluated and refined during FY-2017 and will be subject to down selections made by the FCRD program and its FOA partners following completion of the ATF 1 and ATF 2 irradiation cycles and subsequent PIE. The aggressive schedule that is currently being developed will ensure that ATF-3-1-CAL experiments will be available and ready to install into the TREAT reactor upon its resumption of operation.

In order to ensure that the testing schedule can be achieved, the detailed design for the Multi-SERTTA static environment vehicle was completed in FY-2016. The design underwent the INL's design review process which was completed in September 2016. This design review included all aspects of nuclear/neutronics, thermal, mechanical and reactor safety analysis. This work will support the FY-2017 activities in experiment design and safety documentation required for experiment insertion into TREAT, and prototype vehicle development. Similarly, the detailed Multi-SERTTA design will feed the FY-2017 work in the development and production of a neutronically similar calibration vehicle to support the initial ATF-3-1-CAL calibration experiments in TREAT.



*The ATF-3 Transient Test Series will provide ATF concept developers with essential initial transient performance data required for a concept fuel to be licensed by the Nuclear Regulatory Commission by demonstrating that such fuels provide adequate resistance to fuel and fission product release under RIA and will allow a plant to maintain a coolable geometry of the core as required by the nuclear regulatory guides.*

Figure 4. (Top) Overview of Treat Features, (Bottom Left) Example Core Map, and (Bottom Right) Multi-SERTTA Vehicle in TREAT Core in 1/4 Section View.

## Design and Testing of a Modified Burst Test System for Evaluation of ATF Claddings

Principal Investigator: Nicholas Brown

Collaborators: M. Nedim Cinbiz, Kurt A. Terrani, Rick Lowden, Don Erdman, Daniel M. Wachs

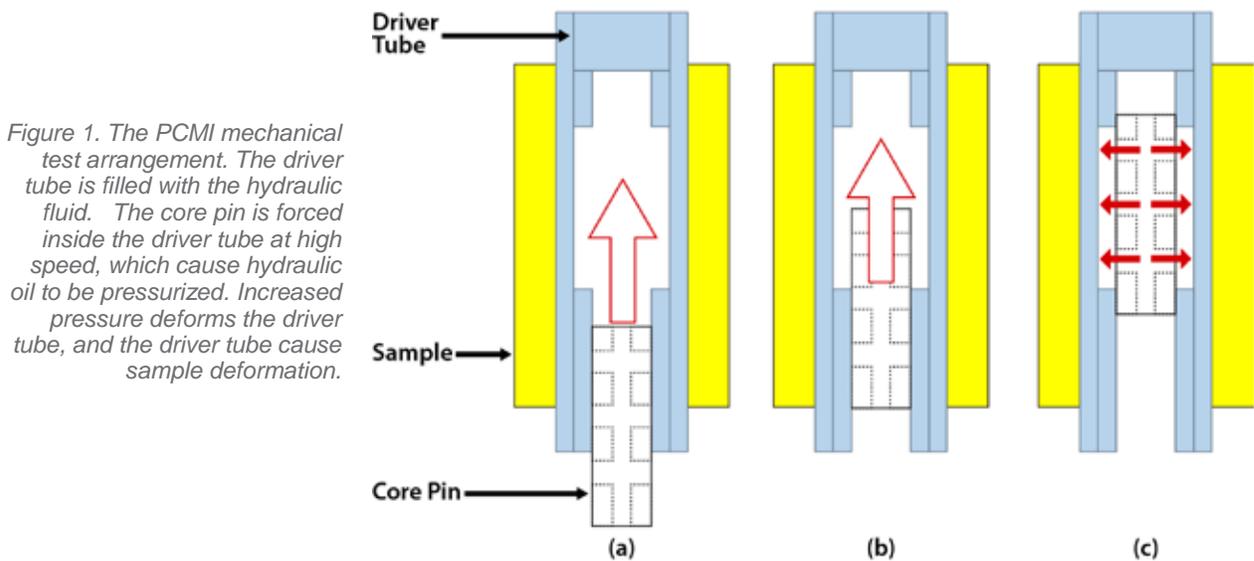


Figure 1. The PCMI mechanical test arrangement. The driver tube is filled with the hydraulic fluid. The core pin is forced inside the driver tube at high speed, which cause hydraulic oil to be pressurized. Increased pressure deforms the driver tube, and the driver tube cause sample deformation.

The reactivity-initiated accident (RIA) is a postulated design basis accident in light water reactors. A potential failure mechanism for medium or high burnup fuel in RIA is pellet cladding mechanical interaction (PCMI). During FY16 initial mechanical separate effects tests of a zirconium-based alloy and FeCrAl alloy cladding failure were conducted at a high strain rate, which supports understanding of how these materials may behave in an RIA. These tests also support development of failure criteria for candidate ATF cladding materials.

A mechanical test rig was developed to provide well-controlled cladding strain and strain rate representative of that experienced in the PCMI phase of a super prompt RIA. In addition, FY16 included studies to enhance the design of the driver tube, which induces the strain for the cladding tube samples, and also to provide boundary conditions that are relevant for ATF cladding candidates.

### Project Description:

A variety of RIA-initiating events are possible, but typically the RIA takes the form of a control-rod-ejection accident in a pressurized water reactor (PWR) or a control-rod-drop accident

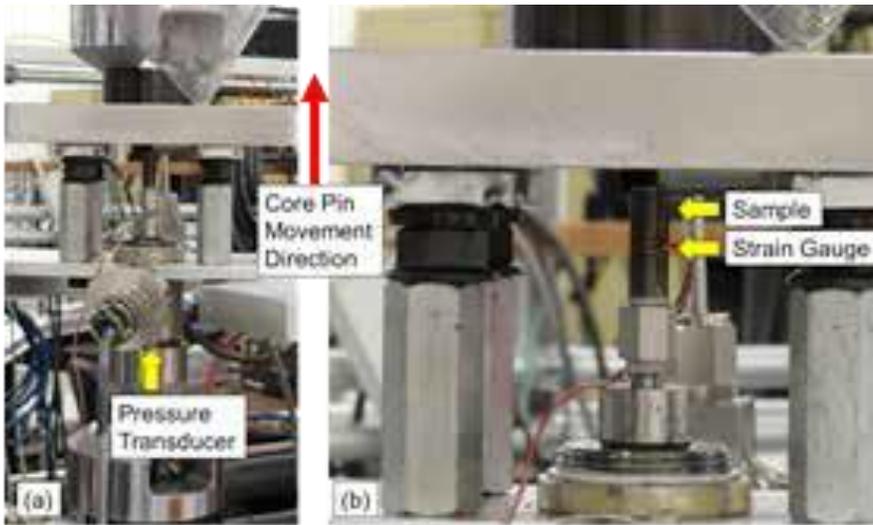


Figure 2. Components of the Mechanical frame (a) and close-up view of the mechanical frame with sample and strain gauge (b).

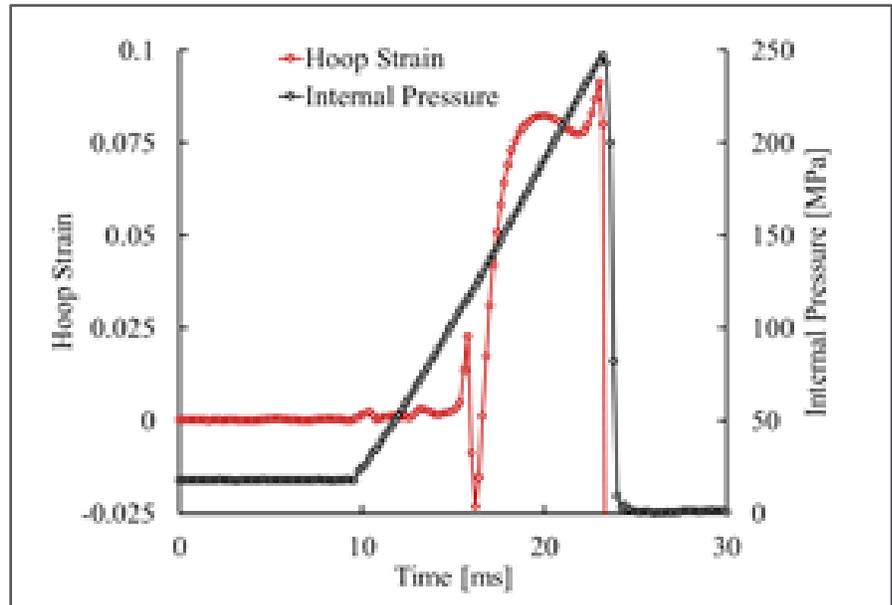
in a boiling water reactor (BWR). RIA events cause an exponential increase in the fission rate density until fuel temperature feedback (Doppler effect) stops the chain reactions. This postulated event occurs in a very short time, but prompt energy deposition in the fuel can cause mechanical loading of cladding at high strain rate due to thermal expansion of fuel under near adiabatic conditions. This in turn may induce PCMI.

The design criteria for RIA are based on the coolant pressure boundary's integrity and on maintaining the core cooling capability<sup>1</sup> as specified by the

US Code of Federal Regulations (10 CFR Part 50). Thus, the cladding integrity under PCMI must be maintained during the RIA transient. This criterion is also valid for the candidate ATF cladding during RIA type of loading conditions. To assess the mechanical behavior of the ATF cladding, both integral and separate effects tests of the candidate alloys should be considered under RIA or RIA-like conditions.

This initial study established a working test rig and investigated PCMI during postulated RIA or RIA-like transients for unirradiated model

Figure 3. Strain evolution on the ZIRLO cladding with 168 wt. ppm of hydrogen up to rupture. The rupture occurred immediately when the internal oil pressure increased up to 250 MPa in 15 ms.



FeCrAl alloy tubes by conducting separate effect tests. To perform mechanical testing for the PCMI phase of RIA-like transients, a mechanical testing rig was developed and used that is capable of working at high strain rates under precise control of the strain. The device is based on an improved version of a modified burst test facility design developed by EPRI 2. Since there is no data for FeCrAl tube mechanical behavior at high strain rates, a zirconium-based alloy that is pre-hydrated has been used, in addition to two different types of FeCrAl alloys, to provide a reference. Scoping experiments are conducted

to determine possible mechanical responses of FeCrAl alloys to the high strain rate PCMI between cladding and nuclear fuel for comparison with industry standard zirconium-based alloys. The results are discussed in terms of the changes in the hoop and diametral strain change.

The schematics of the PCMI driver tube and sample are shown in Figure 1. The core-pin is encapsulated by the age-hardened Inconel driver tube, which in turn is jacketed by the cladding sample. The mechanical system's working principle is based on pressurizing viscous hydraulic oil by the axial movement of the core pin within an age-hardened Inconel 718 tube

at high speed. Pressurization of the hydraulic oil induces radial expansion of the Inconel tube, and the tube's expansion applies a force onto the test sample, which causes it to radially expand, analogous to UO<sub>2</sub> fuel expansion during RIA. Thus, rupture or permanent deformation of the cladding materials occurs if the driver tube expansion induces sufficient strain at the contact area between the sample and the driver tube.

The key components of the mechanical testing system are shown in Figure 2. As the core pin is moved upwards by the load cell, the pressure transducer records the filled hydraulic oil pressure, and the strain gauge records the hoop strain on the sample as a result of the driver tube expansion.

#### **Accomplishments:**

The rapid expansion of the nuclear fuel in an RIA was simulated by driver tube expansion for several samples at room temperature. In FY16, the samples included both zirconium-based specimens (ZIRLO) with hydride formation and FeCrAl alloys B126Y and B136Y. A typical hoop strain test curve profile for a ruptured zirconium-based specimen is shown in Figure 3. For this test, the pressure inside the driver tube rose up to 250

MPa within 15 ms. The hoop strain signal showed a time lag of 5 – 7.5 ms. A fluctuation in the strain response was observed which may have been caused by the strain time lag and the pressure wave initially induced by the fast core-pin movement. This behavior was not observed for other tests. When hoop strain increased up to 0.091, rupture of the sample occurred.

The diametral strain for B126Y obtained after the mechanical test using laser surface profilometry is shown in Figure 4 (a). The maximum diametral strain is analogous to a ductility measurement for tube expansion. Figure 4 (b) compares the diametral strains of different FeCrAl alloys and the ZIRLO cladding along the specimen height. Diametral strains were 0.2, 0.13, and 0.08 for ZIRLO, B126Y, and B136Y. The ZIRLO showed higher diametral strain compared to FeCrAl alloys. As expected, B136Y, which was in the as cold drawn condition without any final annealing, exhibited the lowest diametral strain. Because grain sizes and the composi-

*These separate effects tests on the failure of candidate ATF cladding materials are necessary to determine appropriate safety limits and failure mechanisms, as well as validate high fidelity advanced modeling and simulation tools, such as BISON.*

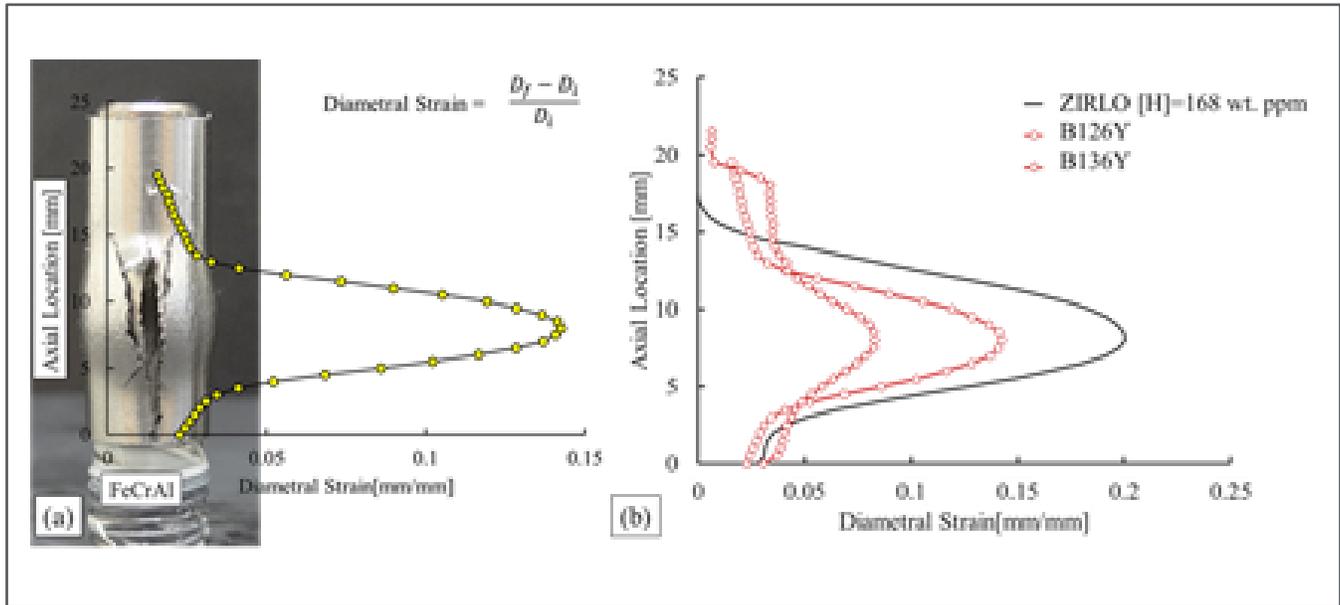


Figure 4. Permanent (plastic) diametral strain distribution along the FeCrAl specimen height after rupture (a), and comparison of the diametral strains for different FeCrAl alloys with ZIRLO 168 wt. ppm after rupture (b)

tions of both FeCrAl alloys were more or less same, the ductility difference between FeCrAl alloys was caused by post heat treatment after the cold drawing process. B126Y underwent a final annealing, relieving the residual stresses, reducing the dislocation density, and increasing the ductility compared to the as-drawn alloy. These numbers are comparable with the uniaxial tensile strength of similar alloys developed at ORNL.

Future work includes implementation of digital image correlation for the strain measurement and development of new driver tube designs for ATF cladding candidates, which have different expected thicknesses and

inner diameters than zirconium-based cladding designs. In addition, improvements to the baseline driver tube design are expected to better reproduce expected strain biaxiality and stress for zirconium-based alloys. Future tests will be conducted using a variety of FeCrAl and SiC/SiC samples. The tests will account for expected differences in strain rate due to the reactor performance and safety characteristics of the candidate materials. High temperature tests and eventual tests of irradiated materials in a hot cell are also envisioned.



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# ADVANCED REACTOR FUEL SYSTEMS

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- 3.1 AR Fuels Development
- 3.2 AR Computational Analysis
- 3.3 AR Core Materials
- 3.4 AR Irradiation Testing & PIE Techniques
- 3.5 Capability Development

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## 3.1 AR FUELS DEVELOPMENT

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### Feedstock NpO<sub>2</sub> Reduction

Principal Investigator: Leah Squires

*The elimination of long lived actinides in spent nuclear fuel would be a great advance in nuclear energy and neptunium metal is needed for the development of transmutation fuels.*



Figure 1. Metal retrieved from oxide reduction run.

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In order to study the feasibility of transmutation fuels it is necessary to fabricate fuels that mimic spent reactor fuel. Pure transuranic (TRU) materials such as neptunium, that are present in spent fuel, are necessary additives to this type of experimental fuel. Currently the bulk of the neptunium that exists within the DOE complex is in the form of neptunium oxide and therefore a process has been developed to reduce neptunium oxide to neptunium metal.

**Project Description:**

The reduction of neptunium oxide to neptunium metal is accomplished using calcium metal as a reducing agent and calcium chloride. First the neptunium oxide starting material is calcined at 1200 degrees Celsius under argon atmosphere. Following the calcining step the material is mixed with an excess of

calcium metal and calcium chloride in a magnesium oxide crucible and heated to a molten state and stirred. The calcium metal pulls the oxygen from the neptunium leaving neptunium metal and calcium oxide which is absorbed into the calcium chloride salt. As the mixture cools the neptunium metal falls to the bottom of the crucible and can be easily identified and retrieved once the materials resolidify.

**Accomplishments:**

The process was previously developed and demonstrated and was published this FY.

## AmBB Experiment Preparation and Planning

Principal Investigator: Leah Squires



Figure 1. Metal retrieved from the last distillation run using the lathe.

*The elimination of long lived actinides in spent nuclear fuel would be a great advance in nuclear energy.*

The americium bearing blanket (AmBB) project is an effort by France to eliminate long lived actinide materials in spent nuclear fuel by re-irradiating the material in the fuel blanket. In support of this effort DOE is funding INL to produce pure americium metal which will be used as an additive to simulated spent fuel for testing of the concept.

### Project Description:

The AmBB project aims to convert americium, a long lived actinide present in spent nuclear fuel, into shorter lived products; thereby, decreasing the time it takes for spent nuclear fuel to decrease in radioactivity. This is important for the advancement of nuclear energy because spent fuel waste is one of the largest obstacles nuclear energy faces in the future. In order to test the method it is necessary to fabricate fuel which mimics spent nuclear fuel containing americium and therefore a reliable source of

pure americium metal is needed. This project employs the specially designed americium distillation furnace in the Fuel Manufacturing Facility at Idaho National Laboratory to make pure americium metal in support of France's efforts to test the feasibility of reducing or eliminating the americium in spent fuel.

### Accomplishments:

Americium distillation runs were performed to determine the optimal conditions for the most efficient production of pure americium metal. These experiments were conducted using a material that is a mixture of americium and neptunium. Due to the relatively low vapor pressure of americium compared to that of neptunium the two metals can be separated once molten by heating the mixture to a temperature at which americium will volatilize while the neptunium will not. This is performed under vacuum to lower the temperature to which the mixture needed to be heated. The process is carried out in a tantalum crucible that is placed inside a



*Figure 2. Shavings of americium metal retrieved from the distillation*

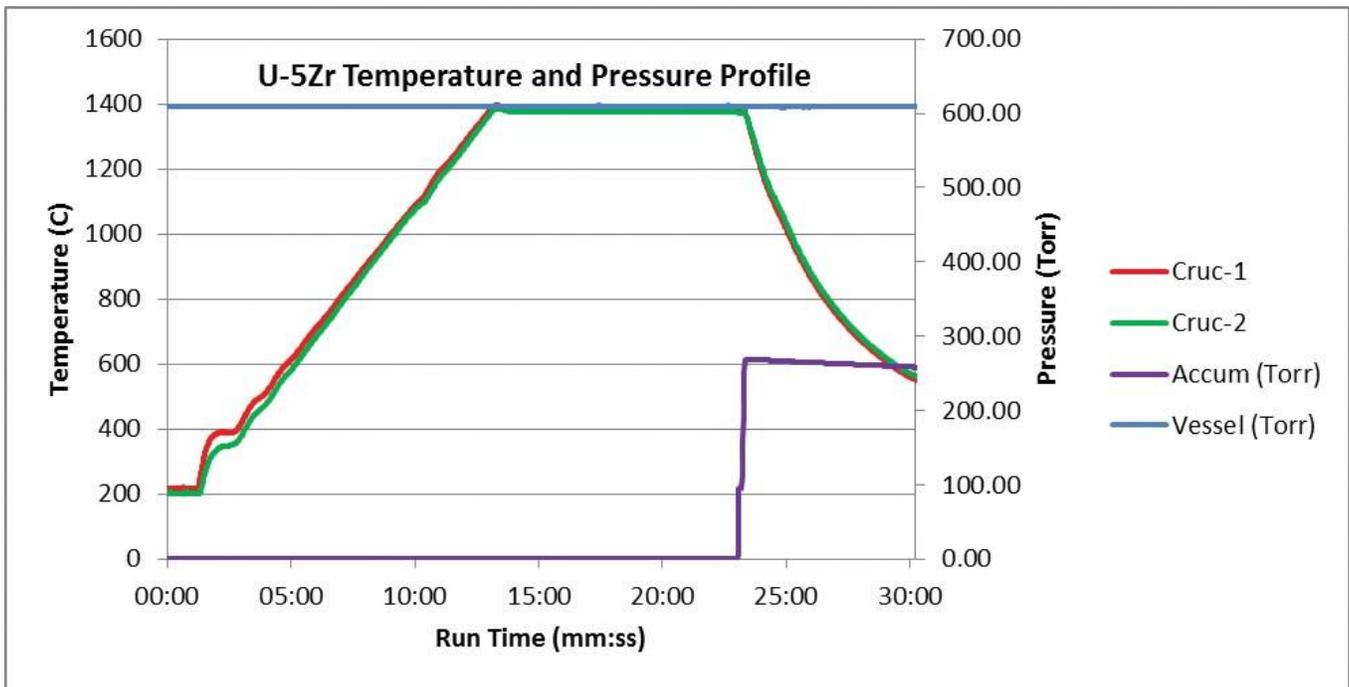
heating mantle style furnace. The volatilized material is collected by re-condensing it on a copper cold finger placed at a sufficient height above the starting material to allow for complete separation. In practice the material tends to re-condense on the walls of the crucible just below the cold finger position. In order to locate and retrieve the material a gamma detector is used to run along the length of the crucible and find the americium signature. Once the americium is located the crucible is cut and the inside is reamed out. A

major advancement in this retrieval process was made this year by the addition of a small hobby lathe to the glovebox. This tool allows for more precise drilling and also protects personnel from radiation exposure by decreasing the time necessary for material retrieval and eliminating the need for personnel to hold the crucible during the process.

## Fluidity Studies on Uranium and Uranium-Zirconium Alloys

Principal Investigator: Randall Fielding

Collaborators: Scott Wilde, Jake Green, Ginger Dexter, Kevin Hays



fluidity test  
Figure 1. Typical heating and pressure profile from a

The AFC program fabricates fuel through arc casting for irradiation tests. It has been seen that how the molten material flowed during the casting process was dependent upon the composition, as well as other factors. During the Experimental Breeder Reactor (EBR-II) fuel fabrication campaigns the experience of the operator was heavily relied upon, especially for experimental alloys, for casting success. These fluidity studies are an initial attempt to begin to quantify alloy flow as a function of composition.

### Project Description:

Fluidity is a general term used to describe how well a molten material flows and doesn't account for individual parameters such as viscosity, reactivity, surface tension, etc., but rather an overall flow property. In traditional foundry operations a fluidity test is conducted on an alloy specific basis where the molten material is poured into a spiral mold to produce a spiral component with a round cross section. The total length of the flow is measured and is called fluidity. Due to a number of factors such as pouring temperature, mold size and shape, alloy composition,

	DU_1	DU_2	DU_3	U-5Zr_1	U-5Zr_2	U-5Zr_3	U-10Zr_1	U-10Zr_2	U-10Zr_3
Length (mm)	244	259	239	181	175	184	189	168	176
Average (mm)	247.3			180.0			177.7		
Std Dev	±10.4			±4.6			±10.6		

etc. there is not a standard design or test procedure therefore the tests are qualitative and are specific to the foundry. In a laboratory setting a similar test can be run using a glass tube as a mold with either gravity or a pressure differential as the flow driving force. These tests can be more repeatable, but are still only semi-quantitative because an individual property is not measured, only the total length of flow.

Based on the laboratory flow tests testing was done on uranium and uranium-zirconium alloys. The tests were designed to determine the length of flow for a given alloy at a specific superheat for a given pressure differential. Because these tests will be done using the instrumented furnace the pouring temperature, total volumes, pressure differentials, and mold dimensions are well characterized. Based on these known parameters and the flow lengths a casting simulation can be developed that can be used to better benchmark the casting models, possibly

be used to determine the relative importance of the material properties that are incorporated into the flow term, and begin to establish a measurement database of how alloying compositions affect the flow of material.

**Accomplishments:**

Three flow tests were done for depleted uranium (DU), depleted uranium-5wt% zirconium (U-Zr5), and depleted uranium-10wt% zirconium (U-10Zr) for a total of nine tests. The furnace was configured to allow a quartz tube to be inserted through the top of the furnace. One end of the tube is connected to a “vacuum accumulator” which was evacuated to <100 mTorr. For each test approximately 200 grams of the starting materials were charged into a yttria coated graphite crucible and heated to 100°C above the reported liquidus shown in the phase diagram. The material was then held for approximately 10 minutes. The U-10Zr tests

*Table 1. Summary of fluidity rod lengths*



Figure 2. Typical cast rods resulting from the fluidity tests a) DU, b) DU-5Zr, and c) DU-10Zr.

were held for slightly less time due to the furnace induction coil temperature limitations. After holding 9.5 minutes a 4 mm inside diameter quartz tube was inserted into the melt. After 30 seconds the quartz was opened to the vacuum accumulator and material sucked into the quartz mold. Figure 1 shows a typical thermal and pressure

profile for a U-5Zr run, however, the general trends seen are typical for all of the compositions. After the casting cooled it was removed from the furnace and the total length of material lifted into the tube was measured.

Figures 2 show typical examples of the cast rods. Lengths were measured from the top of the ingot to the top of the rod. Table 1 shows the lengths of each

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*Based on the results of these tests the actual flow of material can start to be quantified and related to composition and rely less on experience.*

rod along with the average length and standard deviation for each composition. As seen in Table 1 there is a clear difference in flow properties between the pure uranium and the alloyed uranium. Although little difference is seen between the 5% and 10% alloyed materials. This may be partially due to the flows in the rod leading to biased measurements. In Figure 2 a large void is seen near the top of the U-10Zr pin. When the rod was measured this void area was included in the measurement, possibly skewing the lengths to longer than is actual. It is also possible that it

takes only a small amount of alloying to dramatically affect the flow properties and therefore the effects are less noticeable as more alloying material is added. Further testing and characterization is needed in order to draw more solid conclusions. Characterization of these results will continue into fiscal year 2017, as will additional testing including other alloying additions.

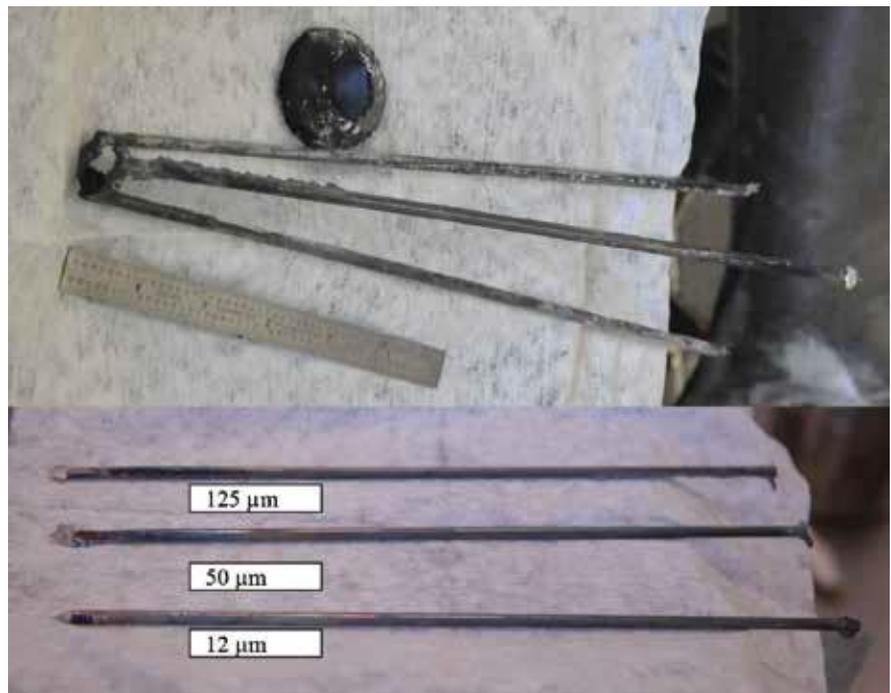
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## Metallic Fuel Casting Trials with an Integral Zirconium Sheath

Principal Investigator: Randall Fielding

Collaborators: Laura Sudberth, Michael Benson, Scott Wilde, Kevin Hays, Ginger Dexter

Figure 1. Above) Cast pins from the mold Below) Separated cast pins showing sheath thicknesses.



Despite the robustness of metal fuels, they are susceptible to fuel cladding chemical interaction (FCCI). FCCI is caused by fission product rare earth elements interacting with the iron based cladding. This reaction effectively reduces the cladding thickness which is a major limiting factor for high burnup metal fuels. If this FCCI could be mitigated possible burnup limits could also be increased. A method of mitigating FCCI in metallic fuels is to incorporate an integral FCCI barrier material, such as zirconium, between the fuel and cladding. The initial feasibility of integral fuel FCCI barrier was

shown earlier during the Integral Fast Reactor program however, during post irradiation examination it was seen that as the fuel swelled the sheath was split and the fuel extruded out. In areas where the fuel was still contained in the sheath there was no evidence of FCCI. A possible strategy to accommodate fuel swelling is to incorporate a sheath that is composed of a foil which is wrapped around the fuel with an overlapping section that corresponds to the amount the fuel will swell. As the fuel swells it the foil will unwrap, however, because of the overlap the zirconium will always be between the fuel and the cladding.

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### Project Description:

An experiment was developed to cast fuel into an integral Zr barrier using the glovebox advanced casting system (GACS). This system is designed to cast three slugs approximately 4.2 mm diameter by 250 mm in length. A charge of U-6Zr was cast successfully into the rolled Zr sheaths. Three different zirconium foil thicknesses were used for the experiment, 125  $\mu$  m, 50  $\mu$  m, and 12  $\mu$  m in order to determine the lower limit of thickness that could be used. The foils were loaded into the conical two-piece GACS mold. The mold was heated to approximately 700°C at the top and upper portion while the lower portion was heated to approximately 415°C. The uranium and zirconium were placed in a yttria coated graphite crucible positioned above the mold. The crucible was heated to 1500°C and held for approximately 13 minutes. After the hold time was accomplished the stopper rod was lifted from the crucible allowing it to drain into the mold. After casting each fuel slug was sectioned for metallographic examination in order to determine the amount of melt/sheath interaction. The second set of experiments was conducted using the arc casting method. The arc casting equipment is designed to cast items of a smaller batch size, consequently the size of

slug mold was approximately 5 mm x 75 mm, which is more prototypic of fuel irradiation test specimens. For these experiments a charge of U-10Zr was prepared through standard arc melting techniques. The foil was loaded into a quartz mold 5 mm in diameter by 89 mm in length. The foil thickness was 25  $\mu$  m. The resulting cast product was removed from the quartz mold, and section for both radial and longitudinal examination using an optical microscope.

Two separate methods were used to produce and load the foil sheaths into the quartz molds. The first technique simply hand rolled the foil prior to loading into the quartz, after loading an attempt was made to smooth the foil using a cotton swab. Despite attempts to smooth the foil, due to the nature of the thin foil a completely smooth sheath could not be produced. The second experiment rolled the foil tightly around an undersized steel mandrel, which was then inserted into the quartz tube. The mandrel was then removed and the foil allowed to spring back against the quartz tube. In this experiment the foil was left 2-3 mm long on the top edge and was then folded down around the outside of the quartz mold. After casting this fuel sample was also sectioned radially and longitudinally for optical microscopy.

*Experiments have shown that casting into an integral FCCI barrier made of rolled zirconium foil is **defi** **feasible***



*Figure 2. Upper portion of the second attempt arc cast fuel slug.*

**Accomplishments:**

Figure 1 shows the three resulting U-6Zr rods from the GACS furnace casting experiment. A visual inspection of all three slugs showed them to be generally sound with a few local flaws or discontinuities. As seen in Figure 1 in some areas the zirconium foil appears to have degraded during the casting process. In some areas the sheath can be clearly seen around the fuel, in other areas a sheath is not discernable; however, metallographic examination clearly showed the zirconium foil in the fuel interior. Both the 125  $\mu$  m and the 50  $\mu$  m sheathed fuel were very similar. Although both pins had areas where the sheath had been moved into the interior of the fuel, particularly at the overlap regions, both pins were completely surrounded by the sheath. However, it was common for fuel material to infiltrate the area of

overlap. In the 125  $\mu$  m pin it appeared that there was more interaction at the bottom section, while the 50  $\mu$  m pin showed less interaction at the bottom. Also it appeared that on the thicker sheath materials the outside edge is better intact when compared with the 12  $\mu$  m sheath material.

In the arc casting experiment the first attempt simply hand rolled the 25  $\mu$  m zirconium foil and inserted into the 5 mm quartz mold. The resulting sheath was quite wrinkled however the casting was still attempted. The slug was approximately 70 mm in length but, there were some gaps along the length. During the de-molding operation the bottom most portion of the pin broke off revealing another gap. As with the GACS pins the sheath position varied along the length based on the loading of the sheath into the mold. At the bottom section of the



mold because the sheath was able to fit more tightly against the quartz mold, fuel alloy did not run between the outside of the sheath and the mold. However, even in the bottom section fuel alloy can be seen in between the overlapped portion of the sheath. Based on these results the process of loading the zirconium sheath was changed to provide a more uniform sheath and overlap joint. The second  $25 \mu\text{m}$  thick sheath was rolled tightly around a steel mandrel and inserted into the quartz mold and allowed to expand against the glass and a small portion of the zirconium foil was folded down over the top edge of the mold. This method of sheath handling provided a much better and more consistent sheath. Casting was done using standard arc casting techniques. The overall pin was made up of several solid sections separated by partially fitted sections. Samples were removed from the upper and lower solid sections. Sections of the fuel were examined using optical microscopy. Figure 2 shows an overall cross section and a high magnification image of the upper portion of the

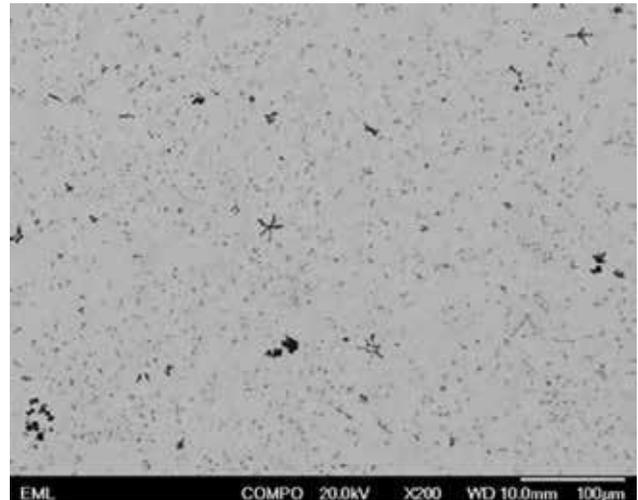
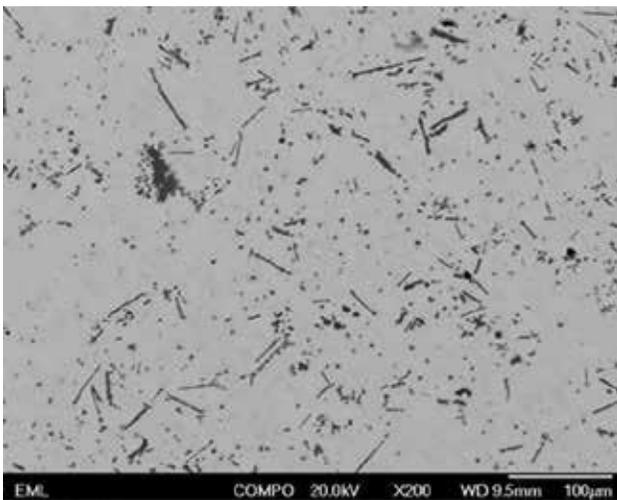
cast pin. Notice on the overall cross section some overlap fit is still seen, although the amount of overlap fit is greatly reduced. It should be noted that the amount of overlap is more than would be required for an irradiation test. Figure 3 shows a macrograph of a longitudinal cross section micrograph. In Figure 3 a consistent cast product is seen over the length of several millimeters. Also visible is the overlapped portion of the sheath which is clearly free, presumably to cover the irradiation induced swollen fuel in an irradiation test.

From these experiments it has been shown that casting into an integral FCCI barrier made of rolled zirconium foil is definitely feasible. Although, feasibility has been shown, additional optimization will be needed especially in the area of sheath handling and loading processes. In both casting techniques the major flaws in the sheath location were caused by sheath placement.

*Figure 3. Longitudinal macrograph of the lower portion of the second arc cast slug.*

## Fabrication of the AFC-3F Metallic Fuel Fabrication Variables Experiment

Principal Investigator: Randall Fielding  
Collaborator: Blair Grover



Arc Cast (Right) Injection cast  
Figure 1. Micrographs showing the microstructure of U-20Pu-10Zr (Left)

The AFC-3F is a standard AFC-OA type drop in test designed to examine the effects of fuel casting technique on overall fuel behavior. 5 rodlets containing either U-10wt%Zr or U-20wt%Pu-10wt%Zr were fabricated and inserted in the Advanced Test Reactor.

### Project Description:

EBR-II fuel was cast using an injection casting process. In this process approximately 15-18 kg of fuel was induction melted in an yttria coated graphite crucible under either an inert purge or vacuum, while the quartz molds were suspended above

the melt. At the end of the melting cycle the furnace and molds were evacuated, the molds lowered into the melt, and the furnace pressurized. The sudden pressurization forced the molten material up into the molds. This process was used to cast pins approximately 16 inches long which were then trimmed to the required fuel length. The AFC-OA fuel slugs are generally fabricated through arc casting. A small charge of approximately 10-25 grams of material is placed on a copper hearth where it is melted by means of an electric arc. After melting the material flows into a quartz mold positioned in the copper hearth. Generally lengths

vary from 1.5-3 inches. The fuel microstructure is dependent on which casting technique was used and the associated thermal history with that technique. Figure 1 shows two fuel slugs cast by the two different methods. The AFC-3F test will place fuels of both U-10Zr and U-20Pu-10Zr fabricated with each method in a side by side test.

**Accomplishments:**

A total of five rodlet/capsule assemblies were made. Rodlet 3F-R1 contained a EBR-II cast U-10Zr fuel slug. The EBR-II cast U-20Pu-10Zr fuel was used in rodlet 3F-R5. The U-20Pu-10Zr was retrieved from an archived slug that was cast for the X521 test. The original slug is shown in Figure 2. The U-10Zr fuel was retrieved from a driver element, therefore the fuel had been through the sodium settling and bonding process. Of the other three rodlets two, 3F-R2 and 3F-R4, were loaded with arc cast U-10Zr and 3F-R3 was loaded with an arc cast U-20Pu-10Zr fuel slug. Figure 3 shows the fueled rodlets before encapsulation into the outer capsules. The capsule have been loaded and inserted into the Advanced Test Reactor for irradiation testing.



Figure 2. Archival X521 fuel slug that was used to supply the injection cast U-10Pu-10Zr fuel slug for AFC-3F.



Figure 3. AFC-3F rodlets ready for encapsulation (3F R1-R5).

*The AFC-3F irradiation test examining the effects of fuel casting techniques on fuel behavior has been fabricated using both arc melting fuel and archival EBR-II fuel which has been fabricated through injection casting.*

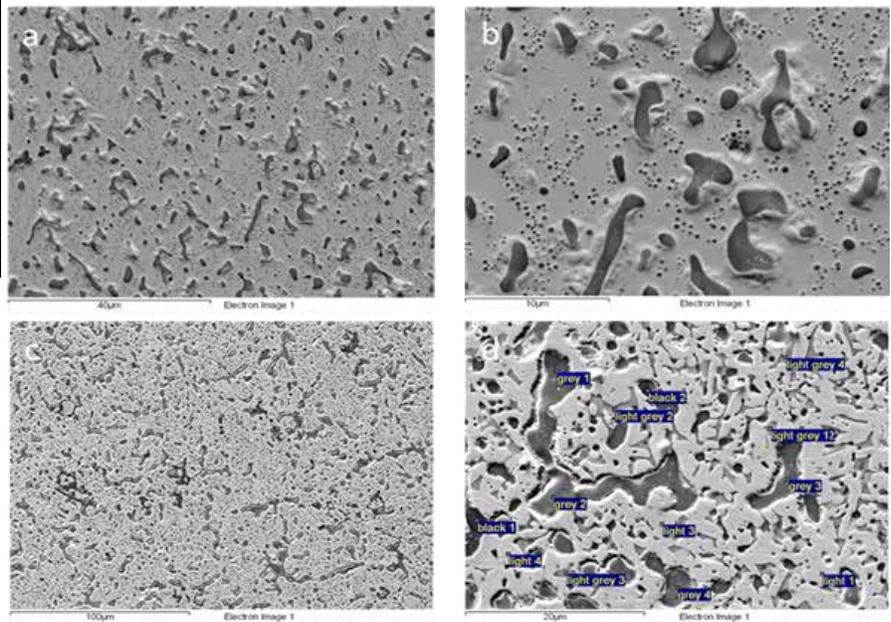
## Alloy Optimization Casting and Characterization Studies

Principal Investigator: Michael Benson

Collaborators: James A. King and Jason Harp

Fuel-cladding chemical interaction due to fission product lanthanides can be prevented using small amounts of Pd.

Figure 1. Microstructure of U-12Zr-4Pd. a and b are as-cast. c and d were annealed at 650°C and quenched in water.



Fuel-cladding chemical interaction (FCCI) occurs when the nuclear fuel or fission products react with the cladding material. A major cause of FCCI in U-Zr and U-Pu-Zr fuels during irradiation is fission product lanthanides (Ln), which tend to migrate to the fuel periphery, coming in contact with the cladding. The result of this interaction is degradation of the cladding, and will eventually lead to rupture of the fuel assembly. Palladium is being investigated as an additive to control FCCI in metallic fuels specifically due to lanthanides. Palladium will prevent FCCI by forming very stable intermetallic compounds with the

lanthanides, thus preventing interaction with the cladding. Studies are underway to characterize the effects of palladium in a metallic fuel.

### Project Description:

The technical objectives of this research are to investigate additives to metallic fuels to improve the performance. Previous work on palladium shows promising results for controlling FCCI. The current work is a continuation of that work to fully characterize palladium as a metallic fuel additive. The specific objectives for this year were to characterize the as-cast and annealed fuel microstructure and perform diffusion couples against iron. This work was carried out using a U-Zr based fuel plus addi-

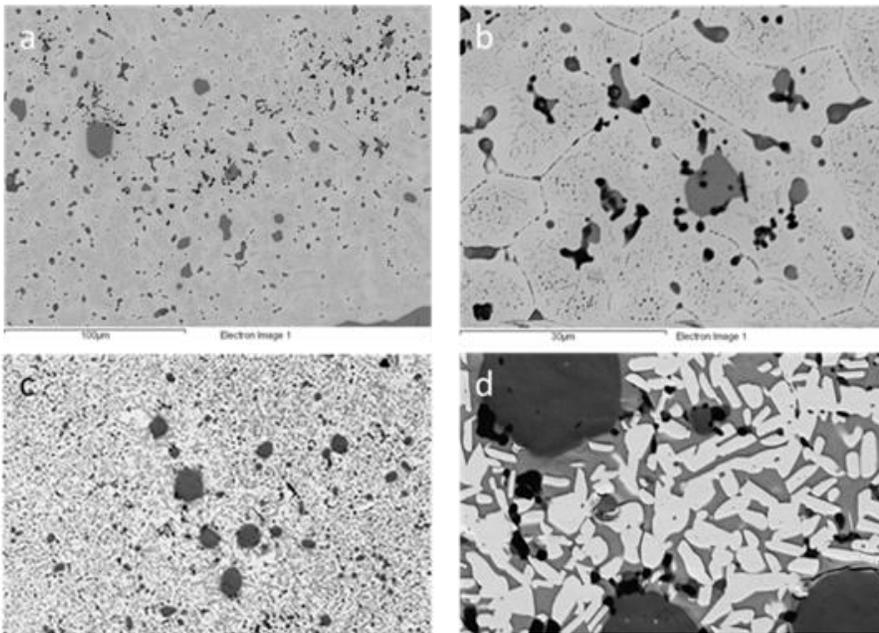


Figure 2. Microstructure of U-12Zr-4Pd. a and b are as-cast. c and d were annealed at 650°C and quenched in water.

tives. In order to investigate additive effects in a Pu based fuel, fuel pins of U-20Pu-10Zr plus additives were fabricated. Characterization and diffusion couples will occur in FY17. An additive that effectively controls FCCI will help the DOE meet its objectives of a safe, reliable, and economic reactor by significantly improving fuel performance. By preventing FCCI due to the fission product lanthanides, cladding ruptures will be prevented, improving fuel safety and reliability, and higher fuel burn-up will be possible, thus improving reactor economics by decreasing the amount of fuel required, and decreasing the amount of nuclear waste generated.

#### Accomplishments:

Three alloys, U-10Zr-4Pd, U-12Zr-4Pd, and U-14Zr-4Pd (always in wt.%, unless noted), have been prepared, and the as-cast alloys have been characterized by scanning electron microscopy (SEM). These alloys are prepared in a step-wise manner, thus the addition order was also explored, and characterized. Two methods of preparation, adding Zr and Pd, followed by U (Figure 1a and 1b), and adding U and Pd, followed by Zr, yielded roughly the same microstructure, whereas adding U and Zr followed by Pd significantly changed

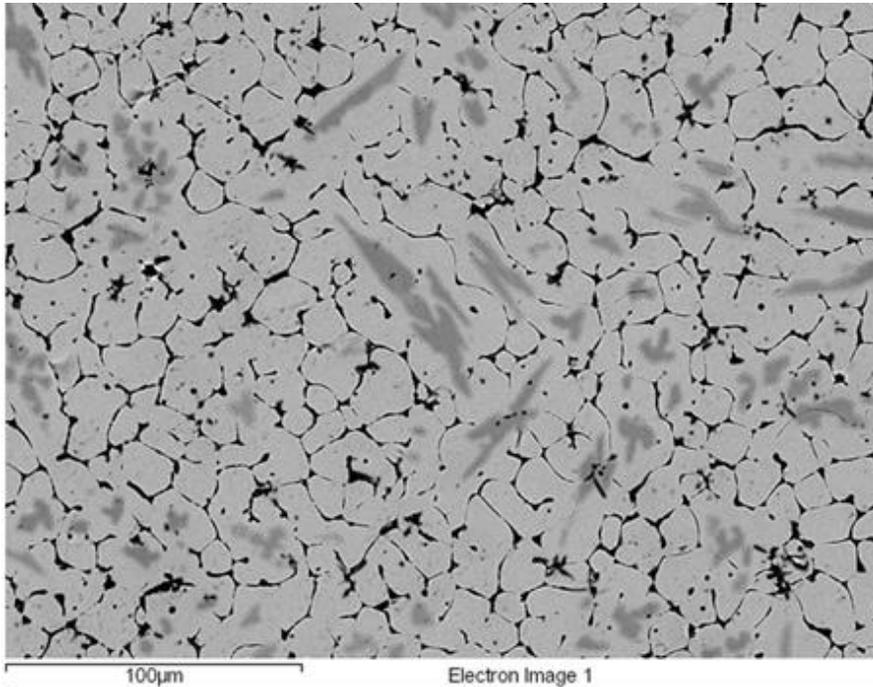


Figure 3. Microstructure of Ln-25Pd.

the microstructure of the alloy. In this alloy, the Pd-Zr precipitates are along the grain boundary. The precipitates are primarily Zr<sub>2</sub>Pd, with a small amount of ZrPd. A small amount of Pd (~1wt%) and Zr (~7wt%) are found in the uranium matrix. After annealing at 650°C for 500 hours (Figures 1c and 1d), the bulk matrix is a combination of  $\alpha$ -uranium and  $\delta$  phase. The  $\alpha$ -uranium phase has less than 0.5wt% Zr and Pd, the  $\delta$  phase is U and Zr, with ~1 wt% Pd, and the precipitates are Zr<sub>2</sub>Pd.

Figure 2 shows images of U-12Zr-4Pd-5Ln, where Ln = <sup>53</sup>Nd-<sup>25</sup>Ce-<sup>16</sup>Pr-<sup>6</sup>La. As-cast images are shown in Figure 2a and 2b, and

annealed are shown in Figure 2c and 2d. In this alloy, U and Zr were mixed first, followed by Pd, followed by lanthanides. The precipitates along the grain boundary are obvious in Figure 2b. The large grey precipitates are the intermetallic LnPd. There is a slight excess of Pd, found in the intergranular boundaries as Zr<sub>2</sub>Pd. There is a small amount of Zr present in the precipitates, although the bulk of Zr is found in the U matrix and in the black precipitates. In the annealed structure (Figure 2c and 2d), the large, dark grey precipitates are LnPd, with less than 0.5wt% Zr. The light grey regions are  $\delta$  phase, and the white is  $\alpha$ -uranium. There is significantly more  $\delta$  phase since Zr is not associated with Pd. This is significant, since it indicates Pd has a definite preference for the lanthanides over the other elements in the fuel.

The primary fission product lanthanides found in metallic fuel are Nd, Ce, Pr, and La. The lanthanides are the primary cause of FCCI, but have never been characterized. Attempts to characterize with X-ray diffraction were not successful, due to the extremely high rate of oxidation of the lanthanides. Due to this, a sample of the lanthanide mix (<sup>53</sup>Nd-<sup>25</sup>Ce-<sup>16</sup>Pr-<sup>6</sup>La) and a sample of the lanthanides with Pd (Ln-25Pd) were sent to North Carolina State University (Ayman I. Hawari and Q. Cai) sealed in vanadium sample cans for neutron diffraction studies. The Ln-Pd ratio was chosen based on recent PIE results of a U-10Zr fuel pin, with

only fission product palladium. The structural data for LnPd and Ln<sub>7</sub>Pd<sub>3</sub> were obtained. These phases can be observed in the SEM image shown in Figure 3. The darker regions are LnPd (~4% of the sample) and the matrix material is Ln<sub>7</sub>Pd<sub>3</sub> (~96%). The dark precipitates in the intergranular region are lanthanide oxides. Although the lanthanides were purchased in mylar bags packed under argon, cerium had some oxidation. Based on the annealed sample shown in Figure 2c and 2d, LnPd will be formed if enough Pd is present, otherwise Ln<sub>7</sub>Pd<sub>3</sub> will be the dominant structure.

Figure 4 shows an SEM EDS map of the interface between U-12Zr-4Pd-5Ln and iron. The diffusion couple was run for 3 weeks at 650°C. There is a Zr rind visible in the Zr map, a common occurrence in U-Zr fuels, although the rind does not extend to the top of the image. At the top of the image there is a LnPd precipitate, in direct contact with iron. The lanthanides have been shown to react vigorously with iron at 650°C, although in the presence of Pd, there is no diffusion into the iron. This is very encouraging for Pd as an additive to control FCCI. A diffusion couple was also run between U-12Zr-4Pd and iron (not shown). There is no interaction between Pd and iron. These results indicate Pd will not contribute to FCCI prior to fission products burning in, and will contain the lanthanides when they do burn-in.

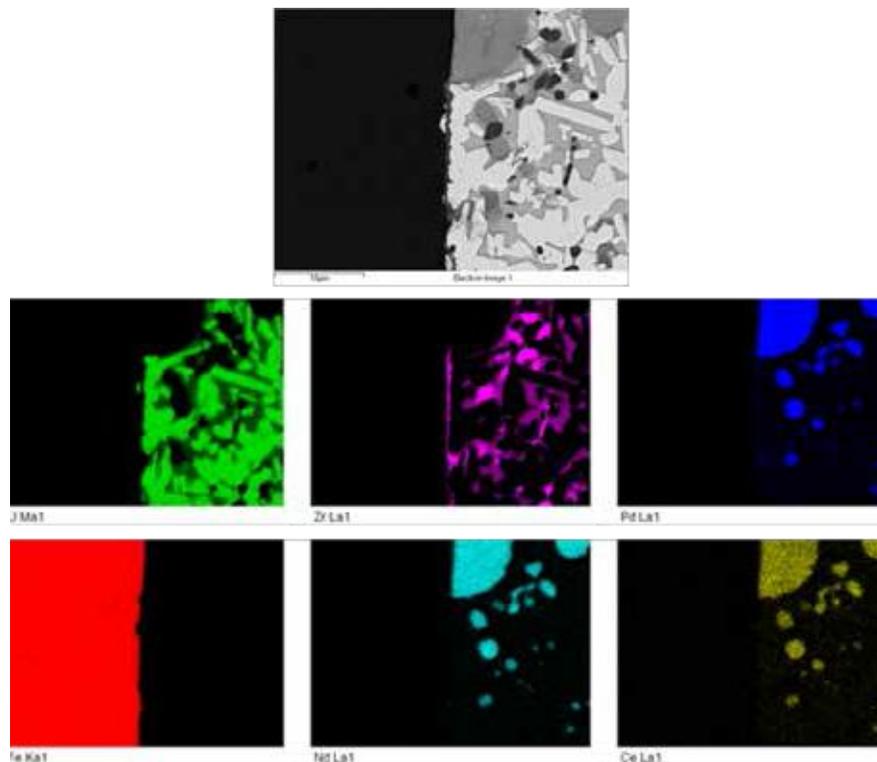


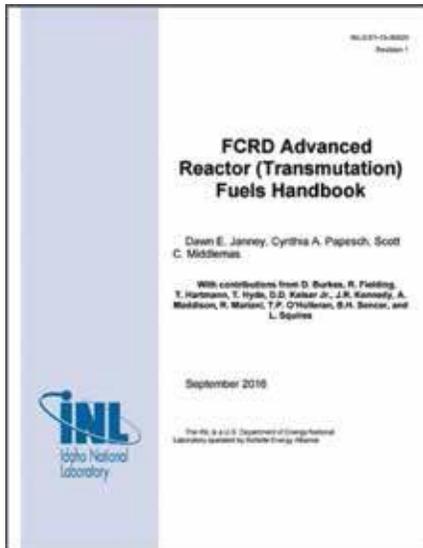
Figure 4. SEM EDS map of interface between U-12Zr-4Pd-5Ln and iron.

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## Update of the FCRD Advanced Reactor (Transmutation) Fuels Handbook

Principal Investigator: Dawn Janney

Collaborators: Cynthia Papesch and Scott Middlemas



Transmutation of minor actinides such as Np, Am, and Cm in spent nuclear fuel is of international interest because of its potential for reducing the long term health and safety hazards caused by the radioactivity of the spent fuel. One important approach to transmutation involves incorporating the minor actinides into U-Pu-Zr alloys, which may also include rare-earth elements (La, Ce, Pr, Nd) if they are used in a closed fuel cycle. It is, therefore, important to understand not only the properties of U-Pu-Zr alloys but also those of U-Pu-Zr alloys that also include minor actinides (Np, Am) and rare-earth elements (La, Ce, Pr, and Nd). However, the existing experimental data is widely scattered, and much of it was published before ~1975. This Handbook summarizes the available experimentally based knowledge of several key properties of U-Pu-Zr alloys with minor actinides and rare-earth fission products.

Transmutation of minor actinides such as Np, Am, and Cm in spent nuclear fuel is important because of its potential for reducing the long term health and safety hazards caused by the radioactivity of the spent fuel. One important approach to transmutation (currently being pursued by the DOE Fuel Cycle Research & Development Advanced Fuels Campaign) involves incorporating the minor actinides into U-Pu-Zr alloys, which can be used as fuel in fast reactors. These fuels are well suited for electrolytic reprocessing, which leads to incorporation of rare-earth fission products such as La, Ce, Pr, and Nd. It is, therefore, important to understand not only the properties of U-Pu-Zr alloys but also those of U-Pu-Zr alloys that include minor actinides (Np, Am) and rare-earth elements (La, Ce, Pr, and Nd) in concentrations relevant for transmutation fuels.

This Handbook contains information about elements, binary, and ternary alloys in the U-Np-Pu-Am-La-Ce-Pr-Nd-Zr system. It summarizes the

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available information about phase diagrams and related information (including phases and phase transformations); heat capacity, entropy, and enthalpy; thermal expansion; and thermal conductivity and diffusivity. In addition to presenting information about materials properties, the handbook attempts to provide information about how well the property is known and how much variation exists between measurements. Although it includes some results from models, its primary focus is experimental data.

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***This handbook provides a single source of information about important materials and properties from widely scattered, often obscure, sources.***

how much variation exists between measurements. Although it includes some results from models, its primary focus is experimental data.

The handbook is intended to serve two audiences. One audience is researchers, who may find it useful to know what has been measured, where it has been published, and what kinds of information are needed. A second audience is modelers, who need a concise summary of information about specific properties, the accuracy with which they are known, and the likely range of variation to be considered in sensitivity analyses.

All contributors to the project are INL employees. They include Dawn Janney (compiler, primary researcher, and first author of the report), Cynthia Papesch (project manager and experimentalist who analyzed and contributed some previously unpublished INL data),

and Scott Middlemas (experimentalist who collected and contributed some previously unpublished INL data). Personnel from the INL Research Library provided invaluable help in finding obscure references, Dr. Pavel Medvedev assisted with Russian-language translation, and Dr. Steve Hayes provided insight and valuable guidance.

It is expected that the FCRD Advanced Reactor (Transmutation) Fuels Handbook will be a multi-year project with new versions available periodically. Each version will update the data in the previous versions, as well as expand the scope of the Handbook. This year's Handbook includes an update to last year's summary of the U-Pu-Zr ternary alloys based on new research and publications. The new scope includes: 1) references and critical reviews of the available data on the phases and phase diagrams of the elements Np, Am, La, Ce, Pr, and Nd; the binary alloys U-Np, U-Am,

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U-La, U-Ce, U-Pr, U-Nd, Np-Am, Np-La, Np-Ce, Np-Pr, Np-Nd, Np-Zr, Pu-Am, Pu-La, Pu-Ce, Pu-Pr, Pu-Nd, Am-La, Am-Ce, Am-Zr, La-Ce, La-Pr, La-Nd, La-Zr, Ce-Pr, Ce-Nd, Ce-Zr, Pr-Nd, Pr-Zr, and Nd-Zr; and the ternary alloys U-Np-Pu, U-Np-Zr, U-Pu-Am, U-Ce-Zr, Np-Pu-Am, Np-Pu-Zr, and Pu-Am-Zr; 2) references and critical reviews of the available data on the heat capacity and related properties (specific heat, incremental enthalpy) for the elements Np, Am, La, Ce, Pr, and Nd; the binary alloys U-La, U-Ce, U-Pr, U-Nd, Pu-Am, La-Ce, La-Pr, La-Nd, Ce-Pr, Ce-Nd, and Pr-Nd; and the ternary alloy U-Ce-Zr; 3) references and critical reviews of the available data on the thermal expansion of the elements Np, Am, La, Ce, Pr, and Nd; the binary alloys U-Np, Np-Pr, Np-Pu, Pu-Am, Pu-Ce, Am-Ce, La-Ce, La-Pr, La-Nd, U-Ce-Zr, and Pr-Nd; and the ternary alloys U-Ce-Zr, Np-Pu-Zr, and Pu-Am-Zr; and

4) references and critical reviews of the available data on the thermal conductivity, thermal diffusivity, Lorenz number, and electrical resistivity of the elements Np, Am, La, Ce, Pr, and Nd; the binary alloys Np-Pu, Pu-Am, Pu-Ce, La-Ce, La-Pr, Ce-Pr, and Pr-Nd, and the ternary alloy Pu-Am-Zr. These alloys are clearly not an exhaustive list, just the only ones for which data of interest for the Handbook has been published.

References considered for the Handbook were published from the late 1930s through 2016. The Handbook has a strong bias in favor of experimental data when available, but supplements the experimental data by results of modeling as needed. It also contains numerous references to other modeling papers.

## Fundamental Property Measurements Supporting NEAMS Validation

Principal Investigator: Krzysztof Gofryk

Collaborators: C. Papesch, Y. Zhang, J. Harp, and J. Lien

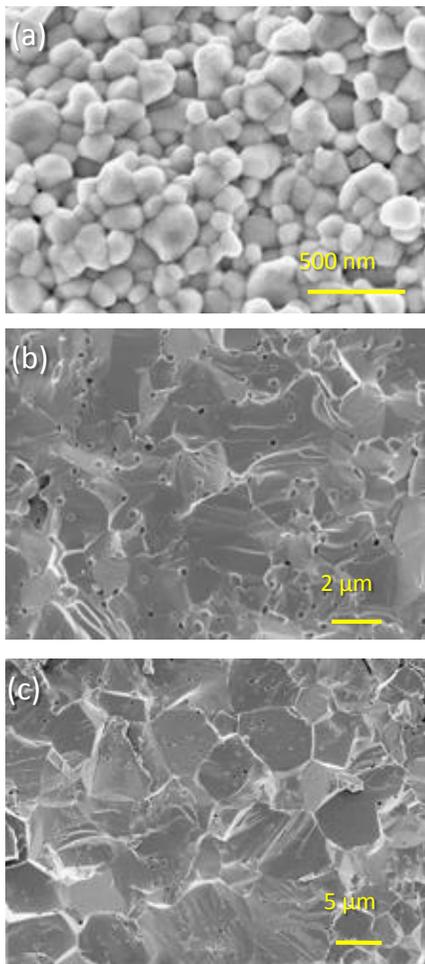


Figure 1. The electron microscope pictures of the UO<sub>2</sub> samples with different grain sizes measured in these studies

Uranium dioxide (UO<sub>2</sub>) is a prime commercial nuclear fuel and uranium silicide (U<sub>3</sub>Si<sub>2</sub>) is considered as a new fuel for the existing LWR fleet. While nuclear fuel operates at high to very high temperatures, thermal conductivity and other materials properties lack sensitivity to temperature variations and to material variations at reactor temperatures, especially related to defect, impurities and grain boundary scattering. These variations need to be characterized as they will afford the highest predictive capability in modeling and offer best assurances for validation and verification at all temperatures.

### Project Description:

Nearly 20% of the world's electricity today is generated by nuclear energy from UO<sub>2</sub> fuel. The thermal conductivity of the fuel governs the conversion of heat produced from fission events into electricity and it is an important parameter in reactor design and safety. Therefore better understanding of scattering mechanisms, especially grain boundary scattering is of paramount interest of nuclear energy research. In order to better understand the impact of grain boundary scattering on the thermal

conductivity of UO<sub>2</sub> and to estimate how much heat is carried by electrons and lattice vibrations in U<sub>3</sub>Si<sub>2</sub>, we have initiated and performed extensive thermal transport measurements of these materials. Uranium silicide, U<sub>3</sub>Si<sub>2</sub>, has been considered as a new fuel for the existing Light Water Reactor fleet. This uranium intermetallic has a number of advantageous thermophysical properties that support its use as an accident tolerant fuel. Because of its high thermal conductivity, U<sub>3</sub>Si<sub>2</sub> can operate at a much lower temperature and experiences lower thermal gradients than UO<sub>2</sub>. The thermal conductivity of the UO<sub>2</sub> and U<sub>3</sub>Si<sub>2</sub> samples have been measured at INL using direct pulse-power "one heater two thermometers" method implemented in Thermal Transport Option (TTO) on the Quantum Design Physical Properties Measurement System (PPMS). The results have been compared to high temperature measurements performed by C. Papesch (INL) and theoretical modeling done by Y. Zhang (INL).

### Accomplishments:

In order to identify which scattering mechanisms are important in nuclear fuels we performed a series of thermal conductivity measurements on selected samples of UO<sub>2</sub> and U<sub>3</sub>Si<sub>2</sub>. The UO<sub>2</sub> samples were synthesized having different grain sizes and the

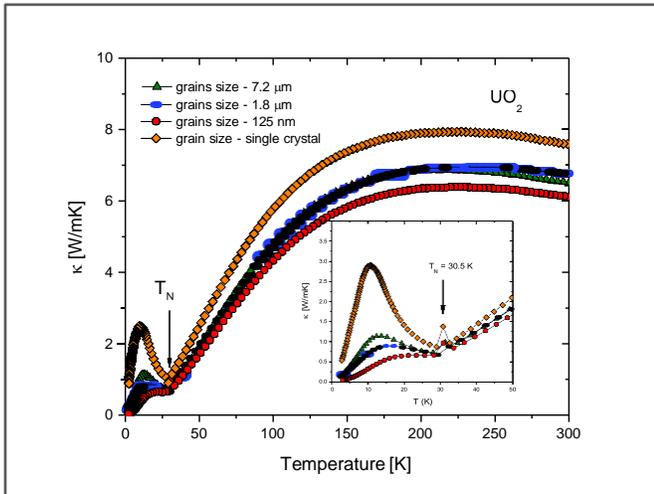


Figure 2. The temperature dependence of the thermal conductivity of RPI samples together with  $\kappa(T)$  of single-crystalline  $\text{UO}_2$ . Inset: low temperature  $\kappa(T)$ . Arrows mark the magnetic phase transition at 30.5 K.

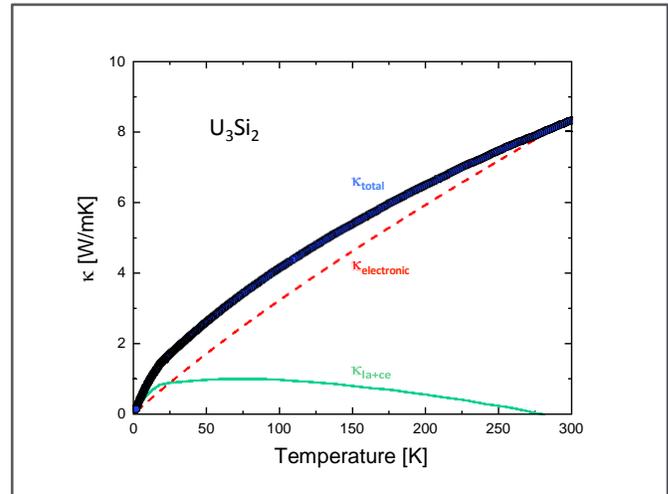


Figure 3. The temperature dependence of the thermal conductivity of  $\text{U}_3\text{Si}_2$ . The dashed and solid lines represent thermal conductivity coming from carriers and lattice vibrations, respectively

measurements were performed below room temperature, where grain boundary scattering should dominate. The samples have been prepared and characterized by Prof. Jie Lian from Rensselaer Polytechnic Institute. Figure 1 shows Scanning Electron Microscope (SEM) pictures of the samples prepared. Our detailed studies indicate impact of grain boundary scattering on thermal conductivity in  $\text{UO}_2$ , especially at low temperatures (see Figure 2) and in agreement with theoretical modeling performed by Dr. Y. Zhang. However, before drawing any firm conclusions on this matter more studies are required involving impact of off-stoichiometry and isotope scattering on the thermal transport in this

system. Also, by performing detailed measurements of the electrical resistivity and thermal conductivity we were able to estimate how much heat is carried by electrons and lattice vibrations in  $\text{U}_3\text{Si}_2$ . The samples for these studies have been prepared and characterized by Dr. Jason Harp (INL). Our preliminary studies indicate that the lattice heat transport is only present at low temperature and is almost entirely governed by electrons above room temperature (see Figure 3). These results are important for thermal conductivity calculations where lattice contributions can be neglected at high temperatures.

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## 3.2 AR COMPUTATIONAL ANALYSIS

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### Fast Reactor Performance -- Am-bearing Blanket Analyses

*Principal Investigator: T.K. Kim*

*Collaborators: N. Stauff and T. Fei*

Heterogeneous Minor Actinide (MA) transmutation in Sodium-cooled Fast Reactor (SFR) based on the Am-bearing blanket (AmBB) concept was proposed by CEA, France, and the irradiation of the AmBB fuels in the Advanced Test Reactor (ATR) is planned. In this study, the transmutation performances of AmBB concept in French and U.S. SFR core designs were evaluated and the irradiation conditions of the AmBB fuels were identified through a detailed pin-by-pin analysis.

#### **Project Description:**

The primary objectives of this study is to evaluate MA transmutation performance based on the AmBB concept and to identify the irradiation conditions of the AmBB fuels to prepare irradiations in the ATR. In this study, the AmBB transmutation performances were compared in terms of French and U.S. SFR core concepts (ASTRID vs. ABR), metal vs. oxide fuels, and homogeneous vs. heterogeneous transmutations. The comparative transmutation

performance results can be used by DOE to develop fuel cycle strategies for MA transmutation. The temperature and flux distributions in the AmBB assembly obtained through a detailed pin-by-pin analysis will be utilized to develop the irradiation plan of the AmBB fuels in the ATR.

#### **Accomplishments:**

Various AmBB transmutation options in the 600 MWe Advanced Sodium Technological Reactor for Industrial Demonstration (ASTRID) reactor developed by the French CEA were evaluated with both self-sustainable and Am-burning modes of operations. The objective of the self-sustainable mode operation is to re-utilize the Pu and Am generated in ASTRID by achieving a conversion ratio of about 1.0. In this case, the recovered Pu and Am are recycled respectively in the driver fuel and in 24 AmBB assemblies loaded with 15% Am content, and other minor actinides are sent to the repository. The purpose of the Am-burning mode operation is to burn the recovered Am from the spent fuel of existing LWRs with the conversion

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ratio less than 1.0. One row of radial blanket containing 66 assemblies in ASTRID was replaced with AmBB assemblies, and the results show that the recovered Am from 1,800 MWe PWRs can be transmuted.

In order to compare the transmutation performance between the heterogeneous vs. homogeneous modes, and metallic vs. oxide fuels, the AmBB transmutation performances were also evaluated in a 1000 MWth Advanced Burner Reactor (ABR), which was developed for TRU transmutation based on a homogeneous transmutation strategy under the GNEP program. The transmutation results in the ABR with the self-sustainable mode inform that the metallic fuel core requires fewer AmBB assemblies when compared to the oxide fuel core because harder spectrum with metallic fuel generates less Am and burns it effectively. Overall, the heterogeneous transmutation based on the AmBB concept does not affect the core performance characteristics and inherent safety

features. However, the decay heat and the rate of helium generation are significantly increased in the AmBB fuels, and a separate study is required to confirm the AmBB fuel integrity during the operation.

A detailed pin-by-pin analysis of AmBB assemblies in ASTRID and in the metallic-fueled ABR core was also conducted to identify the irradiation condition of the AmBB fuels in the ATR. This analysis shows that there are significant neutron flux and fuel temperature gradients in the AmBB assembly. Thus, in order to achieve a uniform temperature and flux profile over the fuel residence time, a rotation of the AmBB assembly was proposed.

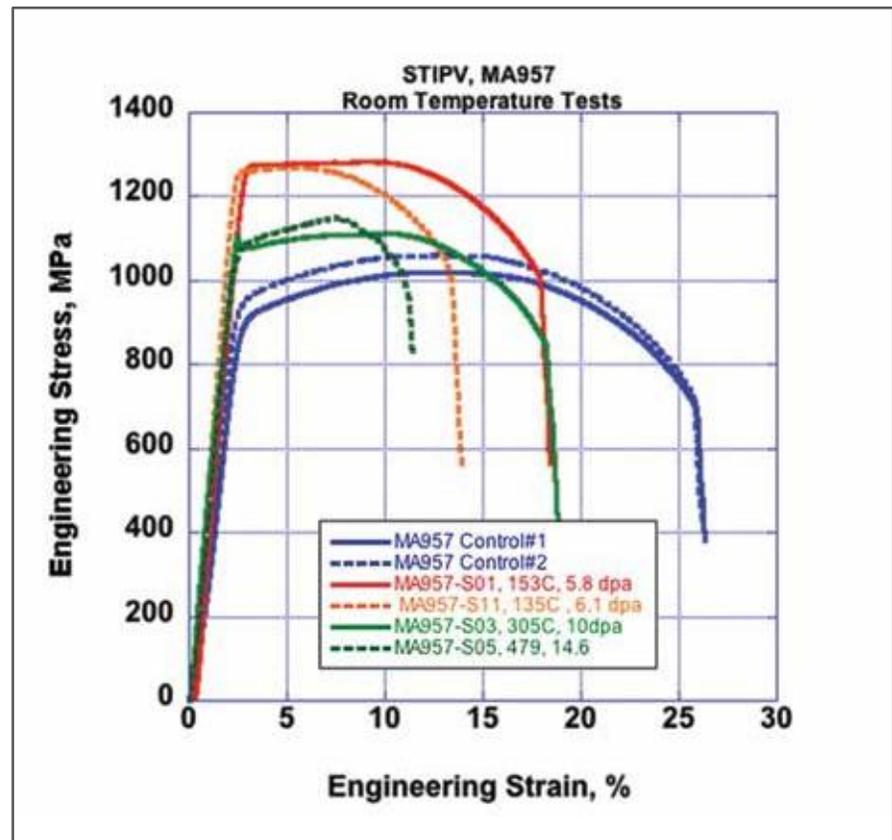
## 3.3 AR CORE MATERIALS

### Tensile Testing of STIP V irradiated Radiation Tolerant Materials

Principal Investigator: Tarik Saleh

Collaborators: Matthew E. Quintana, Toby J. Romero, Stuart A. Maloy, Peter Hosemann, David Krumwiede

Figure 1. MA957 tensile data from the STIPV irradiation, showing excellent ductility up to 14.6 dpa. This is a first generation ODS alloy, that has shown consistently excellent ductility in neutron irradiations.



This research provides insight into the mechanical behavior of fast reactor candidate cladding materials after relevant neutron radiation damage. This work specifically looks the effects of irradiation dose on

mechanical properties after a number of test irradiations, including the STIPV irradiation in which samples of various ferritic/martensitic, FeCrAl and ODS alloys were irradiated to doses of up to 15 dpa under relevant fast reactor irradiation temperatures.

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### **Project Description:**

This work seeks to expand the database of mechanical testing data of candidate advanced fast reactor cladding materials. Research funded under this activity during FY2016 covered irradiations, shipping and mechanical testing of irradiated materials from 3 different irradiations:

1. The STIPV irradiation in the SINQ accelerator at the Paul Scherrer Institut. Covering a variety of advanced fast reactor cladding materials irradiated up to 15 dpa. Mechanical testing was performed on samples from this irradiation.
2. Initial examination of 14YWT NFA 1 samples irradiated up to 8 dpa in the BOR-60 reactor in Russia, including initial nanohardness data collected at UC Berkeley.
3. Preparations and negotiations to receive samples from the MATRIX irradiations from the PHENIX reactor in France, with doses up to 70 dpa. All irradiations provide specimens for high dose mechanical testing data to support development of cladding for advanced fast reactors.

### **Accomplishments:**

Mechanical testing was completed on STIPV irradiated materials (LANL). The stress/strain curves of MA957 from this testing is shown in figure 1. Testing was performed at room temperature. Samples show good retention of ductility after irradiation up to 14.6 dpa.

Samples from a BOR 60 irradiation were received and shipped to partner labs for analysis (IRPTEM specimens) (LANL->ORNL). Initial shear punch testing of BOR-60 TEM specimens was completed at LANL. BOR-60 irradiated 14YWT tubing was received, inspected, cut, and shipped to UC Berkeley for initial nanohardness testing. This work continues into early FY2017. Progress made on receiving PHENIX irradiated MATRIX specimens from CEA (LANL-CEA), this appears to be on schedule for receiving at LANL in FY 2017.

*The AFC-3F irradiation test examining the effects of fuel casting techniques on fuel behavior has been fabricated using both arc melting fuel and archival EBR-II fuel which has been fabricated through injection casting.*

## Update on ODS Tube Processing

Principal Investigator: Stuart Maloy

Collaborators: Eda Aydogan, G.R. Odette, John Lewandowski, Dave Hoelzer

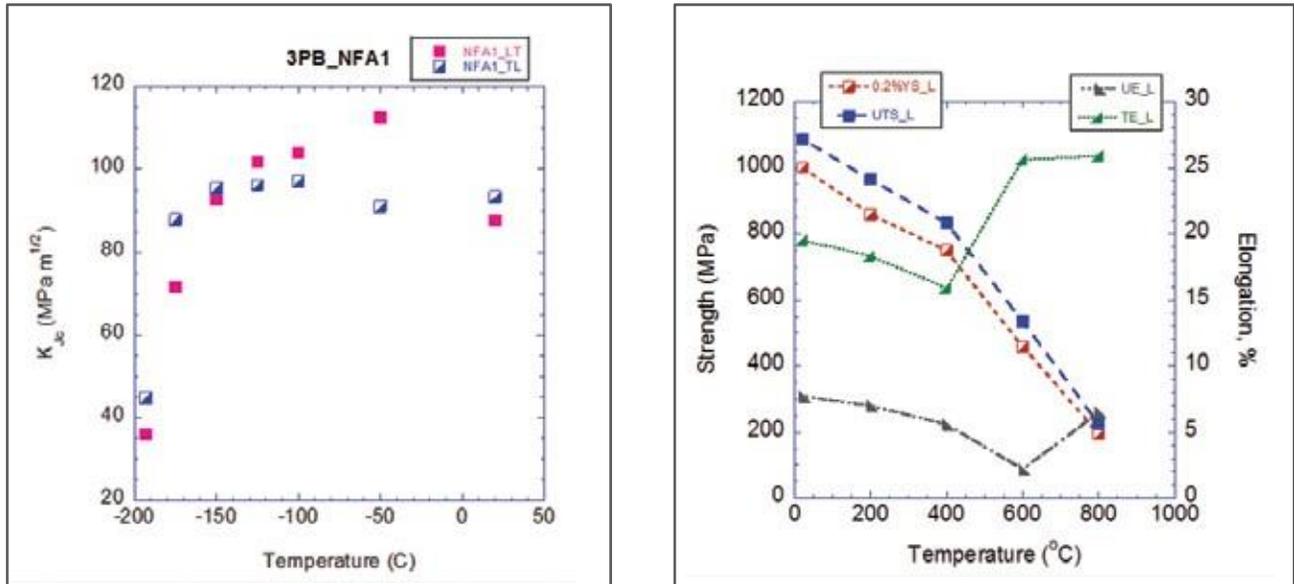


Figure 1. Graphs showing toughness measured to -200C and tensile properties up to 800C measured on FCRD-NFA1

The Fuel Cycle R&D program is investigating options to transmute minor actinides. To achieve this goal, new fuels and cladding materials must be developed and tested to high burnup levels (e.g. >20%) requiring cladding to withstand very high doses (greater than 200 dpa) while in contact with the coolant and the fuel.

### Project Description:

To develop and qualify materials to a total fluence greater than 200 dpa requires development and testing of advanced alloys and irradiations in fast reactors. Specimens of previously irradiated HT9 specimens are

being irradiated in a fast reactor to high doses (>200 dpa). In addition, improvements in the radiation tolerance of HT-9 are being made through minor changes in the composition. Advanced radiation tolerant materials with fine oxide dispersion strengthening are also being developed to enable the desired extreme fuel burnup levels. This fine microstructure provides an alloy with high strength at high temperatures and excellent radiation tolerance (e.g. reduced void swelling and ductility retention at low temperatures) but also increases the difficulty of producing engineering parts (e.g. thin walled tubes) from



Figure 2. Multiple tubes produced by hydrostatic extrusion at CWRU on heat of 14YWT (FCRD-NFA1) shown above.

these advanced materials. Thus, in this project research is underway to produce tubes using techniques such as high temperature hydrostatic extrusion, intermediate temperature plug drawing and low temperature pilger processing.

**Accomplishments:**

Through a collaborative effort between LANL, UCSB and ORNL a large heat (50 kg) of a nano-structured ferritic alloy (14YWT) was produced named FCRD-NFA1. Mechanical tests showed that the material from this heat possessed an excellent combination of high

temperature strength, tensile ductility and low temperature toughness. A summary of these properties is shown in Fig. 1.

Initially, the most promising results have been produced using high temperature hydrostatic extrusion at Case Western Reserve University. During this process, tubes are extruded at 815C with a 4:1 ratio to produce short sections of thin walled tubing (<0.5 mm wall thickness). An example of three of these tubes is shown in Fig. 2.

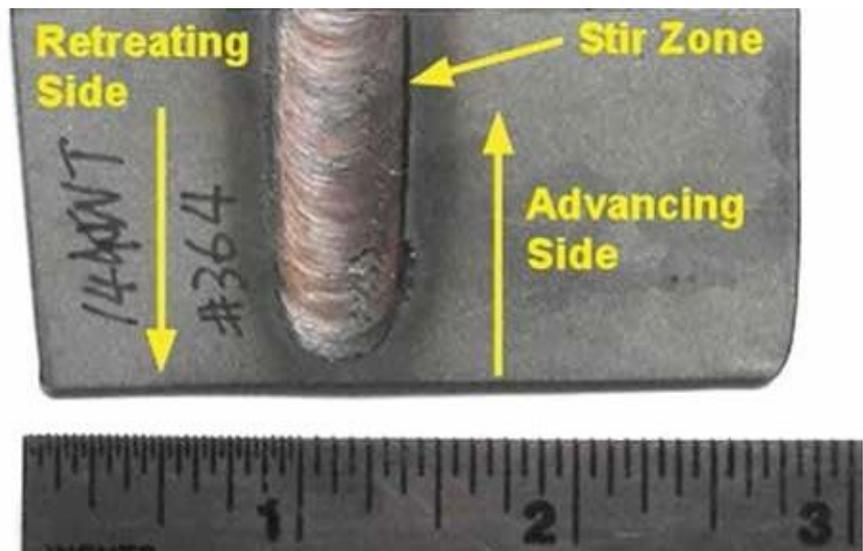
*This research is critical to the application of advanced ODS steels to engineering applications as one of the most difficult tasks is to produce tubing from these radiation tolerant, high strength steels.*

## Weld Development for Thin Wall Tubing of ODS Ferritic Alloys

Principal Investigator: D. Hoelzer

Collaborators: P.D. Edmondson, M.N. Gussev, W. Tang, Z. Feng

Figure 1. Digital image showing the bead-on-plate stir zone produced by FSW on the 1 mm thick plate of 14YWT.



Oxide dispersion strengthened (ODS) ferritic alloys, such as 14YWT, possess excellent high-temperature mechanical and radiation resistant properties to high dose and temperature exposures, but are notoriously difficult to join. Fusion welding techniques melts the materials to produce the joint, but this destroys the oxide dispersion that accounts for the high temperature properties and radiation resistance of ODS alloys. Friction stir welding (FSW) is a solid-state joining

technique originally developed within the aluminum industry that uses heat produced by friction between a moving pin tool rotating at high speeds and the material being joined. For this reason, the FCRD program began exploring FSW for joining thin plates and ultimately thin wall tubing of 14YWT. Although FSW has been shown to maintain the oxide dispersion in numerous ODS alloys, further research is necessary to avoid defects such as porosity, worm-holes, grain growth and tensile residual stresses that can degrade the joint quality.

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### **Project Description:**

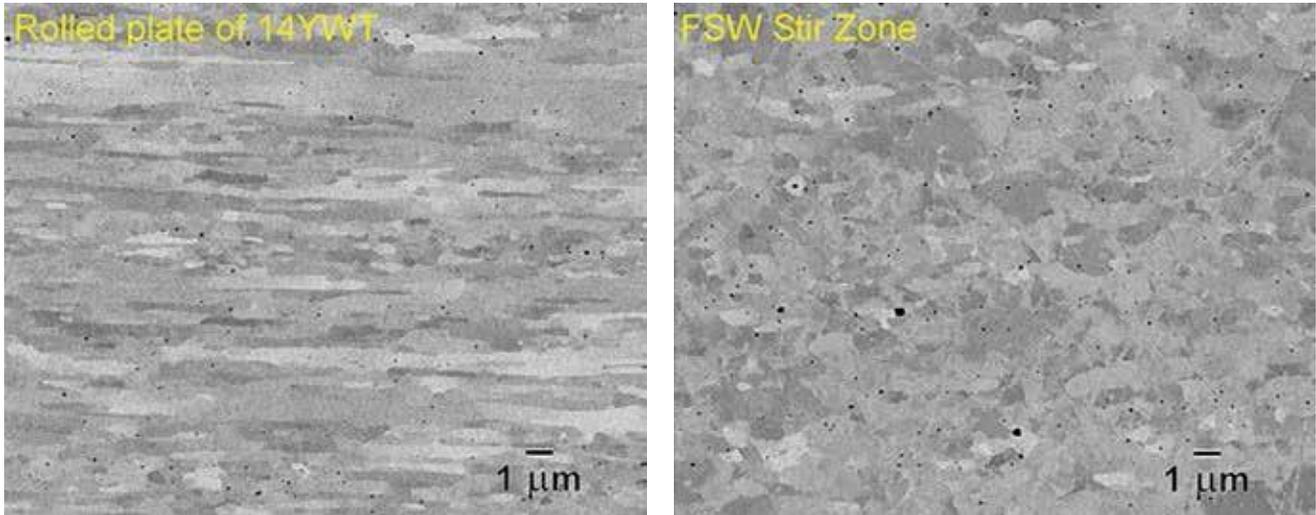
This research is focused on development of friction stir welding (FSW) for joining thin plate and thin wall tubing of the advanced ODS 14YWT. The main task involves developing and optimizing the FSW parameters that will ensure weld joints that are defect free and do not degrade the salient microstructural features of 14YWT that are responsible for the excellent high-temperature mechanical properties that are degradation resistant to neutron irradiation environments at high-doses and temperatures. The most challenging aspect of this research will be the successful joining of plate and tubing of 14YWT with thicknesses that are less than 1 mm. Therefore, this project will be pioneering the knowledge base for joining thin plate and tubing of ODS alloy since the few publications investigating FSW for joining ODS alloys have all used thicker plate samples. This research also includes tasks on characterizing the microstructure and assessing the mechanical properties of specimens prepared from the stir zone produced by FSW in a 1 mm thick plate of 14YWT. Since the microstructure produced in the stir zone is very complex, a novel approach based

on optical extensometry, referred to as digital image correlation (DIC), is being explored for determining the mechanical properties of the stir zone. This technique measures localized extensions, or strains, at a large number of points along the gage of miniature tensile specimen prepared from the stir zone during the tensile test. The results of this research support the main goal of the fuel cycle R&D program on developing fabrication methods for producing thin wall fuel cladding from advanced ODS alloys and together will help maximize nuclear fuel utilization of next generation nuclear reactors.

### **Accomplishments:**

The trial FSW run was successful in producing a non-penetrating bead-on-plate stir zone (SZ) on a 1 mm thick plate of 14YWT. Figure 1 shows a digital image of the bead-on-plate stir zone and the advancing side indicating the direction of the spinning pin tool during the FSW run. A specimen containing a cross-section of the stir zone was prepared and examined using optical microscopy (OM) and scanning electron microscopy (SEM). A separate specimen was

*Friction stir welding is currently the leading technique for successfully joining thin plate and ultimately thin wall fuel clad tubing of advanced ODS alloys that will benefit the development of next generation nuclear reactor technologies.*

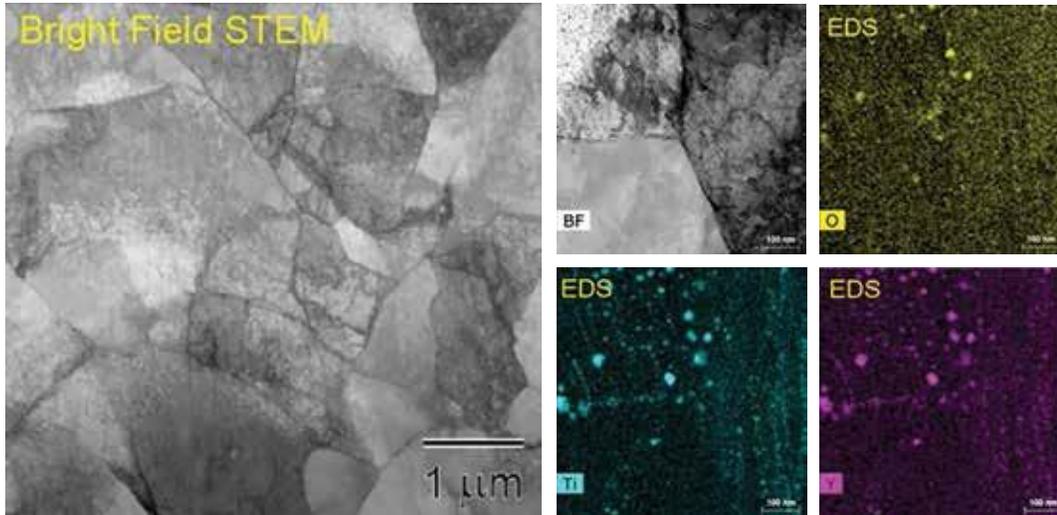


*Figure 2. SEM back scattered electron micrographs showing the grain structures observed in the rolled plate and FSW stir zone of 14YWT.*

also prepared using the focused ion beam (FIB) in-situ lift-out technique and examined using scanning transmission electron microscopy (STEM). As shown in Figure 2, the results of the microstructural characterization study showed the grains present in the SZ were isotropic and irregular in shape but were very similar in size compared to the elongated grains present in the rolled plate of 14YWT. Several cracks oriented horizontally were observed but were mostly located on the retreating side of the SZ. These cracks may have formed due to insufficient pressure being exerted on the top surface of the plate by the shoulder and pin tool during the FSW run. The analysis of the FIB specimen using high resolution STEM and

element mapping by X-ray energy dispersive spectroscopy showed the presence of the Y-Ti-O particles in the SZ and that many of the oxide particles were very small with only a few that appeared to have increased in size. Overall, the FSW parameters used to produce the bead-on-plate SZ in the 1 mm thick plate of 14YWT were nearly optimized and these results demonstrate that FSW is a promising method for joining advance ODS alloys.

Figure 3. Bright-field STEM micrograph showing the grain structure and EDS element maps showing the distribution of Y-Ti-O particles near the triple point grain junction that were observed in the specimen prepared from the SZ of the 1 mm thick plate of 14YWT.



The first attempt in exploring digital image correlation (DIC) as a method for studying the tensile properties of the stir zone produced by FSW in the 1 mm thick plate of 14YWT was successful. Two tests were conducted on miniature (SS-Mini-2) tensile specimens that were fabricated from the 1 mm thick plate of 14YWT. It was found that the specimens exhibited high strength (1035-1130 MPa ultimate stress) and good ductility (13-14.4% total elongation). The magnitude and spatial distribution of strain on the gage section of both specimens was accurately determined during the tensile test and DIC analysis. It was determined that strain localization occurred away from the center of

the gage section in both tensile specimens during the test. The strain localization rapidly increased in magnitude at these locations, which was the location where plastic instability occurred and ultimately where the specimens failed. These results indicate that the DIC analysis is a very promising technique for assessing the tensile properties of the complex microstructure associated with FSW stir zone of joined 14YWT plates. The next DIC experiments will be performed on SS-Minni-2 tensile specimens that were prepared from the SZ of the 1 mm thick plate of 14YWT.

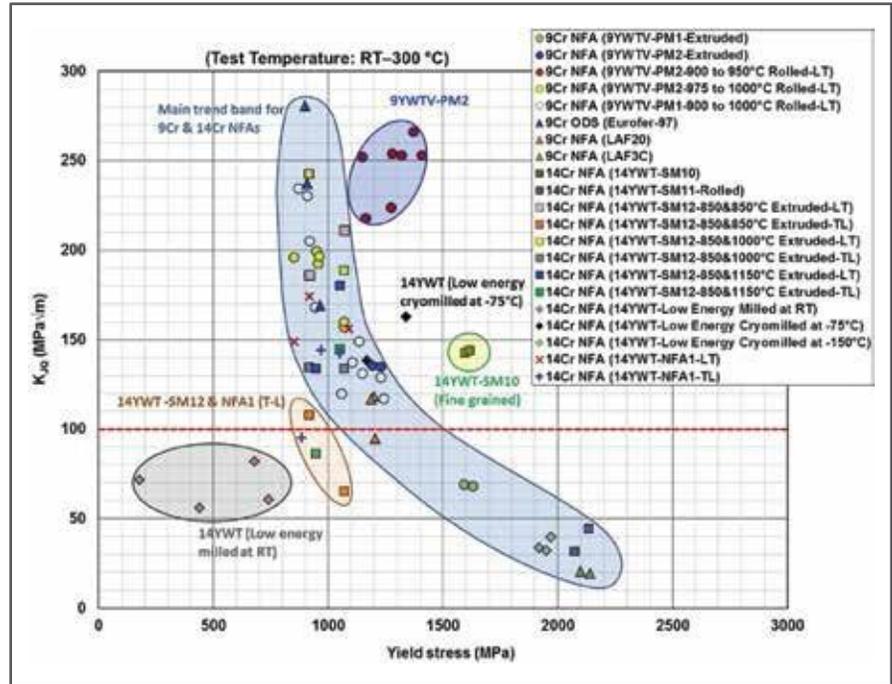
*Figure 3. Bright-field STEM micrograph showing the grain structure and EDS element maps showing the distribution of Y-Ti-O particles near the triple point grain junction that were observed in the specimen prepared from the SZ of the 1 mm thick plate of 14YWT.*

## Fracture Toughness Testing of ODS Alloys

Principal Investigator: T.S. Byun

Collaborators: D.T. Haelzer and S.A. Maloy

Figure 1. Fracture toughness versus yield stress plot for the 9Cr and 14Cr NFAs tested in a low temperature region of 22–300 °C.



The Fe-Cr alloys with ultrafine microstructures are primary candidate materials for sodium cooled fast reactors because of their excellent high temperature strength and high resistance to radiation-induced damage such as embrittlement and swelling. Mainly two types of Fe-Cr alloys have been developed for the high temperature reactor applications: the quenched and tempered ferritic-martensitic (FM) steels hardened primarily by ultrafine laths and carbides and the powder metallurgy-based nano-

structured ferritic alloys (NFAs) by nanograin structure and nanoclusters, which often called advanced oxide dispersion-strengthened (ODS) alloys. Although significant characterization results support possible applications of some new Fe-Cr alloys in various irradiation conditions, however, some structural integrity controlling properties, such as the fracture toughness in the as-fabricated condition, need to be separately assessed because they can limit the fabrication processes of components and such baseline properties also determine much of the in-service performance.

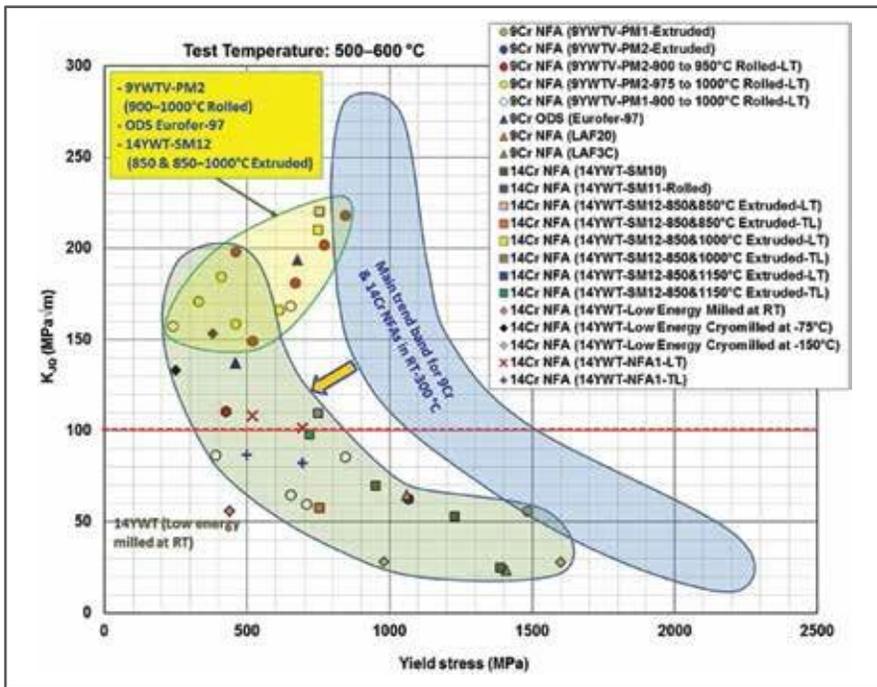


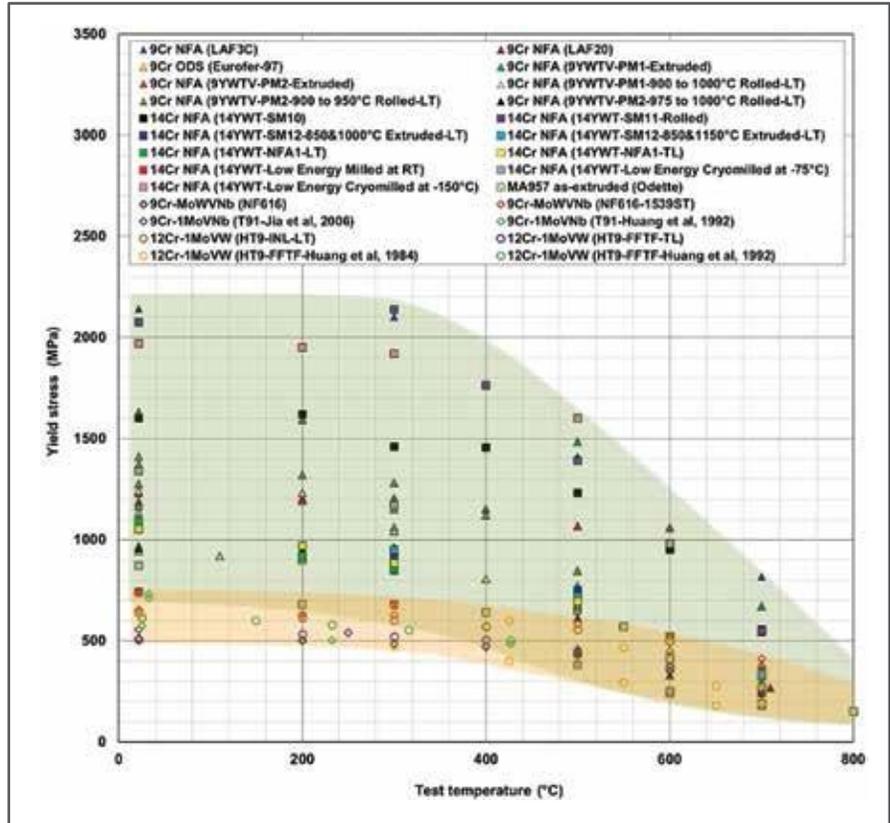
Figure 2. Fracture toughness versus yield stress plot for the 9Cr and 14Cr NFAs tested in a high temperature region of 500–600 °C.

## Project Description

This research aimed to assess the baseline mechanical properties of the Fe-Cr alloys, focusing on the fracture toughness behavior of NFAs. A testing and analysis task was planned to effectively reveal differences and similarities in the temperature and strength dependences of fracture toughness in the Fe-Cr alloys to provide a comparative assessment of their high-temperature structural performance. The task involves integration of a large amount of fracture

toughness and tensile strength data including both the newly-obtained test data and the data from open literature by the authors and others, all of which were produced using miniaturized testing techniques. New static fracture resistance (J-R) tests have been carried out using either 15 mm long three-point bend (TPB) specimens, 12.5 mm diameter disk compact tension (DCT), or 12.5×12.5 mm CT specimens. It is often observed that the fracture toughness within a

Figure 3. Temperature dependence of yield stress in ferritic-martensitic steels and nanostructured ferritic alloys.



materials group is inversely proportional to strength. Such an inversely proportional toughness-strength relationship can be established when the variation in processing route for an alloy does not substantially change its fundamental properties such as grain boundary bonding, alloy composition,

constituent phases, and stacking fault energy. Therefore, the fracture toughness data were plotted against corresponding yield stress data to reveal any true improvement for each materials group and to elucidate the effect of other factors such as test temperature and specimen orientation. Further, the fracture toughness and tensile property data were replotted against test

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temperature to display and compare the temperature dependences of those properties. This mechanical property assessment will help select the best performing materials or improve their properties for application to the next generation reactors.

### **Accomplishments**

A comparative assessment of fracture toughness behavior was successfully performed for Fe-Cr alloys. As seen in Figure 1, the fracture toughness versus yield stress plots confirmed that the fracture toughness was inversely proportional to yield strength. It was found, however, that the toughness data for some NFAs were outside the band of the integrated dataset at a given strength level, which indicates either a significant improvement or deterioration in mechanical properties due to fundamental changes in deformation and fracture mechanisms. When compared to the behavior of NFAs, the FM steels have shown much less strength dependence and formed narrow fracture toughness data bands at a significantly lower strength region. Figure 2 shows that

only a few NFAs including 9YWTV-PM2 and 14YWT-SM12 with proper thermomechanical treatments and ODS Eurofer-97 could demonstrate promising high-temperature fracture performance. The NFAs generally have an advantage of lower DBTT but some suffer a steep decrease in fracture toughness with temperature at above 500 °C, which strongly varies among different NFAs. The vast majority of FM steels have sufficient fracture toughness ( $> 100 \text{ MPa}\cdot\text{m}^{1/2}$ ) over a wide temperature range of RT-700 °C, and the datasets in the low and high temperature ranges from weak fracture toughness-strength relationships within narrow strength ranges.

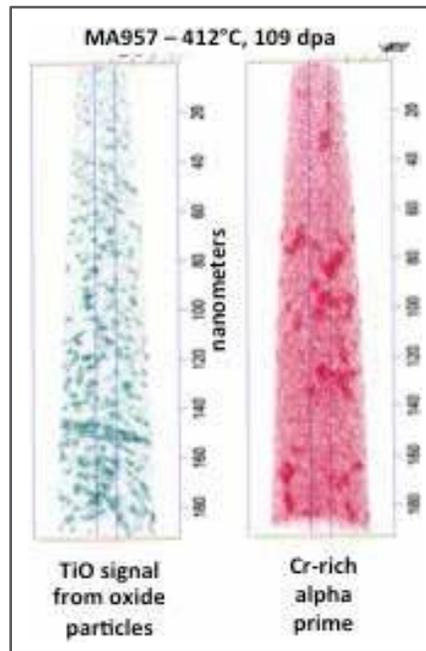
*Fracture toughness versus strength plots can provide a clear assessment of high temperature mechanical performance for advanced high-temperature reactor materials.*

## High Dose Ion Irradiation Testing of Radiation Resistant Fast Reactor Alloys

Principal Investigator: Mychailo Toloczko

Collaborators: Jing Wang, Peter Hosemann, Nathan Bailey, Victor N. Voyevodin, Frank A. Garner

Figure 1. Examples of APT reconstructions showing the distribution of TiO (oxide particles) and Cr (alpha prime precipitates) in neutron irradiated MA957.



The use of ion irradiations to estimate the neutron irradiation response of fast reactor (FR) clad and duct materials is an area of rapid growth. There are a number of reasons for using ion irradiations for materials studies, the most important one being that high ion irradiation dose levels can be rapidly achieved, an aspect that is important because of dwindling availability of fast reactors for materials irradiation performance studies, and because rapid accumulation of dose allows accelerated materials studies. An

important metric for the usage of ion irradiations is gauging how well it estimates neutron irradiation effects on an alloy. The primary means of assessing irradiation effects is by studying the microstructure of the material. The research presented here summarizes the status of an ongoing study aimed at comparing the neutron irradiation and ion irradiation induced microstructures of a relevant ferritic alloy. The data obtained to-date show excellent similarity in microstructural changes for neutron irradiation and ion irradiation exposures.

### Project Description:

The objective of this task is to show the degree of similarity or difference in the effects of ion irradiation and neutron irradiation on prototypic FR clad and duct materials so that the usefulness of ion irradiations for materials development scoping studies can be established. Establishing the viability of ion irradiations for FR clad and duct studies is extremely valuable to gauging which currently available materials are most likely to survive to high dose, and it is also very beneficial for development of more advanced materials. Several comparisons were performed in the 1970s-1980s that suggested substantial differences may occur, but these studies were done under less optimal conditions. Therefore it is important perform new assessments using modern techniques.

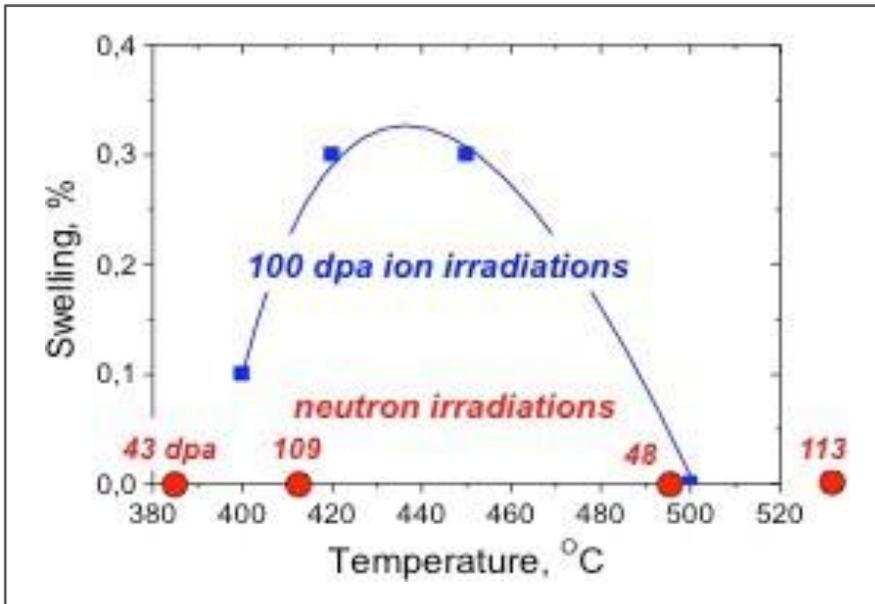


Figure 2. Comparison of void swelling in neutron irradiated and ion irradiated MA957. Either very low swelling (ion irradiations) or no swelling was observed.

*Early results indicate excellent promise for using self-ion irradiations to rapidly and more easily estimate the effect of neutron irradiations on the microstructure of candidate fast reactor clad and duct materials*

**Accomplishments:**

The foundation of this study is an oxide dispersion strengthened (ODS) ferritic alloy called MA957. This class of ferritic alloy was originally envisioned for cladding due to its excellent high temperature creep resistance, and MA957 was the first to be proposed as a candidate ODS ferritic alloy for use as fuel cladding in US-based fast reactors. Development of this alloy took place in the 1980s at PNNL, and short tubing pieces were irradiated in the Fast Flux Test Facility (FFTF) to doses of ~100 dpa over a wide range of temperatures in the late 1980s. Archival unirradiated material was

obtained and ion irradiated with 1.7 MeV Cr ions at the Kharkov Institute of Physics and Technology (KIPT) to 100 dpa for comparison.

Irradiation can cause many different kinds of changes to the microstructure such as the formation of new precipitates, the dissolution of existing precipitates, changes to the dislocation density, changes to the types of dislocations, and compositional in the vicinity of grain boundaries. Several different observation techniques are needed to assess all these possible changes. For the present work, the focus was on void swelling and

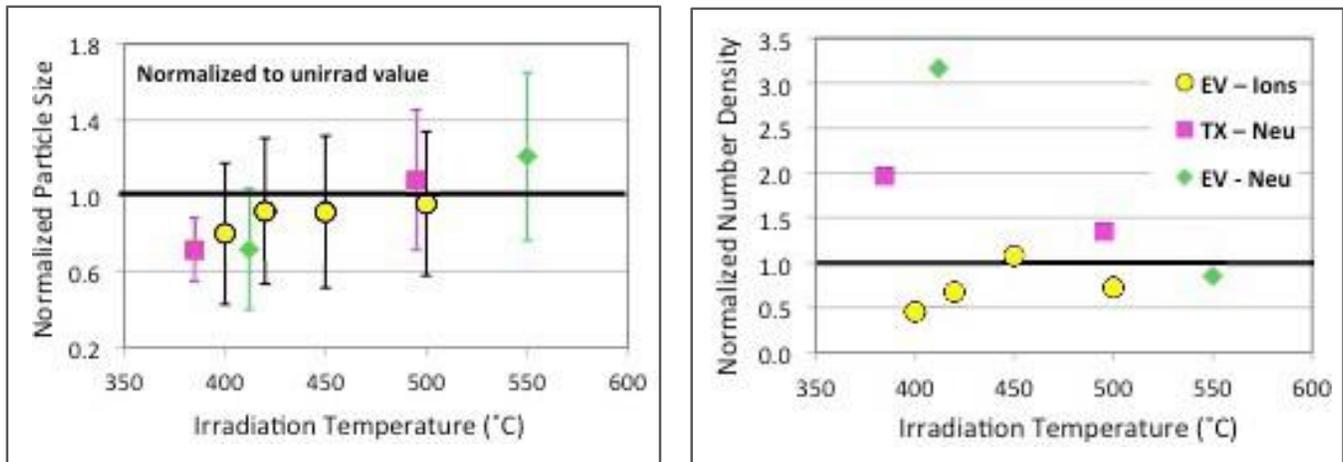
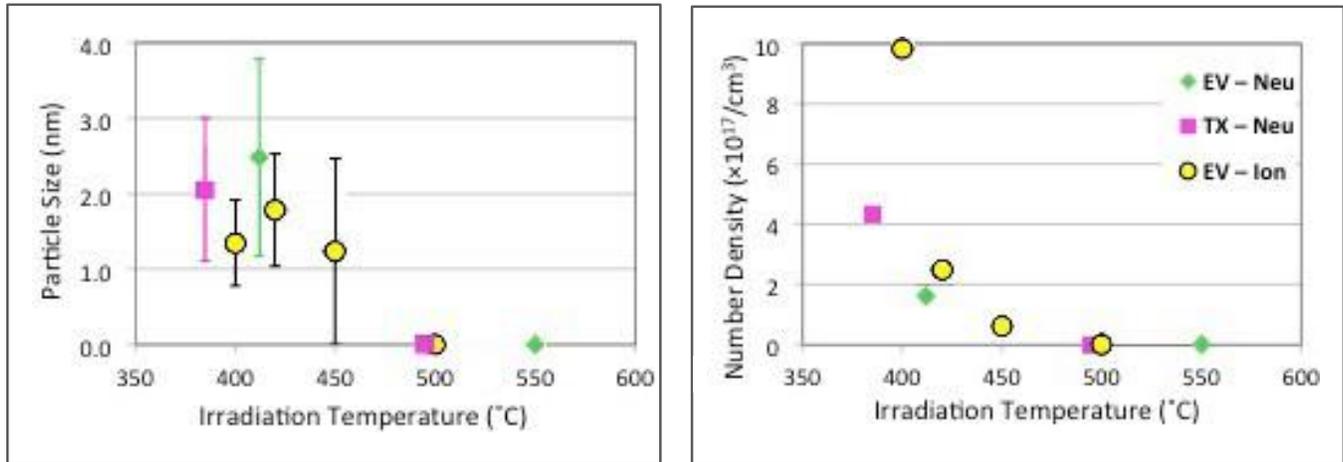


Figure 3. Comparison of oxide particle size and number density distributions in neutron and ion irradiated MA957 as a function of irradiation temperature. Values are normalized to the unirradiated material.

changes to precipitate populations. Void swelling was observed by transmission electron microscopy while the precipitate populations were observed using a technique called atom probe tomography (APT). This technique can differentiate between elements and provides a 3-dimensional atom-by-atom reconstruction of an ~30 nm diameter by 100 nm long cylinder of material extracted from the specimens. Accuracy in atom positions is ~1-2 nm, so a crystalline structure is not seen in the reconstructions, but it does an excellent job of showing the distribution of elements in the analysis volume. An example of two reconstructions showing Cr and TiO distributions within an MA957 needle are shown in Figure 1. Numerous reconstructions are needed and were performed to analyze a sufficient volume of material to generate

statistical confidence in the observed microstructures. These observations were performed at both PNNL and at UC Berkeley. For the ion irradiated specimen, the examinations were performed at a depth of ~150 nm from the surface of the specimen.

Void swelling measurements are presented in Figure 2. The neutron irradiated materials did not exhibit any swelling while the ion irradiated MA957 underwent a very small amount of swelling. While there is some difference between the swelling for the two irradiation methods, this difference is small compared to swelling values of >5% that may be considered problematic for causing interference when removing subassemblies or for maintaining desired coolant flow between fuel pins. The oxide particle populations are presented in Figure 3. The oxide particles are built-in features that add



creep strength to the material, stabilize the grain size, and possibly act to minimize swelling. For the neutron irradiations, two nominally identical batches of MA957 were examined and are noted as EV and TX. These are compared to the ion irradiated material, all of which was from the EV batch. Very good correspondence between neutron and ion irradiations can be seen for the oxide particle size, but some clear differences in number density are apparent. This is a somewhat surprising result because the oxide particles are thought to be very stable and should undergo very little change in population. This is an area where further examinations are planned. Lastly, the radiation-induced alpha-prime precipitate distributions are presented in Figure 4 where it can be seen that there is very good correspondence between neutron and ion irradiations. Not only are size and

number density values similar, but also the irradiation temperature where alpha-prime ceases to form appears to be very similar for the two irradiation methods.

In summary, these results generally show very good correspondence between ion and neutron irradiation induced microstructures in this first comparison, not only for approximate size and number density but also in trends with irradiation temperature. This is an excellent start for showing the viability of ion irradiations and encourages their use, but addition comparative studies are needed to fully explore the degree of similarity of microstructures that are produced. Such studies are underway at PNNL as well as at other institutions participating in the development of ion irradiation methods.

Figure 4. Comparison of alpha-prime precipitate size and number density distributions in neutron and ion irradiated MA957 as a function of irradiation temperature. Values are normalized to the unirradiated material.

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## Tube Pilgering Process Development

Principal Investigators: Ron Omberg, Ryan Webster, and Curt Lavender  
Collaborators: Wendy Bennett

Cladding developed with these processes will produce: (1) a more radiation tolerant fuel cladding and (2) fuel used to reduce the half-life of isotopes in spent or used nuclear that will need to be disposed.

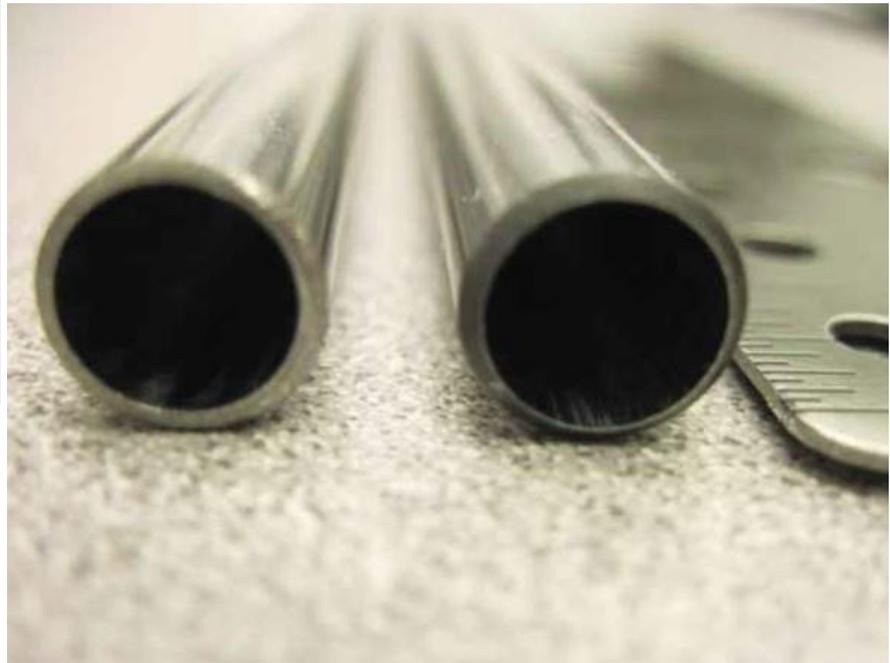


Figure 1. Extruded and Gun-Drilled MA956 Tubing Ready for Pilgering

Radiation effects to displacement doses greater than 400 displacements per atom (dpa) in a reactor environment are challenging to materials used for cladding and ducts. The activity is investigating processes for fabricating promising materials for cladding, specifically using extrusion and pilgering processes in a sequential mode.

### Project Description:

Extrusion and pilgering are promising techniques for fabricating advanced alloys such as MA956 and 14YWT suitable for producing tubing for reactor use. This project is using

the extrusion press located at the Pacific Northwest National Laboratory (PNNL) to perform sequential extrusions followed by pilgering using the pilger mill located nearby as Sandvik Special Metals. The goal and objective is to produce tubing samples with dimensions for in-reactor use as specified by Los Alamos National Laboratory (LANL).

With respect to the next generation of reactors, success in this activity will produce: (1) a more radiation tolerant fuel cladding and (2) a fuel used to reduce the half-lives of isotopes in spent or used nuclear that will need to be disposed. This in turn reduces the burden on any spent fuel or nuclear waste repository.

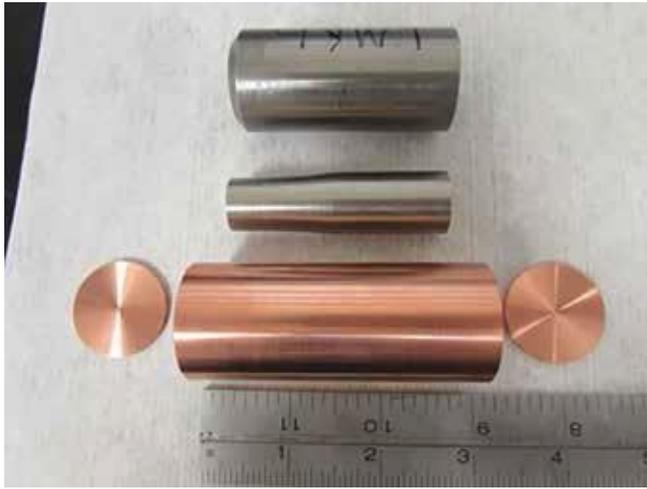


Figure 2. 14YWT Billet along with Components Ready for Assembly for First Stage Extrusion

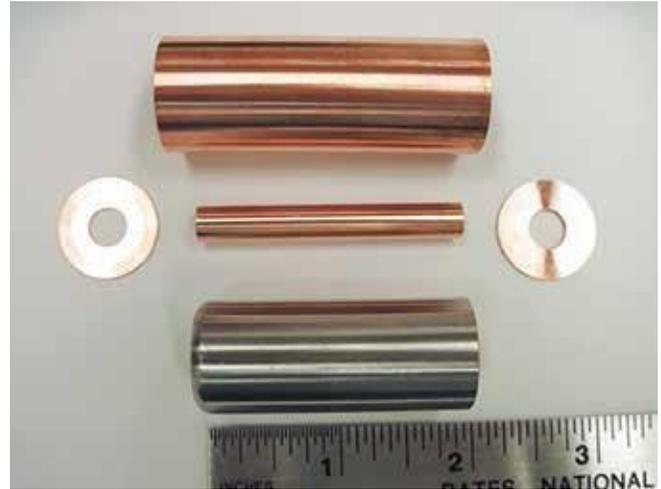


Figure 3. Machined Components Ready for Assembly and Second Stage Extrusion

### Accomplishments:

Accomplishments this fiscal year include proceeding sequentially through the extrusion process using a two stage extrusion process. This allows a sequential volume (and area) reduction to occur with manageable temperatures, pressures, and ram rates. As an initial start, MA956 was extruded and then gun drilled to prepare samples for pilgering. Two such samples ready for pilgering at Sandvik are shown in Figure 1. As follow-on, 14YWT billets have been fabricated for extrusion in a two-stage extrusion process. A 14YWT billet with components ready for

assembly for the first stage extrusion is shown in Figure 2. Subsequent to this, machined components ready for a second stage extrusion are shown in Figure 3. As the second stage extrusion is intended to produce a hollow thick tube, special tooling is required for the extrusion press. This tooling was fabricated and is shown in Figure 4. Subsequent pilgering steps will occur in October when time is allocated to this program by Sandvik on their pilger mill.

## 3.4 AR Irradiation Testing & PIE Techniques

### Progress and Status of Irradiations in the AFC Series in ATR

Principal Investigator: Steve Hayes and Doug Dempsey

Collaborators: Emily Swain, Brian Gross, Dan Chapman, Cody Hale, Susan Case, Misti Lillo



Figure 1. AFC – 3F capsules ready for reactor insertion

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**Project Description:**

The Irradiation Testing work package is responsible for new experiment design, shipments of both new and irradiated experiments, and activities associated with keeping experiments in the ATR for the ongoing irradiation tests.

Irradiation Testing is one work package in support of the overall technical objectives of the FCRD/AFC program.

**Accomplishments:**

Two new experiments were designed and fabricated this Fiscal Year; AFC-3F and AFC-4C. AFC-4C commenced irradiation and is expected to be in the irradiation phase of the experiment until FY-2020. AFC-3F

was designed, fabricated, and shipped to ATR for reactor insertion but will begin irradiation in early FY-2017 due to unanticipated ATR outage delays. AFC-3F is significant in that it is the first U-Pu experiment fabricated in several years due to facility unavailability.

Experiment AFC-4A was removed from the irradiation phase of the experiment and shipped to the Hot Fuel Examination Facility (HFEF) where Post Irradiation Examination (PIE) has commenced.

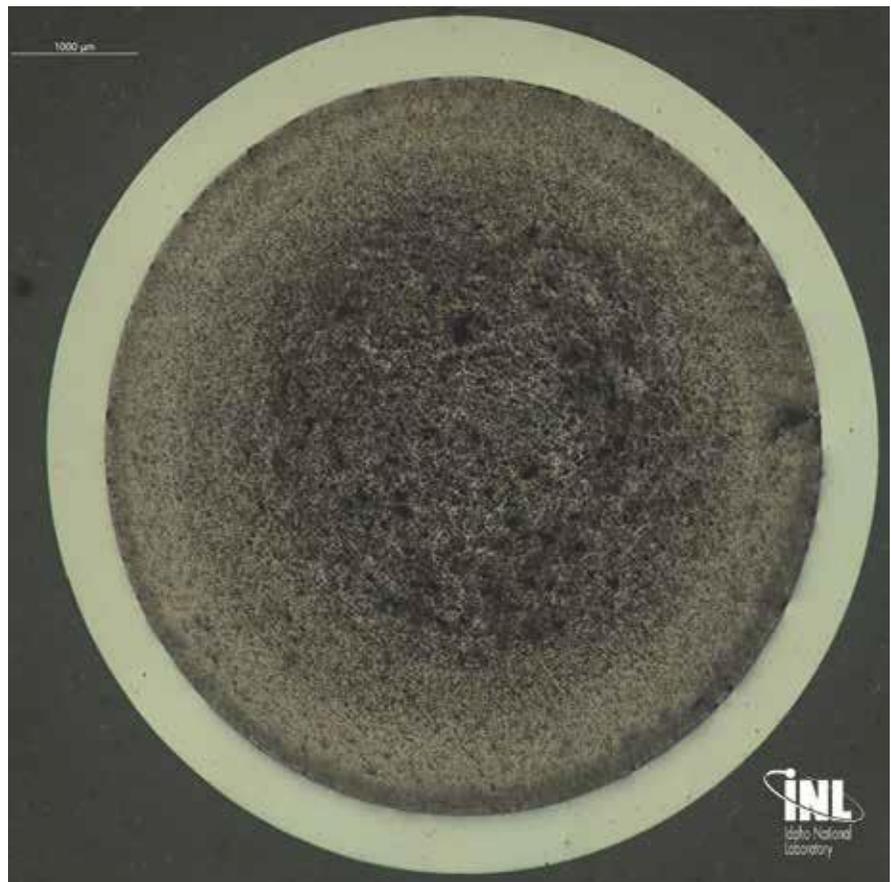
*Observing the effects of irradiation on materials and differing material compositions provides crucial information regarding the direction for further research and ultimately determining the solution*

## Postirradiation Examinations of FUTURIX-FTA

Principal Investigator: Jason Harp

Collaborators: Heather Chichester, Luca Capriotti, Douglas Porter

Figure 1. Optical microscopy montage collected from a polished cross section of FUTURIX-FTA DOE1 (U-28.3Pu-3.8Am-2.1Np-31.7Zr)



Postirradiation Examination (PIE) results from the baseline examination of the 4 FUTURIX-FTA irradiation experiment rodlets was completed and documented in “Baseline Postirradiation examination of the FUTURIX-FTA Experiments” (INL/LTD-16-40088). This experiment was conducted in the true fast neutron spectrum conditions of the Phénix sodium fast test

reactor in France. The FUTURIX-FTA irradiations provide a validation case that helps validate if ATR pseudo-fast spectrum testing adequately reproduces fuel performance behavior in a true fast spectrum reactor like Phénix or if there are tangible differences that need to be considered when evaluating fuel performance based on ATR testing.



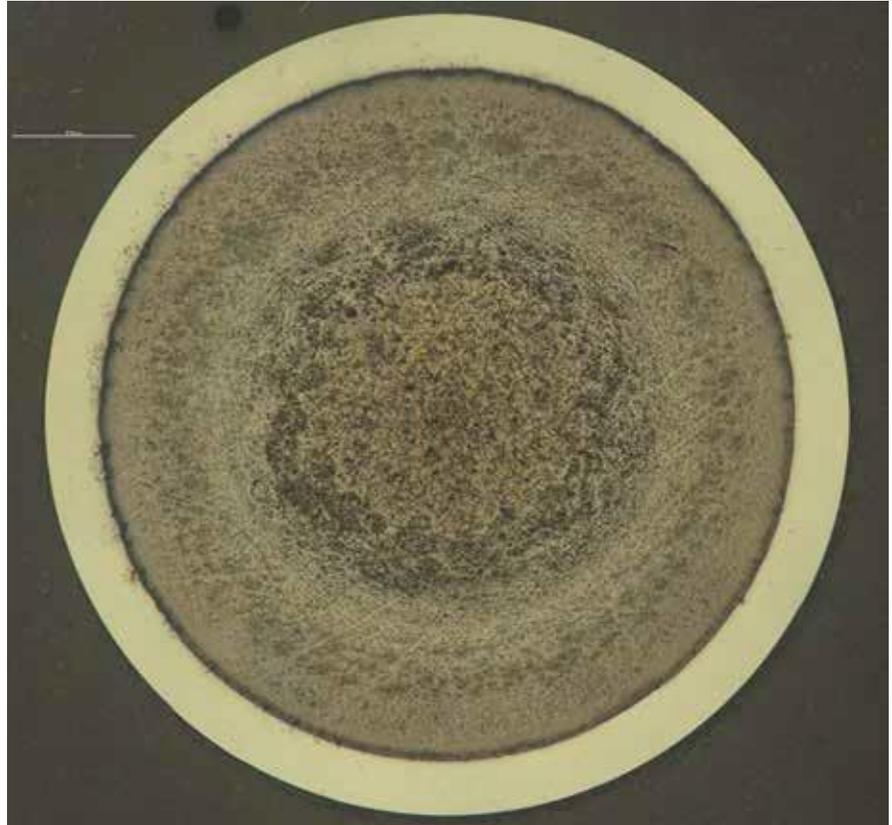
### **Project Description:**

The baseline PIE of the FUTURIX-FTA rodlets is designed to evaluate the macro to microscopic fuel performance properties of the irradiated non-fertile and low-fertile metallic and nitride fuel compositions. This is accomplished by applying the existing available techniques including gamma spectrometry scanning, neutron radiography, dimensional inspection, fission gas release analysis, optical microscopy, and chemical analysis. Fuel performance in metallic fuels includes fuel cladding chemical interaction (FCCI), fuel alloy redistribution, swelling, and fission gas release among others. The results of the FUTURIX-FTA PIE are important to

demonstrate the applicability of Cd shrouded quasi-fast spectrum testing in the INL Advanced Test Reactor by comparing the results to sister pins irradiated in the AFC-1 irradiation test series. Any tangible difference in fuel performance between the two systems will need to be understood and considered as quasi-fast spectrum testing continues. The compositions irradiated in the FUTURIX-FTA tests also will provide important fuel performance data on potential low-fertile and non-fertile fuel alloys that could potentially be used to transmute and eliminate plutonium and other minor actinides in a future fast spectrum reactor.

*Figure 2. Higher magnification detail of radial microstructure revealed in FUTURIX-FTA DOE1 cross section.*

*Figure 3. Optical microscopy montage collected from a polished cross section of FUTURIX-FTA DOE2 (Pu-10.5Am-0.3Np-41.6Zr)*



#### **Accomplishments:**

Baseline PIE is now complete for the FUTURIX-FTA Pins. All indications are the fuel performed well during the irradiation. These exams include visual inspection of the pins, neutron radiography, dimensional inspection of the fuel pin diameter, gamma spectrometry examination of the pins, fission gas release analysis, optical microscopy and chemical burnup analysis. The results of all these exams will be summarized in

documented in “Baseline Postirradiation examination of the FUTURIX-FTA Experiments” (INL/LTD-16-40088). Non-destructive examinations of the FUTURIX-FTA pins were completed in FY’ 15 and the destructive exams were completed in FY’ 16. Prior to destructive exams gamma tomography scans were performed on select areas of the metallic fuel pins. This revealed the radial distribution of several fission and activation products including Cm-243, Co-60, Cs-134, Cs-137,

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Eu-154, Mn-54, Ce-144, Ru-106, and Sb-125. The distributions of different fission products can be used to infer some information about the stability of different fission products in the fuel matrix and may have some impact on other fuel performance properties such as FCCI. The fission gas release from the fuel pins was measured and was found within expectations from historic fission gas release. In these transmutation fuels He release from the decay of Am is also important and was measured as well. This measure of He release (62% for the low-fertile metallic pin and 64% for the non-fertile metallic pin) in the metallic transmutation fuel containing Am is quite unique.

The microstructure of the fuel pins was evaluated by optical microscopy. The cross section from the low-fertile DOE1 MNT-20Y (U-28.3Pu-3.8Am-2.1Np-31.7Zr) pin is shown in Figure 1 and in greater detail in Figure 2. The local porosity in Figure 1 is evenly distributed locally and is spherical in shape throughout most of the fuel, but the very outer periphery of the fuel the porosity is smaller and somewhat lenticular. This would tend to indicate that the underlying crystal structure of the fuel material is cubic everywhere except the outer 500  $\mu$  m. In a binary or ternary metal fuel lenticular pores are formed in fuel at tempera-

tures below the  $\gamma$  phase transition temperature (776°C). The cladding temperature of DOE1 was predicted to be approximately 550°C, so there should be a region of the fuel showing non-spherical porosity that was irradiated below the  $\gamma$ -U phase transition temperature. There is also a small (~20  $\mu$  m) interaction layer at many locations between the fuel and the cladding. Optically this cannot be positively identified. This may be the initiation of a FCCI layer, or it may be an artifact of fabrication. There was a Zr rich layer at the edge of the as-fabricated samples.

The cross section from non-fertile DOE2 MNT-21Y (Pu-10.5Am-0.3Np-41.6Zr) pin is shown in increasing detail in Figure 3, and Figure 4. The cladding has several spots with debris and tarnishing from polishing. The black marks on the cladding in Figure 3 should not be mistaken for cladding degradation. This cross section shows evidence of constituent redistribution and phase separation. There are several rings of microstructure present in Figure 3 that suggest different phases that were present during irradiation and these phases are likely driven by different thermal conditions present in the fuel during irradiation. The Pu-Am-Zr system is not as well understood as the U-Zr

**Initial Postirradiation Examinations complete on fast reactor irradiated fuel samples that will be used to validate current transmutation irradiation testing in US test reactors.**

or the U-Pu-Zr system, but many of the same observations made on the DOE1 cross section can be made and tied back to known properties of the Pu-Zr system. As-fabricated, XRD of the fuel revealed the predominant microstructure to be  $\delta$ -(Pu,Zr). During irradiation the cladding temperature was about 550°C, so it is likely that during irradiation both  $\delta$ -(Pu,Zr) and  $\epsilon$ -(Pu,Zr) were present in the fuel. Both of these phases are cubic and the porosity structure suggests a cubic crystal structure. As in DOE1 there is a  $\sim 20 \mu\text{m}$  layer that is likely a Zr rich layer from fabrication, or it could be a FCCI interaction layer. There are at least 5 major zones of microstructure in the fuel. The first three from the outer radius of the fuel inward about 1mm all have small porosity and varying amounts of what appears to be phase separation supposed by different colors in the microscopy which often indicates various different levels of oxidation. Certain layers oxidize faster than others presenting a different color. In the next 750

$\mu\text{m}$  the porosity of the fuel changes significantly and becomes much larger. The color of the fuel matrix also suggests that this is a more homogeneous phase in the fuel. The interior of the fuel has a great deal of phase separation. The orange phase tends to cluster and is surrounded by a lighter matrix phase. The matrix phase is shown in detail in Figure 4. If it is assumed that the appearance of the stacked structure is indication of a different chemical phase not a different crystallographic orientation in the material, the matrix phase has a stacked structure that is suggestive of the decomposition of  $\nu$ (U,Zr) into  $\alpha\text{U}$  and  $\delta\text{UZr}_2$ . The stacked structure in this fuel could be the decomposition of  $\epsilon$ -(Pu,Zr) into  $\delta$ -(Pu,Zr) and  $\alpha\text{Zr}$ . If Zr redistribution did drive additional Zr up the temperature gradient to the center of the fuel this explanation is also more likely. The exact nature of these phases and the location of the Am in the fuel will require further investigation likely with at least an EPMA exam. Micro-XRD and the preparation of transmission (TEM) lamella by Focused Ion Beam (FIB) would also be helpful to fully understand this system.



*Figure 4. Detail of phase separation present in the central region of FUTURIX-FTA DOE2*

Burnup and fission density measurements were also performed on the 4FUTURIX-FTA rodlets. The determination of burnup and fission density was performed using the measured mass of a specific fission product in the fuel, the cumulative fission yield of that specific fission product, and the total mass of actinides present in the sample. This method is sometimes referred

to as the "Fission Product Monitor Residual Heavy Atom" technique. The measured burnup was found to be in fair agreement with the predicted burnup from simulations. The greatest discrepancy was found in the non-fertile rodlets. This is not surprising as the nuclear data associated with Pu and Am are less well known than the U data.

## Postirradiation Examinations of AFC-3,4 Experiments

Principal Investigator: Jason Harp

Collaborators: Heather Chichester and Douglas Porter

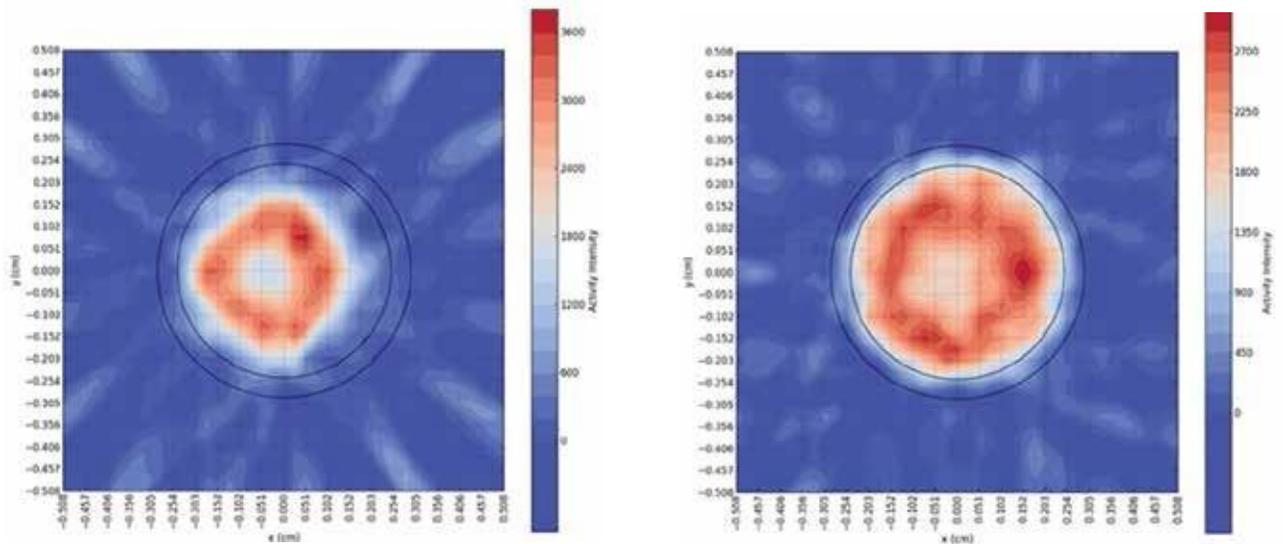


Figure 1. Gamma tomography showing the relative distribution of Ce-144 in (a) AFC-3A R4 and (b) AFC-3A R5. The black circles in the plot represent the cladding around the fuel. Intensity beyond the cladding is an artifact of the technique

Postirradiation examination continued on the AFC 3 and 4 series of irradiations. This series of transmutation fuels experiment is focused on investigating novel fuel forms and alloys that can extend the lifetime of transmutation fuel beyond the maximum lifetimes achieved historically (20 atom % burnup). Currently the results of the AFC-3A/B experiment which included U-Zr, U-Mo, and U-Zr-Pd alloys, with solid and annular geometries, and smear densities of 75 and 55% are being finalized and advanced PIE is being performed if possible. The

non-destructive PIE of AFC-4A which includes U-Zr, U-Mo and a U-Mo-Ti-Zr alloy with and without Pd additions have also been completed.

### Project Description:

The objective of the transmutation fuels campaign is to investigate promising fuel forms that can be used to reduce the long term radiotoxicity of a spent fuel repository. Once a fuel form has been analyzed for acceptability from a pre-irradiation properties perspective, it is necessary to assess the irradiation fuel performance. There are a variety of fuel performance criteria used to judge a fuel form including fuel cladding

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chemical interaction (FCCI), fuel redistribution, swelling, and fission gas release to name a few. Other factors such as the ability to remotely fabricate a fuel form and actinide utilization/distribution are also used to limit the scope of considered fuel forms. The fuel performance of any fuel form under investigation must also be compared to the historic performance of solid, sodium bonded, 75% smear density, U-10Zr that was used very successfully as one of the driver fuels for EBR-II. The new fuel forms investigated in the AFC-3 and AFC-4 would ideally out-perform U-10Zr and be viable beyond 20 atom % burnup, where FCCI becomes a major concern for U-10Zr fuels, and allow fast reactors to reach high levels of actinide utilization. With current data, the most promising fuel form under investigation appears to be U-Pd-Zr alloys. Although high burnups have not yet been investigated the performance of solid, sodium bonded, 75% smear density, U-10Zr with 1 or 2 wt.% Pd appear to have performed well and SEM examinations of this fuel show the Pd is collocating with rare earths preventing them from interacting with the iron in the cladding causing FCCI.

### **Accomplishments:**

In this fiscal year, PIE continued on different rodlets from the AFC-3 and AFC-4 irradiation tests. The baseline examination of AFC-3A/B were completed in the previous fiscal year, but reporting and advanced PIE examinations continued. Highlights from the PIE of AFC-3A/B were published at the American Nuclear Society Annual Meeting and included previously unreleased gamma tomography evaluations of the final product distribution of Ce and Cs in both solid U-Pd-Zr alloy and annular U-10Zr alloy fuel (see Figure 1). The entire baseline PIE results from AFC-3A/B are planned for journal publication next fiscal year. Two SEM mounts from AFC-3A rodlets were also prepared for analysis. One of these mounts from AFC-3A R5 (U-1Pd-10Zr) was examined by SEM. Elemental analysis mapping analysis of this mount confirmed that rare earth

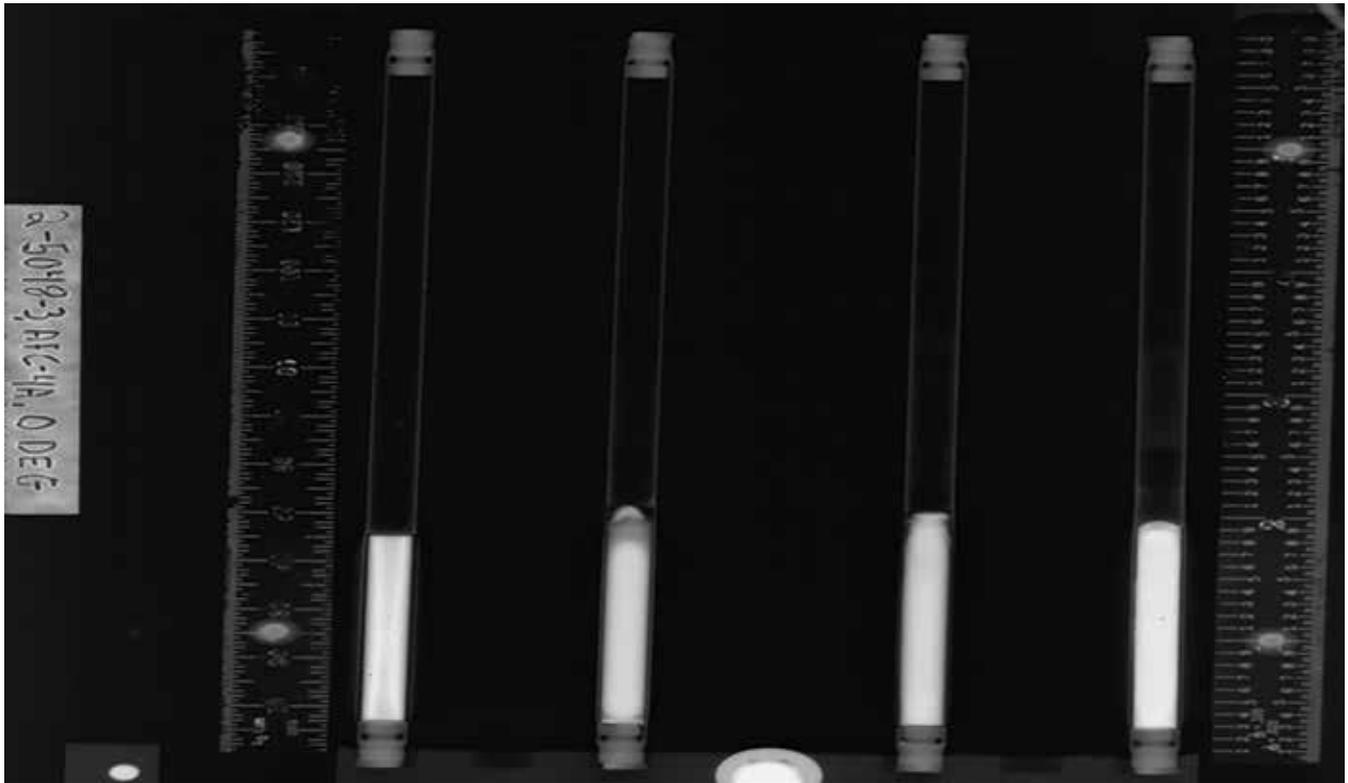


Figure 2. Neutron Radiography of the AFC-4A rodlets from left to right:

AFC-4A R1 (U-10Mo, annular 65% smear density),

AFC-4A R3 (U-5Mo-4.3Ti-0.7Zr, solid, 75% smear density)

AFC-4A R4 (U-5Mo-4.3Ti-0.7Zr-2Pd, solid, 75% smear density), AFC-4A  
R5 (U-10Zr, solid, 75% smear density)

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fi products (Nd, Ce, La, Pr) have formed stable intermetallic compounds with the available Pd present in the fuel. Rare earth fi product migration to the martensitic stainless steel cladding and subsequent attack of the cladding resulting in FCCI is a major life limiting phenomenon in metallic fast reactor fuels. The next round of AFC rodlets available for examination are from the AFC-4A irradiation. Non-destructive irradiation of these rodlets was initiated. This irradiation contained an annular, He bonded U-10Mo rodlet which is not anticipated to perform well based on the FCCI observed in U-10Mo fuel in AFC-3A/B. The performance of U-MTZ alloy in AFC-4A which is designed to retain the favorable FCCI properties of U-10Zr,

but not redistribute the alloying metals, was initially evaluated in these exams. Nothing catastrophic has been observed in the available data. Capsule examinations, disassembly from the capsule, visual exams, neutron radiography and axial gamma spectrometry scans were performed on the AFC-4A rodlets. Some interesting swelling of the MTZ alloy was observed in the neutron radiography shown for one angle in Figure 2. Optical microscopy among other exams next fi year will be necessary to fully evaluate AFC-4A. A U-MTZ+Pd alloy and U-10Zr control were also irradiated in AFC-4A.

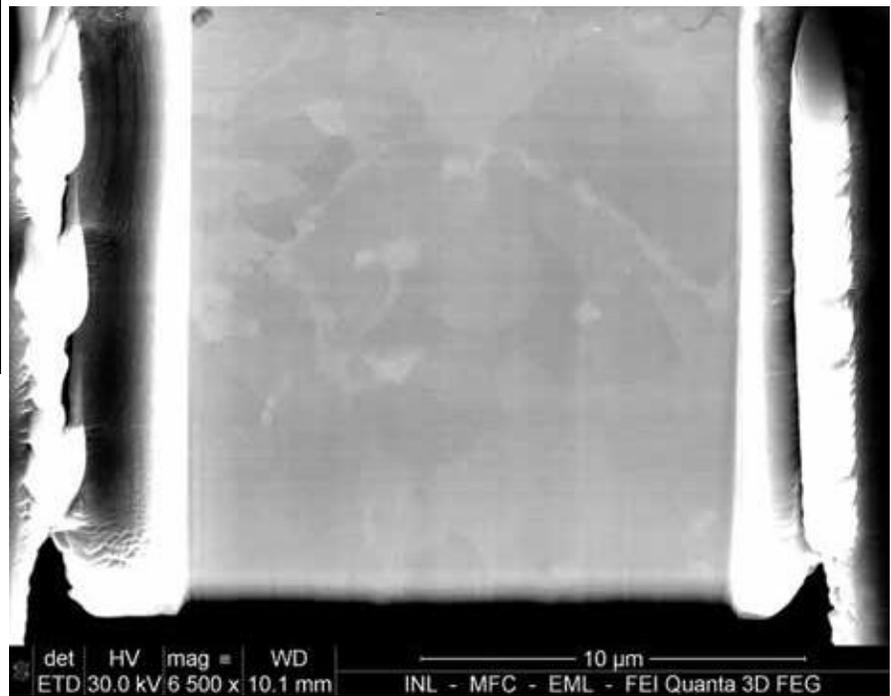
## Electron Microscopy of AFC-2,3 Experiments

Principal Investigator: Jason Harp

Collaborators: Brandon Miller, Tammy Trowbridge, Douglas Porter

Electron microscopy of irradiated fuel helps researchers understand the underlying physical phenomena driving macro-scale fuel performance in transmutation fuels.

Figure 1. FIB liftout from HT-9 cladding of a cross section taken from the MFF3 irradiation test. Rare earth fission products have attacked along intergranular pathways in this sample



Samples were prepared to examine with a scanning electron microscope (SEM) portions of fuel from an irradiated AFC series experiment. Some SEM was possible on subsized samples from the AFC-3A R5 irradiation test. Work also continued on a historic sample prepared from fuel irradiated in the Fast Flux Test Facility (FFTF). Additional SEM examinations were made of the FFTF sample and Transmission Electron Microscope (TEM) samples were prepared by Focused Ion Beam (FIB) and examined to better understand the phenomena observed in SEM exams.

### Project Description:

Until recently the dose rates of AFC experiments have been prevented electron microscopy examination of samples from the AFC series of irradiation tests. This has prevented fully understanding the distribution of major and fission product elements in the microstructure of the irradiated fuel. Understanding the distribution of major elements (U, Pu, Zr) and fission products is essential to fully comprehend the performance of irradiated fuel. Any measurement of irradiated transmutation fuel is fairly unique and often either fission of a kind or near fission of a kind type of measurement.

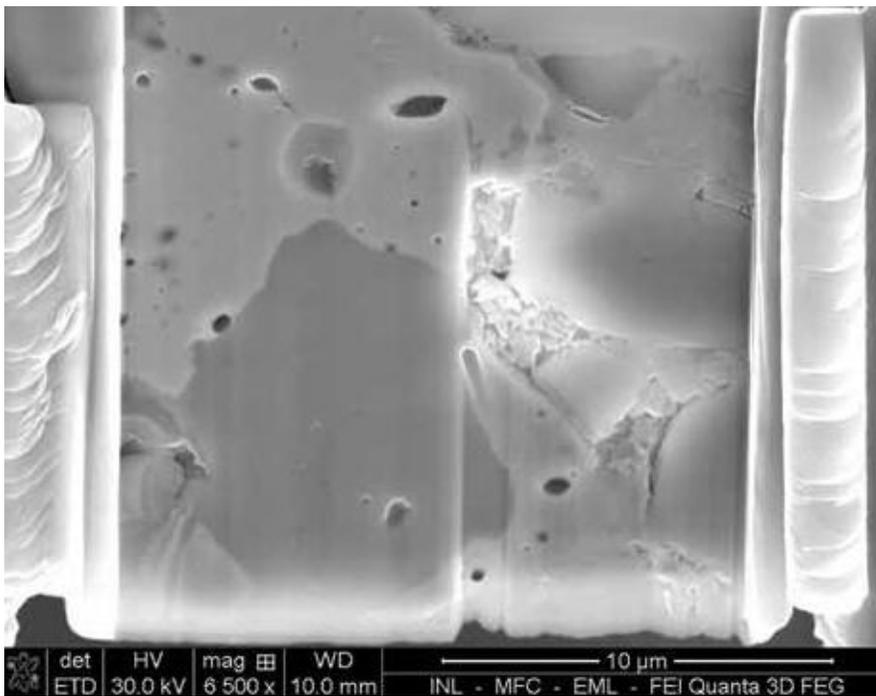


Figure 2. FIB liftout from the outer radius of fuel from a cross section taken from the MFF3 irradiation test.

### Accomplishments:

Three SEM samples were prepared from AFC-3A R5 (U-1Pd-10Zr, solid), AFC-3B R2 (U-4Pd-10Zr, annular), and AFC-2D R6 ((U<sub>0.75</sub>,Pu<sub>0.20</sub>,Am<sub>0.03</sub>,Np<sub>0.02</sub>)O<sub>1.98</sub>). In order to lower the dose to acceptable levels these samples were cut into approximately 1mm wide strips across a diameter, mounted and polished. The sample from AFC-3A R5 was examined on the Idaho National Laboratory Electron Microscopy Laboratory SEM. Energy dispersion spectroscopy mapping of this sample confirmed that rare earth fission products formed during irradiation had formed precipitates with the Pd alloying element present in the fuel.

This result is encouraging for the continued investigation of Pd as an alloying element that can prevent Fuel Cladding Chemical Interaction (FCCI).

Additional SEM and TEM examinations were performed on a cross section from the MFF3 experiment (irradiated U-10Zr). The SEM measurements were made to collect additional information on the redistribution of constituents in this fuel and the structure of FCCI layers. The FIB was used to create TEM lamella (see Figure 1 and 2). TEM analysis of these lamella will be used to better understand the migration of fission products in FCCI. The TEM analysis will also help determine differences in rare earth fission product behavior in the FCCI layers.

## Progress in Advanced NDE Development and Demonstration at LANL

Principal Investigator: Mark Bourke

Collaborators: Adrian S. Losko, Sven C. Vogel, Ken.J. McClellan, Stewart L. Voit

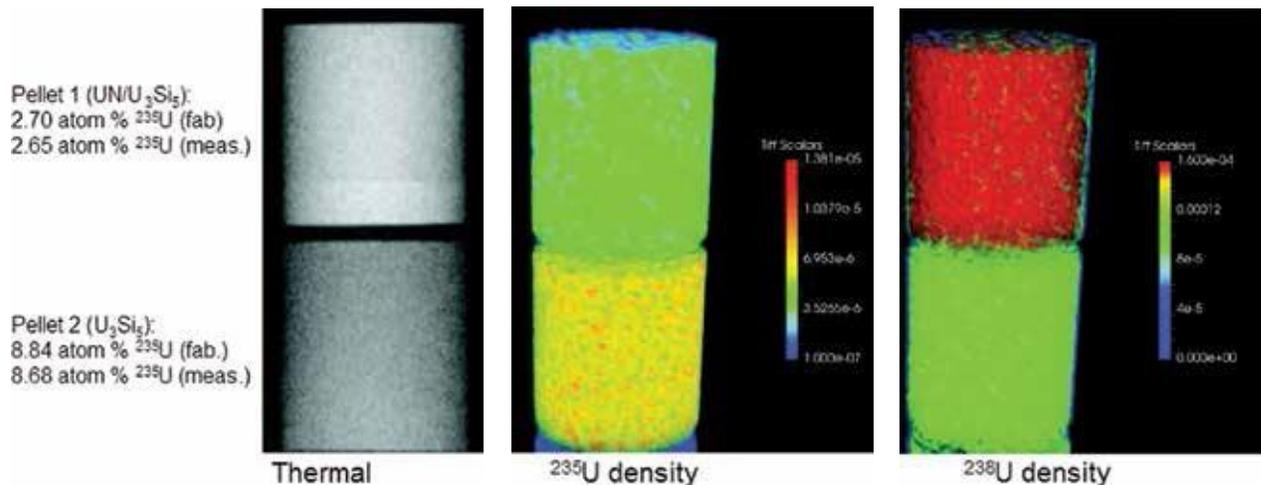


Figure 1. Neutron radiographs and tomographic reconstructions of two pellets with different  $^{235}\text{U}$  enrichment; Thermal radiographs (left)  $^{235}\text{U}$  tomographic reconstruction (center)  $^{238}\text{U}$  tomographic reconstruction (right) Pellets are 9.4mm long and 8 mm in diameter.

Imaging and diffraction measurements of ceramic and metallic nuclear fuels have been completed at the Los Alamos Neutron Scattering Facility. The measurements are non-destructive and provide insight about phase composition, defects, enrichment, microstructure and texture. A series of UN / U-Si pellets of varying stoichiometry and isotopic enrichments were examined this year. The techniques span length-scales from the crystal cell to the microstructure to macroscopic features. The goal of the measurements is to provide insight on fabrication, document the pre-irradiation condition of test samples and to develop techniques that could be applied to irradiated fuel. Imaging and spectroscopic measurements are performed at the Los Alamos Neutron Science Center (LANSCE) pulsed

spallation neutron source using a pixilated time-of-flight neutron imaging detector that can record thermal and epithermal energies simultaneously. Isotopic enrichment is determined from fitting absorption resonances and tomographic reconstructions provide 3D distributions of features and isotopics.

### Project Description

The technical objective of this project is the development and improvement of a suite of neutron based nondestructive characterization tools that can be applied to fresh and, potentially, irradiated nuclear fuel formulations. Energy-resolved neutron imaging and absorption resonance spectroscopy measures isotope distributions. By performing multiple measurements tomographic characterization is possible. When combined with neutron diffraction the data provide non-destructive multi-

lengthscale characterization opportunities. Current objectives include improving the sensitivity of measuring neutron absorption resonances to provide spatially resolved characterization of isotopic enrichment. Opportunities for neutron induced prompt gamma analyses are being assessed as is the development of laser-driven pulsed neutron sources which would enable deployed application of these techniques rather than requiring the neutron production infrastructure at Los Alamos.

The techniques are being applied to composite Accident Tolerant Fuel (ATF) candidates that are fabricated in the Fuels Research Laboratory at Los Alamos. For example crystallographic unit cell dimensions indicate whether mixing occurs between the composite constituents. The formation of U(N,Si) compounds in UN/U<sub>3</sub>Si<sub>5</sub> composites is apparent from changes in lattice parameters of the constituent phases. Texture measurements indicate whether preferred orientation is introduced during sintering (which influences thermal stresses and crack formation). Tomography verifies homogeneity of pellets as well as measuring gaps between pellet and cladding or between rodlet and capsule (which determine the thermal conditions experienced by pellets in reactor).

By maximizing the insight obtained from irradiation testing this work contributes to the accelerated certification of new fuel formulations. The current activity is focused on docu-

menting the pre-irradiation condition of fuels before irradiation in the Advanced Test Reactor. However the unique probe characteristics of neutrons mean that the same techniques could be applied to irradiated fuels to provide pre- and post-irradiation characterization of exactly the same material. This could provide unique insight on microstructural evolution that takes place during irradiation and guide the application of destructive techniques to representative and atypical regions of behavior.

#### **Accomplishments**

Neutron tomography and neutron diffraction characterizations were performed on nine pellets; four UN/U-Si composite formulations (two enrichment levels), three pure U<sub>3</sub>Si<sub>5</sub> reference formulations (two enrichment levels), and two reject pellets with visible flaws (to qualify the technique). The <sup>235</sup>U enrichments ranged from 0.2 to 8.8 wt. %. The nitride/silicide composites are candidate compositions for use as Accident Tolerant Fuel (ATF). The monophase U<sub>3</sub>Si<sub>5</sub> material was included as a reference. Pellets from the same fabrication batches will be inserted in the Advanced Test Reactor at Idaho during 2016.

Analysis of neutron resonance measurements (averaged over the centerline of 8 mm diameter pellets and with 45 minutes of data collection enrichment levels) yielded enrichment levels that

***New pulsed neutron techniques offer quantitative 3D, non-destructive pre irradiation characterization of isotopics, density, crystallography and microstructure in fresh fuel pellets.***

agreed with the nominal fabrication levels to within 0.1 wt%. Tomographic reconstructions of the <sup>238</sup>U and <sup>235</sup>U isotopic components demonstrated homogeneity of the pellets to at least 100 microns. Thermal neutron tomography demonstrated the ability to measure dimensions of pellets and the rodlet capsule gap double-encapsulated in ATR irradiation capsules. Spatially resolved microstructural analysis by neutron diffraction complemented the imaging and provides archival characterization of the pre-irradiation state. Advances in the analysis of the neutron imaging relied on the ENDF/B-VII.1 database, in conjunction with the ORNL nuclear cross-section analysis code SAMMY.

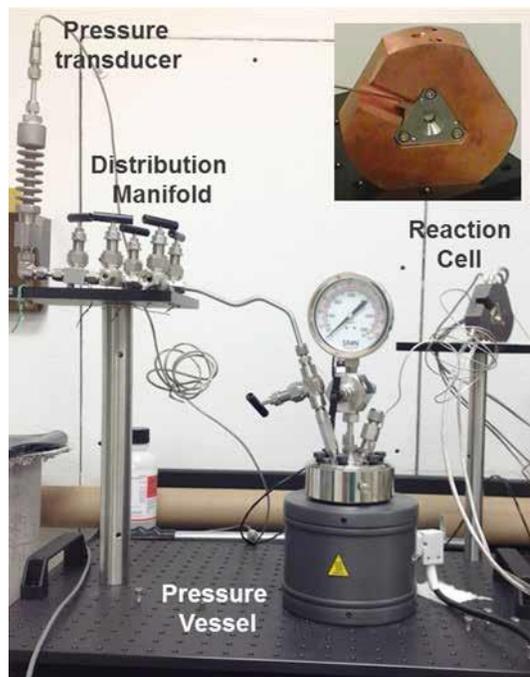
## 3.5 CAPABILITY DEVELOPMENT

### Investigating corrosion at interfaces under extreme environments using synchrotron methods

Principal Investigator: Simerjeet K. Gill

Collaborators: Mohamed Elbakhshwan, Lynne Ecker, Raul Rebak

Figure 1. In situ sample environment developed at BNL, showing the pressure vessel for steam generation, distribution manifold and pressure transducer which communicates with lab-view based platform for remote pressure monitoring. Inset shows the reaction cell where sample is mounted and copper heater with thermocouple for remote T monitoring



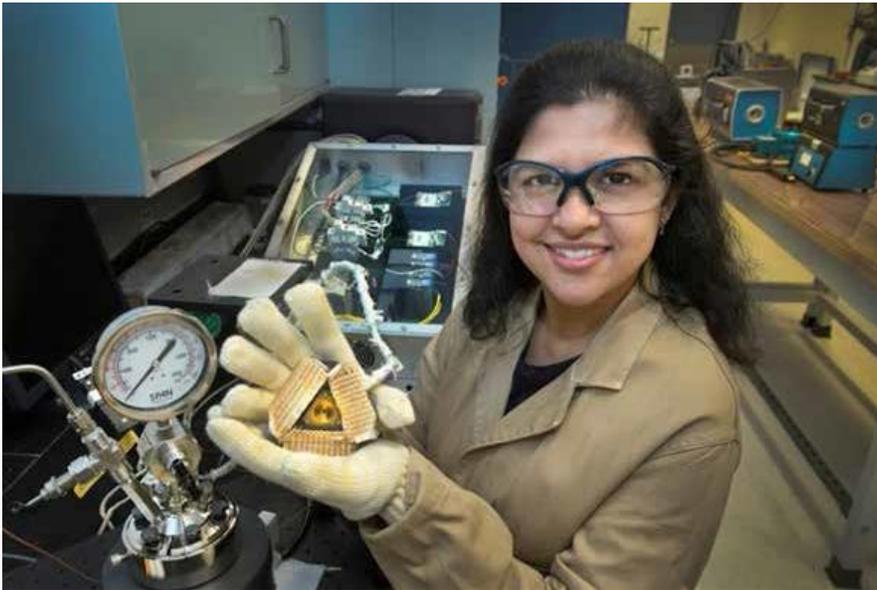
There is a lack of fundamental understanding of oxidation and associated corrosion mechanisms that occur at nuclear cladding interfaces under extreme conditions of temperature, pressure, and corrosive environments. High synchrotron X-ray energy and brightness, coupled with advanced sample environments, present an exceptional opportunity to understand the fundamental interfacial processes involved in the degradation of materials in extreme environments. In our work, a new in situ sample environment has been

designed and developed to study the interfacial interactions of nuclear cladding alloys with high temperature steam, using synchrotron X-ray diffraction (XRD) for in situ structural analysis. The use of the in situ sample environment is exemplified by monitoring the oxidation of metallic zirconium during exposure to steam. In addition, the structural and chemical analysis of oxide layer formation on advanced FeCrAl based cladding (Alloy-33) using synchrotron based XRD, X-ray photoelectron spectroscopy (XPS) and X-ray fluorescence methods (XRF) is discussed.

#### Project Description:

Following the accident at the Fukushima nuclear power plant, the U.S. Department of Energy is focusing on understanding the materials degradation and performance of current zirconium based claddings and developing nuclear fuels and cladding with an enhanced tolerance of accidents. The broad technical objective of the current research is to develop technology and high resolution synchrotron methods for fundamental understanding of corrosion behavior of zirconium based and advanced accident tolerant claddings.

Corrosion of current zirconium alloy fuel cladding in water or steam and the associated hydrogen pickup is a limiting factor for increasing fuel



*Figure 2. Principal Investigator Simerjeet K Gill showing the in situ sample environment*

burn-up in current and future reactors. Zirconium alloys with slightly different compositions and microstructures show widely different corrosion behaviors. There is limited information about the structural changes as well as the changes in the elemental distribution at the early oxidation stages of oxide-film growth. In our work, our objective is to develop advanced sample environment where material interactions and behavior at the surfaces and interfaces can be studied in situ under corrosive conditions utilizing synchrotron methods.

Further, Advanced steel alloys offer enhanced resistance to high-temperature oxidation in steam environments due to the formation of protective oxide layers of chromia and/or alumina oxides, which enhance their stability at high temperatures by acting as an effective diffusion barrier, resulting in low rate of oxidation. These alloys present a great opportunity to explore alternative cladding materials with better accident tolerance for commercial light water reactors. The second technical objective of our work was to evaluate the microstructural and chemical evolution of accident

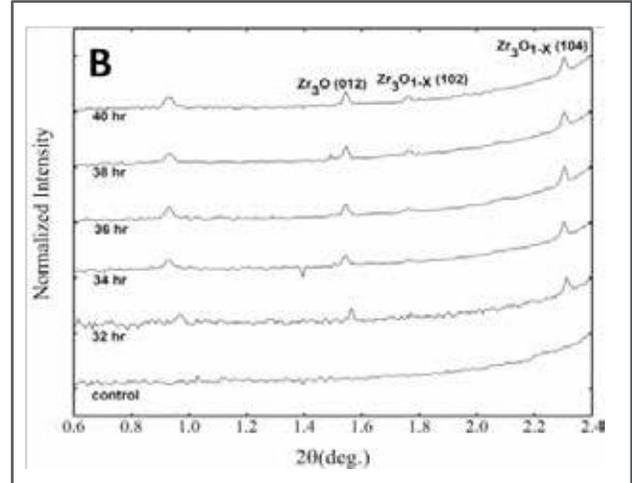
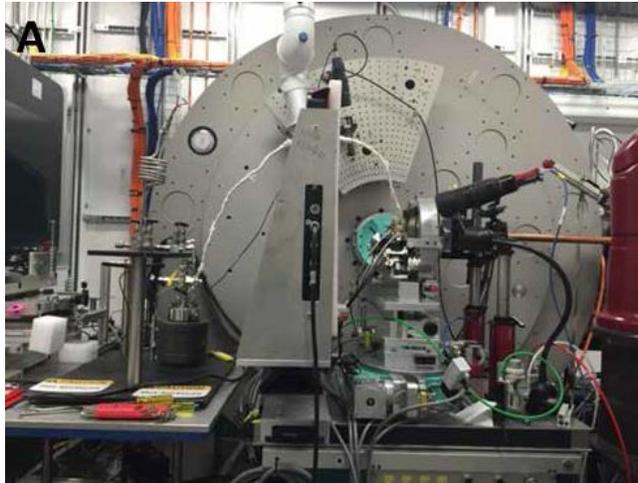


Figure 3. A) In situ sample environment installed at XPD beamline, NSLS II for Reflection Geometry XRD measurements in steam environment, B) XRD patterns of Zr metal exposed to steam inside the in situ sample environment at 350°C and 50 psi up to 40 hours.

tolerant claddings such as Alloy 33 in high temperature environments by utilizing synchrotron methods.

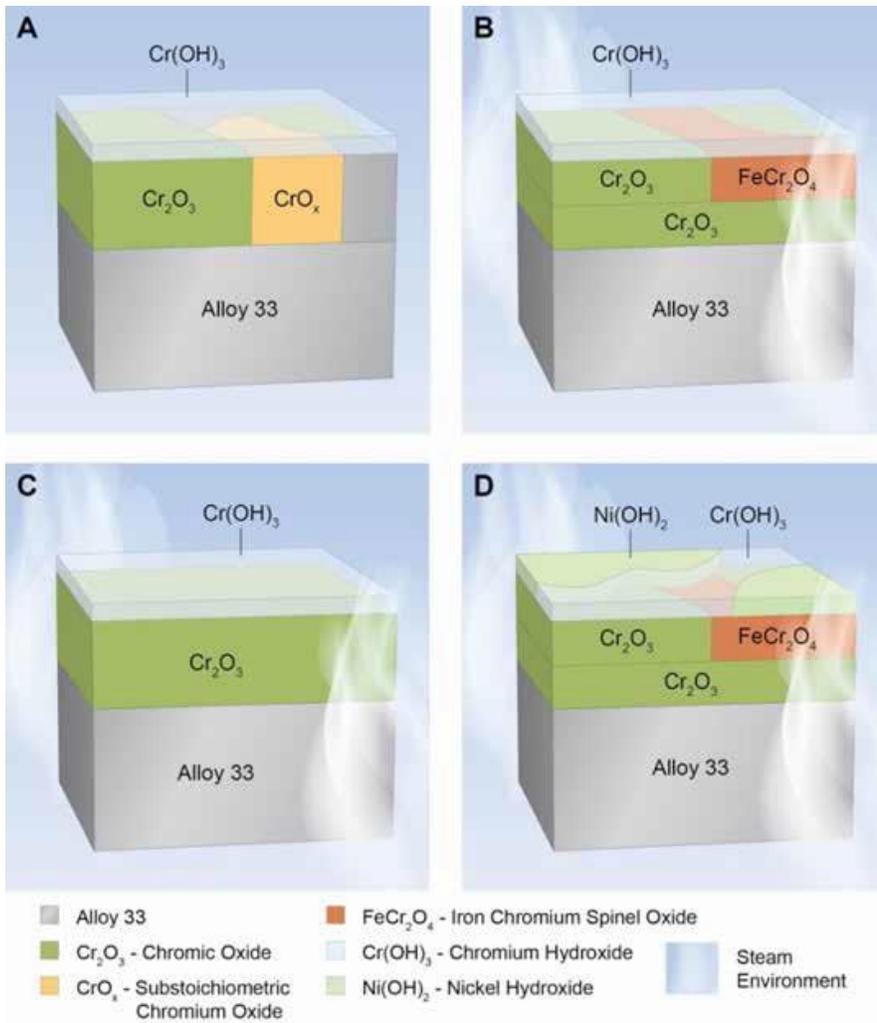
The fundamental and mechanistic understanding provided by high resolution synchrotron structural and chemical data on corrosion processes of nuclear claddings is vital for development of improved and reliable models for predicting the materials performance and lifetime.

**Accomplishments:**

In our work, a novel in situ sample environment for monitoring structural changes and oxide growth on nuclear cladding steam interfaces using high resolution synchrotron methods was designed and built. The sample environment is highly corrosion resistant and optimized for synchrotron X-ray diffraction studies for in situ structural analysis (Figure 1-2). The first results of in situ data collection are presented here as an initial step toward developing a technique capable of in situ studies of variety of zirconium-based and advanced cladding alloys in

corrosive environments. We were able to successfully monitor the substoichiometric oxide phases formed during the initial corrosion of zirconium in steam at 350 C (Figure 3). The in situ sample environment design complies with G2 ASTM standards for studying corrosion. Hence, the data is collected under industry relevant conditions. The design allows for studies of numerous interfacial phenomena including oxidation of thin film, hydride formation, and detection of early oxidation forms. Such information can be gained not only in pure steam environments but also with different additives and under various environments including supercritical carbon dioxide. The ability to couple synchrotron methods for in situ analysis opens up a large number of possibilities for mechanistic corrosion studies.

Mechanistic corrosion studies of advanced FeCrAl alloy (Alloy 33) exposed to high temperature steam were performed using XRD, XPS and XRF methods to understand the



*In situ sample environments coupled with high resolution synchrotron methods offer exceptional opportunity to understand and predict corrosion kinetics and surface oxide composition for nuclear claddings.*

*Figure 4. Schematic illustration of the oxide layer formed on the surface of Alloy 33; (a) control, after steam exposures at 800°C for (b) 8 and 24 hours, and (c) 48 hours, and at 1000°C (d) for 8, 24, and 48 hours*

structural and chemical composition of the surface oxide. Our results demonstrate that a compact and continuous oxide scale was formed consisting of two layers, chromium oxide and spinel phase (FeCr<sub>2</sub>O<sub>4</sub>) oxides, wherein the concentration of the FeCr<sub>2</sub>O<sub>4</sub> phase decreased from the surface to the bulk-oxide interface. Further, the thickness of the oxide followed a parabolic

curve and demonstrated self-protective behavior. Such detailed structural characterizations of surface oxide at Alloy 33 have not been undertaken before and are crucial for fundamental understanding of corrosion kinetics and for deploying advanced steels as alternative cladding materials.

## Status of Thermal Conductivity Microscope Development

Principal Investigator: Dave Hurley

Collaborators: Robert Schley

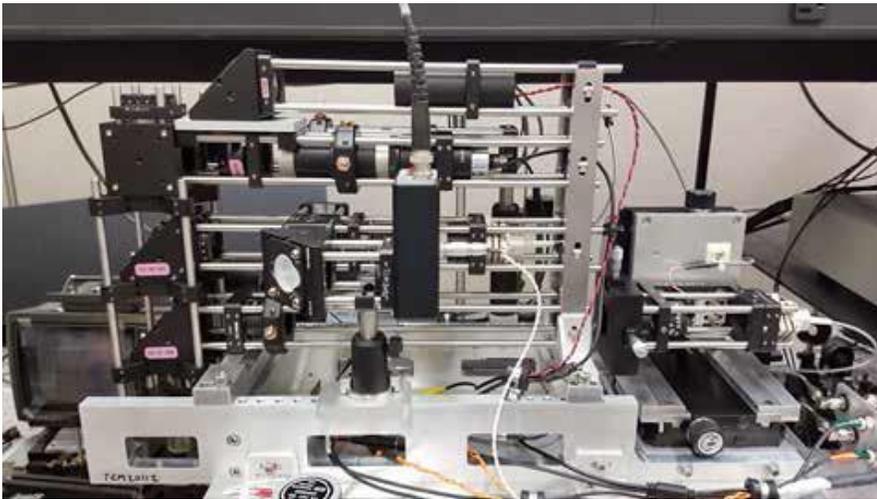


Figure 1. The Thermal Conductivity Microscope under development. This instrument is slated to be installed in the Irradiated Materials Characterization Laboratory.

**The development of the TCM connects closely with INL's larger PIE effort to provide new validation metrics for fundamental computational material science models.**

The function of the fuel in a nuclear reactor is to produce heat through fission. This heat must be transferred through and out of the fuel for eventual energy conversion. With increasing burnup, the thermal transport properties of nuclear fuel degrade due to changes in microstructure brought about by neutron irradiation. The character of the microstructure depends strongly on the local environment and can change drastically over a few millimeters from the fuel element center to the fuel element rim. Assessing the influence of microstructure on thermal transport requires the further development of field deployable instruments that can accurately measure thermal transport characteristics on length scales commensurate with microstructure heterogeneity.

### Project Description:

The Thermal Conductivity Microscope (TCM) is a modulated thermoreflectance instrument developed to directly measure thermal conductivity and thermal diffusivity. The measurement principle is based on using an amplitude modulated laser beam to locally heat a sample. The temperature rise is measured by monitoring small temperature induced changes in reflectivity with a second probe laser. The TCM is being designed to operate in a radiation hot cell environment via remote control manipulation. The TCM provides micron-level thermal property information that is commensurate with microstructure heterogeneity. A picture of the TCM is shown in Fig. 1. This instrument is slated to go into stage one mockup in FY17.

### Accomplishments:

Thermal Transport Measurements in Single Crystal UO<sub>2</sub> - Recently it has been reported that thermal transport in single crystal UO<sub>2</sub> is anisotropic (doi:10.1038/ncomms5551). To verify this finding, we used the Thermal Conductivity Microscope (TCM) to characterize 3D heat flow in single crystal UO<sub>2</sub>. The sample was grown at the Air Force Research Laboratory by Dr. Mathew Mann using the hydrothermal growth method. The measured lattice constant of  $a=5.475$  angstroms indicates that the sample is stoichiometric. The measured conductivity along the [011] direction [001] directions is 7.8 and 7.7 W/m\_K respectively. A total of

20 measurements were taken in both directions and the standard deviation in both directions is (0.4 W/m\_K). These results are significant for two reasons. First, using the TCM to measure local thermal properties circumvents having to consider sample to sample variations. This is not the case for bulk property measurements that require preparing different samples with specific orientations. Second, the conductivity ratio measured using the TCM of 1.01 is much smaller than the ratio of 1.13 reported in doi:10.1038/ncomms5551.

Spatially resolved measurement of thermal transport in multiphase materials - We mapped the thermal properties of a U<sub>3</sub>Si<sub>2</sub> fuel surrogate sample using the Thermal Conductivity Microscope (TCM). The TCM was used in the time-domain mode in order to spatially resolve the thermal properties of small (5-10 μm) 2nd phase precipitates. This approach uses a coaxial pump-probe arrangement and involves recording the time domain thermoreflectance response due to square wave laser excitation. Figure 2 shows typical reflectivity transients observed from two different locations on the surface of the sample. An SEM micrograph, shown in the inset, reveals the presence of second phase precipitates believed to be uranium oxide. The thermal response is similar to the behavior observed when a square wave is sent through a low pass filter. In this configuration, materials with higher thermal conductivity will

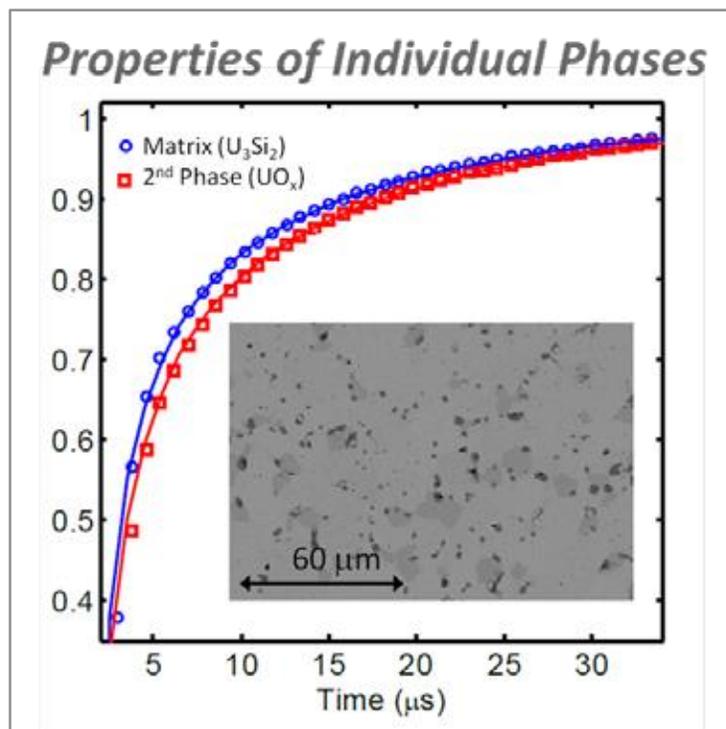


exhibit a shorter time constant. The response of the matrix has a faster response and consequently has a higher thermal conductivity than the second phase precipitate. This observation suggests that the emergence of irradiation induced 2nd phase precipitates in silicide fuel may have a significant impact on thermal conductivity. A simple model of the thermal response (solid line) was constructed using a summation of thermal waves at multiples of the excitation frequency.

Figure 2. Typical reflectivity transients observed from two different locations on the surface of the sample. An SEM micrograph, shown in the inset, reveals the presence of second phase precipitates believed to be uranium oxide.





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# APPENDIX

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- 1.1 The Advanced Fuels Campaign Team
- 1.2 From the Director (Campaign Overview)
- 1.3 International Collaborations