2014 | ANNUAL REPORT

Nuclear Science User Facilities

NSUF Nuclear Science User Facilities

Nuclear Science User Facilities
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FROM THE NSUF DIRECTOR

J. Rory Kennedy
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This marks my first annual report letter to you as director of the Advanced Test Reactor National Scientific User Facility. It is also the last letter I will write to you as director of the Advanced Test Reactor National Scientific User Facility (ATR NSUF).

Later this year, the official name of the ATR NSUF will change to Nuclear Science User Facilities. This new designation more accurately reflects just how much the entire public-sector nuclear research effort in the United States has grown and matured. While our former name has served us well since the user facility was established in 2007, it referred to only the test reactor and its associated facilities at Idaho National Laboratory (INL).

Our new designation reflects what we are today: a network of facilities throughout the United States that are available to researchers for the study and advancement of nuclear science and engineering.

Today, our 11 partner facilities—including the Westinghouse Materials Center of Excellence, our first member from the private sector—provide their cutting-edge equipment and scientific expertise to researchers seeking to develop nuclear energy as a safe and reliable resource.

As exciting as our name change is and what it represents, there are a number of other tasks the ATR NSUF has undertaken or will undertake either under our own initiative or at the behest of the U.S. Department of Energy (DOE). They include project-forward funding, in which an entire research project is funded at the time it is first approved; consolidating our calls for proposals into the Consolidated Innovative Nuclear Research (CINR) Funding Opportunity Announcement (FOA), which includes those of the Nuclear Energy University Programs (NEUP) and Nuclear Energy Enabling Technologies (NEET), to give users the opportunity to apply for R&D and NSUF no-cost access in one application; opening project principal investigators to others besides just university researchers, including those from industry, national laboratories and small businesses; increasing our partner facilities interaction and promotion by simulcasting the Users Meeting to and from all the partner facilities as well as updating our conference and meeting exhibits; continuing to enhance the sample library of irradiated materials; and establishing a searchable database of all Nuclear Energy (NE)-accessible capabilities available to users through the partner program to increase utilization efficiencies. We envision the sample library and capabilities database will become invaluable references for both NE and researchers in the future.

These are all exciting endeavors intended to better support our users and advance nuclear research, not only for the United States but the entire world. And while I will help lead the effort to fulfill them going forward, the foundation for their implementation has been laid by my predecessors: Dr. Mitch Meyer, Professor Todd Allen, Professor Jeff Terry, and Frances Marshall, who served as interim director during my transition. Their innovative thinking, hard work, and dedication to the job and to the ATR NSUF have enhanced the organization, the partnership program, and indeed, the entire nuclear industry further than we could have anticipated. I am grateful to them and hope I can continue the impressive legacy they have established.

Sincerely,

Dr. J. Rory Kennedy
Director


PLE PREPARATION
HOT PLATE
PUL TOR TRAINING

RESEARCHER PROFILES

Mahima Gupta

After graduating from high school in her native India in 2006, Mahima Gupta moved to the U.S. to attend the University of Michigan with every intention of studying genetic engineering. But even the best plans often don't go as expected.

"I was always interested in physics," says Gupta, "so I was taking an honors class. My teacher was this crazy German guy, very intelligent, and his class just blew my mind."

She spent the following summer working with that professor on one of the first programs in the U.S. investigating Bose-Einstein condensates. When she resumed her undergraduate studies in the fall, her advisor suggested she look into nuclear engineering.

"It was instant love," Gupta says. "I loved that you could use nuclear power to create carbon-free electricity. It was clean, renewable—the whole deal."

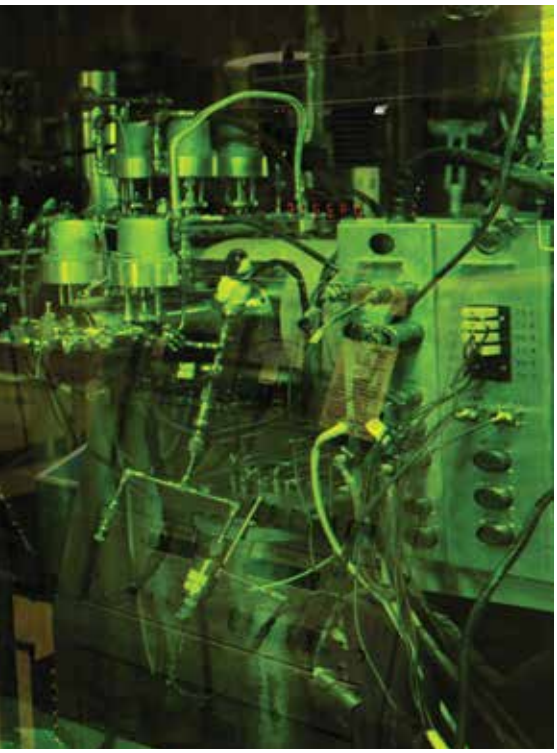
During her undergraduate time at Michigan she served as president of the local American Nuclear Society chapter. At one of the society's conferences she met Dr. Todd Allen. She mentioned she wasn't sure where she wanted to go to graduate school and he asked what she wanted to study. When she said fuels, he suggested she come to the University of Wisconsin – Madison (UW) and work with him. She agreed, and received her Ph.D. in April 2015, after spending the last two years finishing her thesis at INL.

"I was finished with all my classes in the summer of 2013," says Gupta, "so I moved to Idaho. I worked at the Materials and Fuels Complex (MFC) for a year and then started an internship with ATR NSUF. It's been a great experience."

She wasted no time taking advantage of the NSUF's proposal process. With two proposals already accepted, the first of which is included in this annual report, and a third submitted, she is already making her mark as an up-and-coming researcher. The initial proposal, a first-of-its-kind experiment, examined the effects of irradiation on uranium-dioxide (UO_2) at the atomic level.

"Most studies on irradiation damage in UO_2 are usually done on a larger scale," explains Gupta. "We're looking at angstrom and sub-angstrom scales, so the physics is totally different. One of my goals is to see if there is any relationship between these really tiny scales and slightly larger scales."

She spent the first two years of the project irradiating her samples in the Ion Beam Laboratory at UW, one of the NSUF's partner institutions. It was one of the first ion accelerator irradiations completed on UO_2 at the UW lab. She also helped write the protocols for those irradiations and, as a result, UW now has a preparation facility that can handle irradiated



"You get results right away, with state-of-the-art instruments, and with the full support of the CAES staff. It's a great way to work on your Ph.D."

samples for future research. Next, the samples were prepared and examined at CAES using a focused ion beam (FIB) and transmission electron microscopy (TEM).

"One thing we're seeing is that oxygen defect clustering increased with ion beam irradiation on a larger scale that is seen in TEM micrographs," says Gupta. "This leads us to believe that there is a whole different class of defect clusters, larger than individual point defects but smaller than what can be detected in the TEM."

To explore that possibility, a systematic series of experiments was conducted utilizing the Stanford Synchrotron Radiation Laboratory (SSRL). Samples were studied using X-ray absorption fine-structure spectroscopy (XAFS) to see how the atomic structure changes in UO_2 after irradiation. After creating minimally radioactive samples using ion accelerators, Gupta utilized a technique called micro-focusing XAFS. It was the first time the process was used on irradiated specimens. Using the instruments at CAES, she and her colleagues picked out samples about 10 x 15 microns in size and sent them to SSRL. By micro-focusing the X-ray beam, another first for nuclear fuel samples, they were able to raster a micron-sized beam across the samples to find the UO_2 signals and perform the XAFS analysis.

Gupta is quick to point out the advantages of these rapid turnaround experiments (RTEs).

"I submitted the proposal right when I started my internship at INL," she says. "It was approved a couple of months later and we were able to start work instantly. You get results right away, with state-of-the-art instruments, and with the full support of the CAES staff. It's a great way to work on your Ph.D."

Having finished work on the second proposal, Gupta is poised to start the third phase of her project, but now that she's graduated, a job search has taken priority.

"I'm trying to figure out how to make the transition. I can still continue my work here, but I feel like if there were other Ph.D. students who were interested, someone could write their entire thesis on these samples."

Isabella van Rooyen

After five years as an early career researcher at the South Africa Nuclear Energy Corporation, Dr. Isabella van Rooyen went to work on her country's pebble bed modular reactor program. When the government withdrew funding from that program in 2011, she decided the U.S. offered the best opportunities for her to pursue her passion for studying nuclear materials, particularly tristructural isotropic (TRISO)-coated fuel particles. Partly because of its involvement with the Next Generation Nuclear Plant (NGNP) program, and partly because of her broad, respected background in the nuclear field, INL welcomed her with open arms.

"I had also gained a lot of experience working in the industrial sector," van Rooyen says. "That turned out to be a big advantage, because now I not only think about what I'm working on at that moment, I think about what the application of that product will be for the end user."

When she first came to INL, she split her time between studying the viability of TRISO-coated particles and working on the light water reactor (LWR) sustainability program. Her success with TRISO fuels soon won out, and this year she and her colleagues, Dr. Thomas Lillo and Dr. Yaqiao Wu, received an INL Outstanding Paper award for "Identification of Silver and Palladium in Irradiated TRISO-Coated Particles," which was published in the Journal of Nuclear Materials in 2014.

"I had started trying to solve the silver transport mechanism problem in South Africa," says van Rooyen, "and I learned a lot there, but I could never go to the next stage because we couldn't study irradiated particles."

That hurdle was overcome at INL, and resulted in a breakthrough discovery.

"We always knew from gamma spectrometry that under irradiation silver is released outside these tiny, silicon-carbide-coated particles," van Rooyen explains," but we never knew where and how. Now that we know the path it follows, we can work toward a solution to prevent that release from happening."

While they have not yet tracked the full mechanism, they feel very close to gaining a new set of knowledge.

"Using transmission electron microcopy, we are currently looking systematically at the orientation and grain boundary characteristics of each grain that is adjacent to the silver we have found," says van Rooyen. "Once we find the grain characteristics we can manipulate the release process in such a way that does not favor the driving transport mechanisms."

"So by funding this research, the NSUF has not only helped me, along with other researchers and INL, it is also helping to build the next generation of researchers. I hope we can continue that as we move forward."

The question of silver transport has been a nagging one for the Nuclear Regulatory Commission because of its impact on the licensing of high-temperature gas reactors. Being able to change the microstructure of the silicon-carbide layer will be a huge step forward. Van Rooyen's study has also provided valuable information on a question that researchers in the LWR accident-tolerant fuel program have been longing to answer in their investigation of TRISO-coated particles as a potential fuel for that program.

Van Rooyen is quick to point out that their discovery was made possible by the excellent microscopes available at Center for Advanced Energy Studies (CAES), as well as the excellent scientists who operate them.

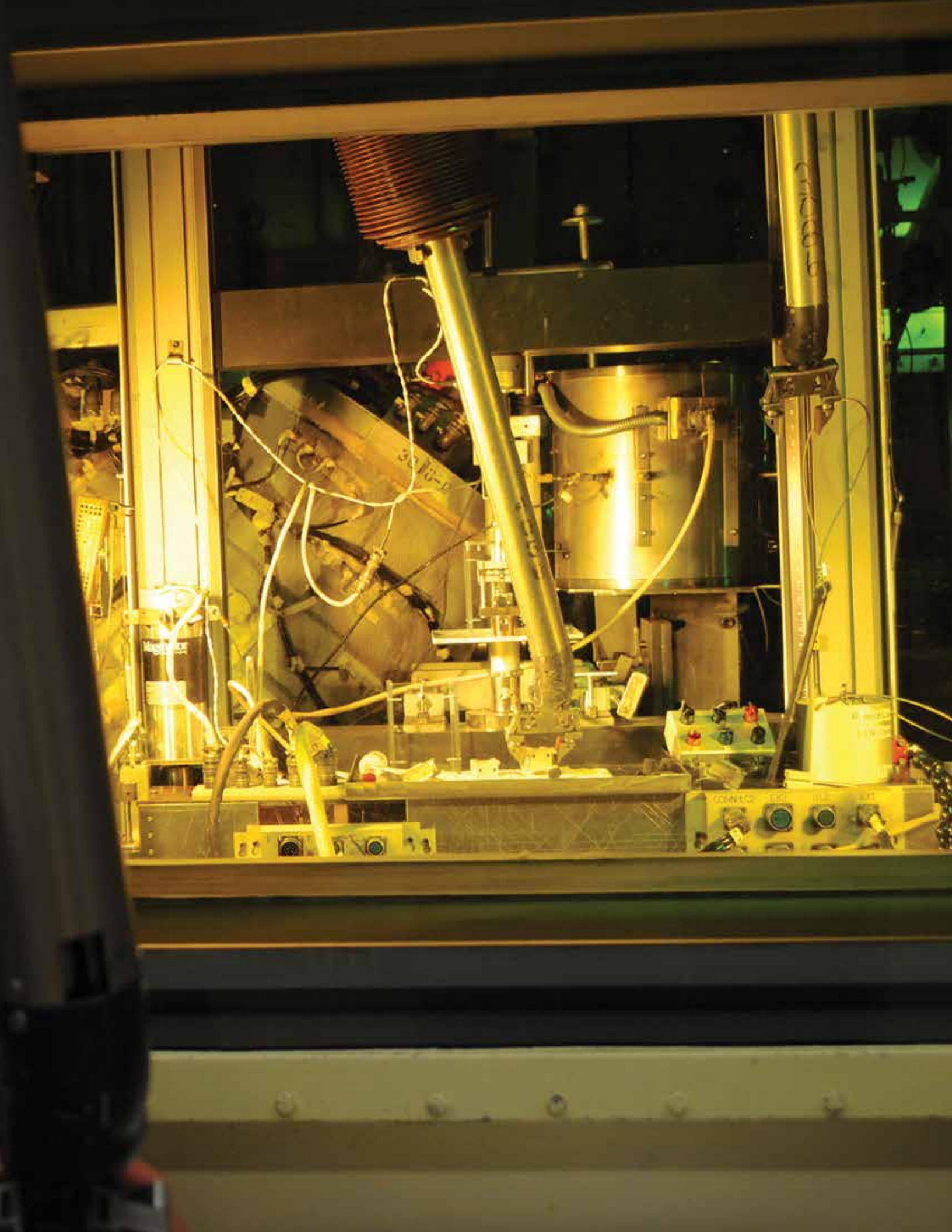
"This project was really innovative on the part of our whole team," van Rooyen said. "Many people said we couldn't accomplish it because of the high radioactivity in the parent material, but we persisted. It shows other researchers that they must not stop. It is no longer an excuse for nuclear material scientists to say they don't have the availability of equipment. Because of the brilliant partnership program at ATR NSUF, we can all use the newest stuff."

Already scientists as far away as the United Kingdom, France, China and South Korea have begun using the nanostructural and microstructural information on silicon-carbide provided by this breakthrough study to pursue further research.

"Every time I present my work at a conference I realize the impact the NSUF has had in my role as a scientist here at INL," said van Rooyen. "One of my presentations at User's Week two years ago stimulated collaboration with a university and I became a part of their research team. Helping us to build our networks is one of the missions of the NSUF, and I can truly say I am living proof that it is working."

Those networks offer benefits beyond any one particular project.

"One of the students that worked with us as an intern on this latest project is now applying for a permanent position at INL," van Rooyen said says with satisfaction. "So by funding this research, the NSUF has not only helped me along with other researchers and INL, it is also helping to build the next generation of researchers. I hope we can continue that as we move forward."



Janelle Wharry

Every student has that fork-in-the-road moment that defines the rest of their life, and more often than not, it's a good teacher who provides it. Boise State University Assistant Professor Janelle Wharry always wanted to be an engineer, she just didn't know what kind. The summer after her junior year in high school she left home in Hawaii to attend a two-week program at the University of Illinois, and all that changed.

"It was like a camp," said Wharry "They had kids from all over, and we all got to learn a little bit about the different engineering majors the university offered. They had a nuclear engineering program that really got me interested in the medical applications, especially cancer research."

She ended up going to the University of Michigan (UM), where her focus shifted to nuclear energy, and finished her master's degree in 2005. She didn't really know where she wanted to go from there, so she took a job with Duke Energy in North Carolina for two years as a core design engineer.

"What I learned there was that the neutronics of a nuclear system are reliable and very well understood, but the main issues we were always having with the plants were the effects of radiation on structural materials. So I decided that's where I would focus my thesis."

She went back to UM and received her Ph.D. in 2012. While doing a six-month postdoctoral study there she started looking for a job. She knew academia was where she wanted to be, and after all the moving around she had done over the years she just wanted to settle down somewhere and make a life for herself. When Boise State made her an offer it seemed like the perfect fit.

"I love it here," Wharry said. "My background is mostly irradiation effects in materials, but since I came here and started my own program I've expanded into metallurgy and mechanical properties. The main focus of my research group is to study how mechanical properties evolve in correlation with microstructure under irradiation."

Materials science and engineering is a relatively new area of study at Boise State. The undergraduate program began barely 10 years ago, and shortly after that, with the help of Micron Technology, a master's curriculum was added. In 2012, thanks to a $13 million grant from the Micron Foundation, a Ph.D. program was instituted. The first year just over 20 students were seeking their doctorates, and today there are more than twice that number, along with 50 undergraduates and more than 40 master's students.

Boise State is not yet an NSUF partner, but along with the University of Idaho and Idaho State University, it is a partner in the state-funded CAES.

"One of my long-term goals for BSU is to establish a partnership with ATR NSUF as well," Janelle says. "We're in the process of buying some pretty cool equipment for in-situ mechanical testing experiments in the transmission electron microscope (TEM) that I'm hoping will bring a unique capability to CAES. Hopefully that will be a major step toward that goal."

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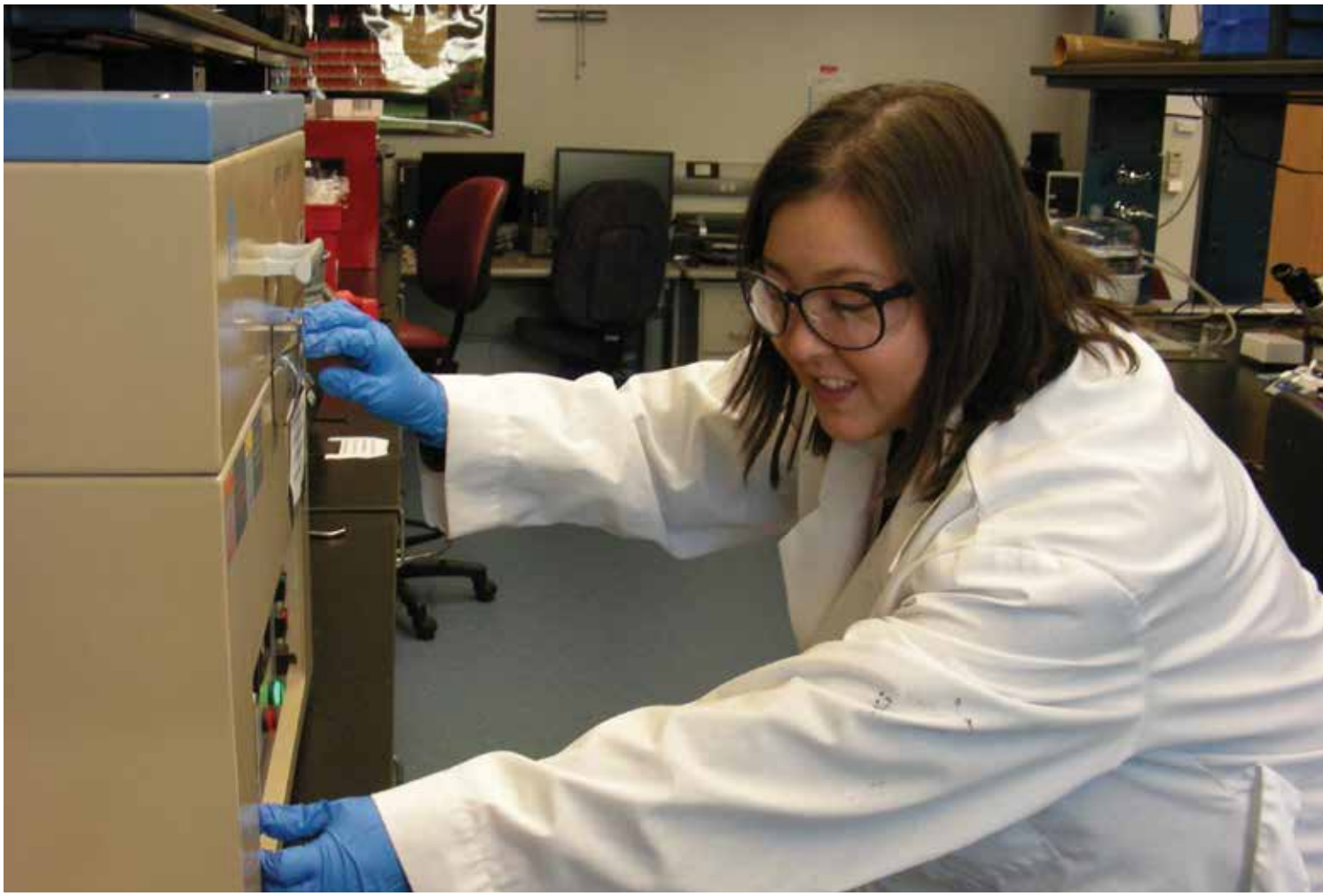


Adding a Hysitron PI-95 picoindenter and new thermal-aging capabilities to her BSU laboratory this year will be a big help in one of her main research efforts: looking at the role solid-solution strengthening plays in the mechanical strength of advanced ferritic-martensitic steels and oxide-strengthened dispersion steels.

"My real interest is in trying to improve steels," says Janelle, "not just in irradiation environments, but in general. Can we harness properties that are desirable for radiation tolerance and use them to develop better steels in nonirradiation environments? I think it's important to broaden our horizons and look at what people are developing for non-nuclear applications as well, and how we can use those advances in the nuclear environment."

One National Science Foundation project she's currently working on is a prime example of that theory.

"We're looking at titanium-dioxide (TiO_2) as a potential lithium-ion battery anode material," explains Wharry. "Irradiation creates a high concentration of defects in a material, which we believe will provide more space for lithium ions to intercalate. So if we can harness the power of irradiation to create vacancies in the TiO_2 we may be able to improve the functionality and efficiency of batteries. Irradiation is always talked about as damage. I guess it's the optimist in me, but I don't believe it can be 100 percent bad. There has to be something useful for everyday life that can come from irradiating materials."

A PASSION FOR DIVERSITY
PNNL

Collaboration and teamwork are the keys to achieving success in almost any endeavor, and in scientific research and development, they're critical. This spirit typifies research at INL, and is, in turn, carried out to other nuclear research institutions through a distributed partnership program overseen by NSUF. Begun in 2009, the program now comprises 10 national laboratories and universities, along with one industry partner: Westinghouse. One of the first to join this comprehensive national network of scientific research capabilities was Pacific Northwest National Laboratory (PNNL), in Richland, Washington.

Since its inception, PNNL has been operated by the Battelle Memorial Institute. It is one of five U.S. laboratories stewarded by DOE's Office of Science, but it serves all of DOE's missions: science, energy, environment and national security. As such, the passion of its scientists and researchers for innovative solutions to problems faced by the U.S., as well as other countries, extends to the U.S. Department of Homeland Security, the National Nuclear Security Administration, and a host of other government agencies, universities and industry customers.

It was born in 1965 as the Pacific Northwest Laboratory (PNL) to conduct research and development for the nearby Hanford nuclear site. There, its scientists and engineers designed and conducted research at the Fast Flux Test Facility, testing fuels and materials for the Liquid Metal Fast Breeder Reactor program to expand the nation's nuclear energy capabilities. Research

soon expanded into areas as diverse as holography and heart transplants, cybersecurity and environmental studies. PNNL (the second N, for National, was added in 1995) researchers pioneered compact disc technology, invented the first portable blood irradiator for leukemia treatments, and were chosen to help study lunar materials retrieved during National Aeronautics and Space Administration's (NASA) Apollo program, to name just a few from a long list of scientific accomplishments. Today, more than 4,300 scientists, engineers and nontechnical staff are responsible for a legacy that includes 2,342 patents, 104 of which were awarded in 2014 alone, and a total of 81 Federal Laboratory Consortium awards for technology transfer since 1984.

The lab is divided into four major technical directorates: Energy and Environment, National Security, Fundamental and Computational Sciences, and the Environmental Molecular Sciences Laboratory. Two major clients, the Nuclear Regulatory Commission (NRC) and DOE's Office of Nuclear Energy (NE), of which NSUF is a part, comprise the core of PNNL's work in nuclear energy, while a third area, nonconventional, helps keep the U.S. engaged in commercial as well as international programs and gathers insight from those collaborations.

"We have a very broad portfolio of science and technology interests," says Steve Unwin, PNNL's Nuclear Market Sector Manager. "Just looking within those three big blocks in nuclear power we tend to be very eclectic. For example, we're heavily involved in helping implement the new generation of risk-informed regulations, and we were very proud to have led the environmental reviews on behalf of the NRC for the first two plants to receive construction permits in the U.S. in over 30 years, in South Carolina and Georgia. We've also been a leader in materials science and nondestructive examination, a very important field given the aging fleet of reactors that must be kept vital to meet the nation's energy needs."

On the NE side, the lab's biggest effort goes toward fuel cycle research and development. In addition to a long history of expertise in spent fuel qualification and being involved in used fuel reprocessing research, its scientists are also studying new fuels that are more accident-tolerant and more economically attractive.

"NE is looking at the next generation of reactors," Unwin says, "and we've been developing technologies that will help make them safe and economically attractive. Also, small modular light water reactors are showing great promise as a viable solution to energy needs, and we helped the NRC develop the regulatory structure to license them."

Part of the value of all this expertise is the creative thinking that goes along with it, and the experience accumulated through the years—50 years to be exact. In 2015, PNNL celebrates its Golden Anniversary, a milestone that brings both the past and the future into sharper focus.

"I don't think we have fulfilled our potential in the nuclear arena yet," says Unwin. "Our vision is to contribute to that potential by ensuring the success of our clients' missions as well as that of the nation, and we want to bring all our ideas, innovations and technical expertise to the table."

Another area where PNNL scientists have particular depth is working towards a nuclear spent fuel repository.

"We have the right types of expertise to help streamline that process in areas such as waste form qualification and risk assessment," Unwin says. "We do a lot of work, for example, around ensuring the integrity of spent fuel systems from storage and transportation to final disposition."

Advanced reactor technology is another, equally important part of the diversity of research that is a hallmark of PNNL.

"The beauty of some of these advanced reactor designs is that they actually generate their own fuel," Unwin explains, "so we can minimize both the consumption of fuel resources and the creation of waste. The data and expertise we gained from the operation of the Hanford Fast Flux Test Facility is allowing us to bring valuable insight to the new generation of reactors. All the analysis shows is that if we're going to meet national energy needs, it really has to be an all-of-the-above policy, of which nuclear power—clean, reliable, and carbon-free—is a critical component. At the end of the day it's going to be the commercial sector building these reactors, so our work with industrial clients opens up a conduit for new technology developed by NE."

Remaining at the forefront of all this cutting-edge research requires state-of-the-art equipment, and that, in turn, requires investing heavily in the future of the nuclear industry.

"I would say that stock in the nuclear portfolio is higher than it has been in recent years," says Steve Schlahta, director of the Nuclear Science Project Management Office at PNNL. "We're committed to a significant internal investment leveraging our materials science, specifically our spectroscopy capabilities, which we feel are hallmarks in this area. We recently invested over $2 million in a JEOL aberration-corrected transmission electron microscope, one of the few in the world. We're also putting a FIB in the Radiochemical Processing Lab (RPL), as well as several other instruments, and adapting a 30-ton INSTRON load frame to work inside a shielded hot cell, all of which make the RPL one of the gems of the Office of Science stable."

In the past the RPL's mission was largely Hanford-related. Today, PNNL management assesses it as a billion-dollar asset. And in order to make sure it remains such a vital part of their portfolio, it's important to make sure the lab is being used. One of the ways they do that is by taking part in NSUF's partnership program. Staff Engineer David Senor has been PNNL's chief liaison with the NSUF over the past five years, and will complete his term as chair of the Users Organization, the group that oversees that program, in June 2015.

"It's tough to pin down PNNL's specialties because we're so diverse," says Senor, "but I think the biggest benefit we bring to NSUF is extra post-irradiation examination (PIE) capacity. If you look at these types of experiments, the bottleneck usually comes after the irradiation is done. So the more PIE capacity a user facility can put together the more responsive they can be to the PI's in getting the results of irradiation experiments out in a timely fashion. Another major piece of that is our irradiation experiment design and fabrication capability. Very few places in the country retain that expertise these days. So, besides playing a role in preparing for PIE, if there are limitations at the user facility on the front end of irradiation experiments, we can help alleviate a log jam there as well."

"We're pretty nimble facility-wise, too," adds Schlahta. "We have a lot of flexibility with what we can put into our modular hot cells, especially with the new INSTRON load frame, and that's not the case with some of the other NSUF partners."

On the other side of the equation, Schlahta agrees that being an NSUF partner also helps PNNL.

"I think this is a great example of how the DOE system should work," he says. "We have some very specific capabilities that give us an entrée, and partnering with NSUF gives us some excellent opportunities to actually demonstrate what we can do."

One such opportunity currently on PNNL's PIE plate is a fuel project headed by Dr. Mehdi Balooch at the University of California, Berkeley (UCB). The objective is to develop a hydride fuel that would potentially replace the uranium-dioxide (UO_2) fuel currently used in LWRs, and to explore the use of a liquid metal as a replacement for helium to fill the pellet cladding gap.

"The problem with UO_2 is that it has a low conductivity," explains Balooch, "so if you want to get the most energy out of it you have to go close to 2,000°C. The hydride fuel we are proposing has a conductivity five times higher than UO_2, so in order to get the same amount of energy out of it you only have to go up to around 600°C. It also has better properties in terms of neutronics and safety."

Beginning in 2010, the fuel elements for the experiment were made at UCB. They were put into the Massachusetts Institute of Technology (MIT) reactor in 2011, and in the spring of 2014 three capsules of irradiated samples were sent to PNNL. All of these institutions are NSUF partner facilities. Only one capsule has been disassembled for PIE. The other two will be shipped to INL's sample library where all that material will become available to other researchers. The multifacility track followed by this experiment is a perfect example of the kind of collaboration fostered by NSUF's partnership program.

"We received the capsules at PNNL in March 2014," Senor said, "then we had to build the fixturing and figure out the best way to take them apart. Disassembly was done in the fall and now we've started cutting up the samples and getting into the examination phase."

Experiment Manager Andy Casella organized groups of PNNL scientists with specific areas of expertise to examine the samples.

"Mehdi has a list of questions he would like to get answered with these samples," said Casella, "so we're trying to figure out the best way to use the facilities we have to generate those results. That way we can tie the beginning of the experiment to the conclusion so it can be presented in a way that is easily digested."

"This is not the end of the story," said Balooch. "We're hoping to get to the point where a decision can be made as to whether it's worth it to continue this research or not, and 90 percent of that takes place here, in the PIE work. It's

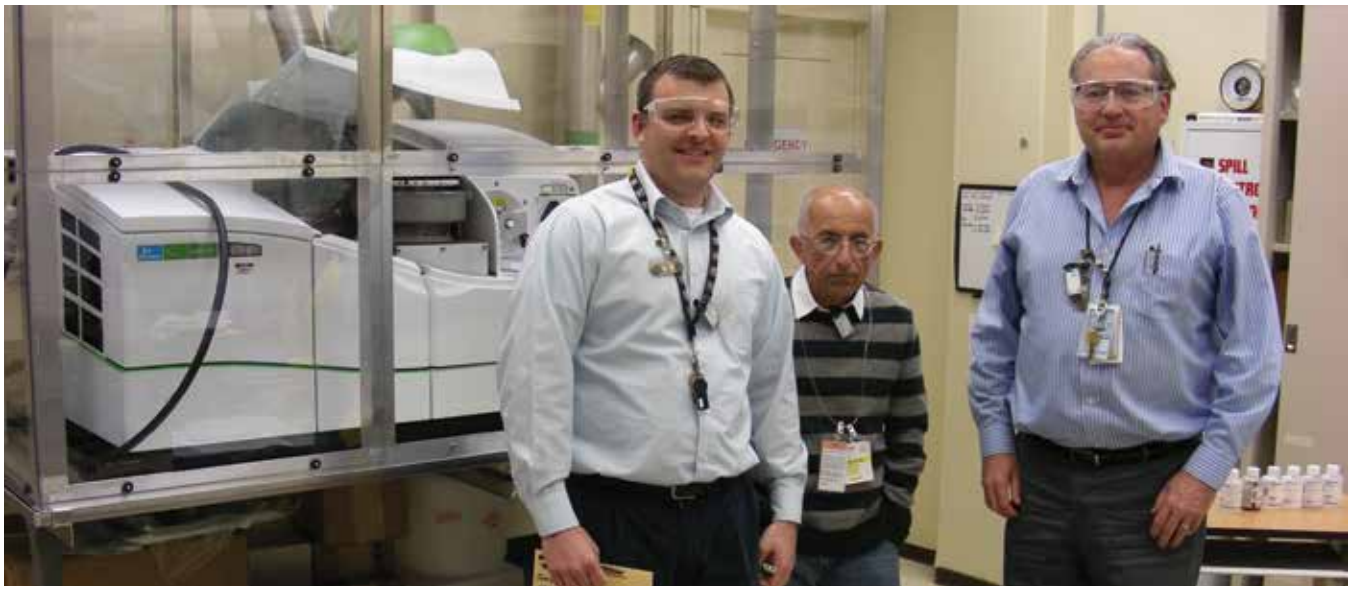
not going to answer all of the questions as to whether we can replace the UO_2 with this new fuel, that's just not going to happen with this small project and small budget."

"And in the course of answering these initial questions," Senor said, "we've already started to generate a whole bunch of new questions, which will lay the foundation for follow-on studies if someone decides to take this to the next step. To introduce a new fuel or a new cladding material you're looking at a 15 to 20 year time frame to accomplish all the development and testing that's going to be required to license that new material with NRC. This project is really just step one of that process."

Step two, according to Balooch and the PNNL team, would likely be further irradiation testing, perhaps on slightly larger rodlets, and at higher temperatures. But first, they'll collaborate on a series of papers and reports outlining all the data collected from this experiment. From there, it's up to someone else to pick up the ball and run with it, perhaps through an NSUF proposal that would utilize the unique capabilities and expertise available through its partnership network.

"I graduated from MIT," said Balooch, "but I never thought that at some point MIT and UCB would work hand-in-hand together. This is one project where it actually happened. UCB got the ball rolling, MIT got involved, PNNL is involved, INL is involved, and one big advantage is that a lot of graduate students have benefitted from it. They come together, work together, and that has helped this project quite a bit. It's a great idea."

"There are really very limited opportunities for professors and their students to interact with a test reactor and to do an experiment and follow it all the way through," Senor said. "The NSUF creates those opportunities, and we're proud to be one of their partners."



SCIENTIFIC REVIEW BOARD

A Guiding Hand for a Maturing Program

Every year during the summer, nine distinguished nuclear scientists and engineers, each of them an expert in a particular field of nuclear science or education, gather at the CAES in Idaho Falls, Idaho. Together, they comprise the Scientific Review Board (SRB) of what, until recently, was known as the Advanced Test Reactor National Scientific User Facility (ATR NSUF).

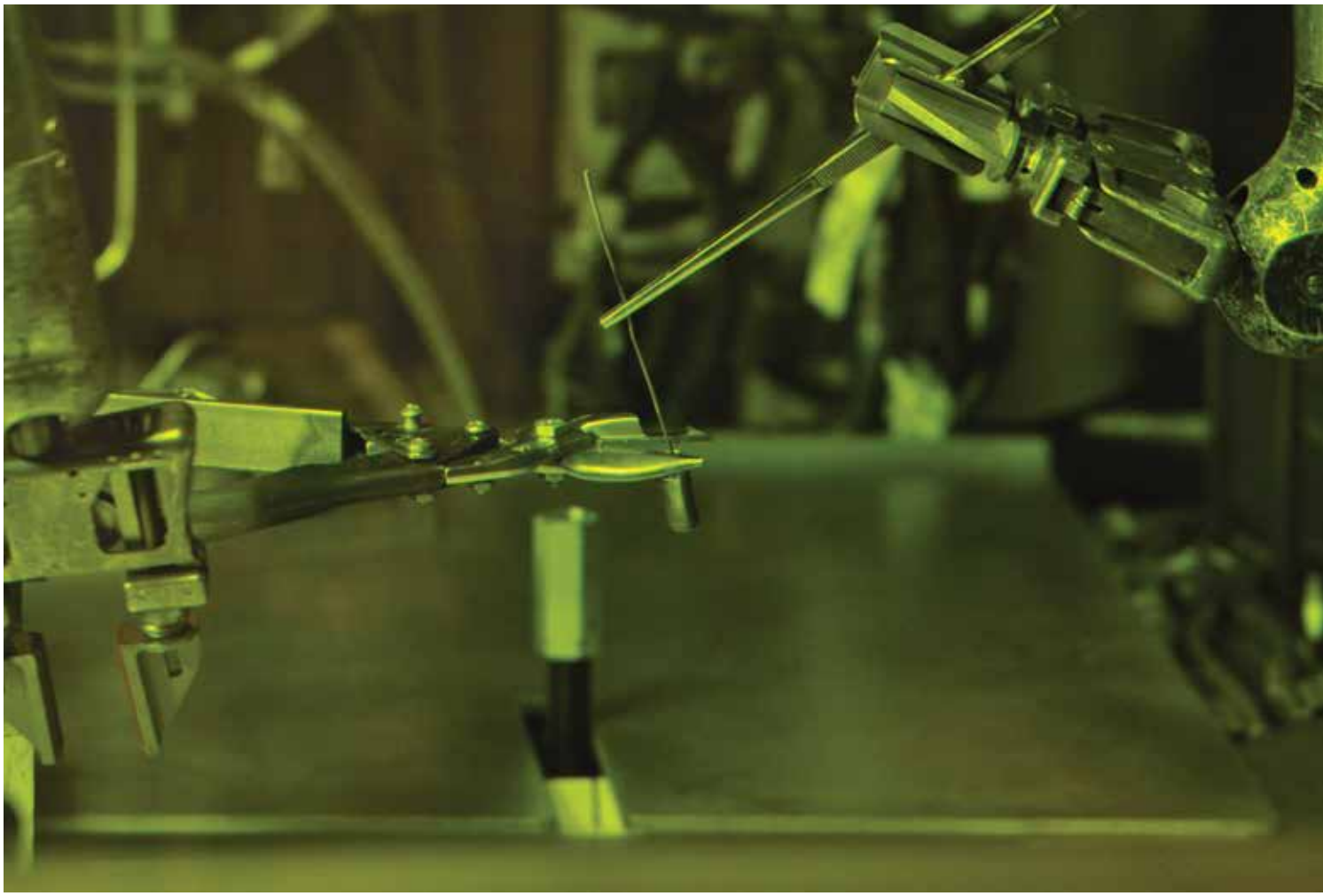
Dr. John Sackett is a former associate laboratory director from Argonne National Laboratory. Now retired, he acts as the SRB's chairman. "The NSUF mission," he says, "is to support the advancement of nuclear science and technology in the United States by providing nuclear energy researchers access to world-class testing facilities. The SRB is an advisory group that helps the NSUF fulfill that mission in the best way possible."

"The board brings together experts in customer service, materials, irradiation, post-irradiation examination, and fuels, who want to make the NSUF as useful to its users as possible," explains Dr. Sean O'Kelly, who until recently served as deputy director of the National Institute of Standards and Technology (NIST) Center for Neutron Research (NCNR) in Gaithersburg, Maryland.

O'Kelly is a good example of one of the experts he refers to. The NCNR has a users program that is considered one of the best in the world due to the reliability of its facility and the high-quality support it provides users. "The users the ATR NSUF wants to attract," he points out, "are similar to the users the NIST reactor provides its own services to, so I can offer insights on what's been successful for us."

As part of his duties in the NIST users program, O'Kelly served as chief of reactor operations and engineering, so he also brought a deep understanding of reactors and how they operate to the board. "Basically, I kept the NIST reactor operating efficiently for the user's program, which used the reactor as a source of neutrons for experiments. We maintained and operated the reactor at NIST 24 hours a day and the NCNR has a very good reputation for the reliability of operations and the predictability of schedule, which users appreciate when they travel long distances to perform experiments."

This past February, O'Kelly accepted a position with INL as associate laboratory director for the Advanced Test Reactor, meaning he is no longer an outside expert, so he has relinquished his seat on the board. "Sean's input



on what makes a very good users program even better was invaluable to us," said Sackett. "He will be hard to replace."

On the other hand, Andrew Klein, professor of nuclear engineering and health sciences at Oregon State University (OSU) is not going anywhere. He brings to the SRB a unique perspective on the world of higher education, especially with regard to one of the NSUF's primary user groups, university researchers and their graduate-level nuclear science and materials students. Take a quick look at his curriculum vitae and you'll see he's the head of the Department of Nuclear Engineering and Radiation Health Physics at OSU, the director of educational partnerships at INL, and a member of the board of directors for the Foundation for Nuclear Studies.

He also brings a certain amount of self-interest to his position. "I'm a university researcher myself, so I'm a potential user of the NSUF," he explains. "I have a vested interest in being the champion of its 'customers' and finding ways that the program can serve them as completely as possible."

To that end, while the members of the SRB support the NSUF and the role it plays in developing nuclear energy as a safe, reliable source of energy, they're not cheerleaders. "We're an independent advisory board," says O'Kelly, "so we do criticize. We don't hold anything back."

"Our recommendations are only that—recommendations," Sackett said. "We don't have the authority to require the NSUF to take any specific action."

But those recommendations do carry the weight of a higher authority, as the SRB actually represents the mission of the United States DOE, the government agency that oversees the NSUF. "We report to the DOE on our view of the quality of the technical and administrative work being done at the NSUF," Sackett continues. "And every year, we've given it very high marks."

As an example of a recent recommendation the board made that the NSUF acted upon, O'Kelly points to an improvement suggested to Rory Kennedy, the new director of the NSUF. "The board noticed that getting samples irradiated at the ATR and then shipping them to graduate students working on projects was taking far too long," he says. "After all, graduate students only have so many years to find a project they feel comfortable working on. And the longer they have to wait for samples, the less time they have to complete their theses. So the review board recommended that the ATR NSUF improve the throughput of its samples."

The user facility did better than that. It increased student access to the sample library of already irradiated material leftover from previous experiments. "The board agreed this was an excellent idea," said Klein. "Students can now get their irradiated samples much faster than if they had to wait their turn on the reactor."

Of course, there is no dearth of projects still going through the reactor, but for students anxious to start their training, having test materials readily in hand is a big advantage.

New Voices. New Perspectives.

Although the NSUF was initially primarily accessed by universities and national laboratories, now the SRB and NSUF are seeking more participation from the private sector and the international community.

The board includes a member from the United Kingdom.

"International participation on the board has always been very important," Sackett said. In fact, when the board was originally formed, it included a member from Japan. Today, Simon Pimblott, professor of radiation chemistry at the University of Manchester in England, serves on the SRB. Pimblott's presence is significant for two reasons. First, it promotes a cooperative effort between the NSUF and the United Kingdom's newly founded National Nuclear User Facility (NNUF).

Secondly, building a new nuclear test facility is a very expensive proposition. The United States government could pursue a truly robust nuclear power development program on its own, but it is more cost-effective to leverage resources from national laboratories, universities and international partners.

"At present," Sackett said, "efforts to develop relationships with partners overseas are constrained by limited budgets on both sides, but we all hope that funding increases from our respective governments are in the works, because there is so much value in sharing our test facility capabilities."

By the same token, having representatives from the private sector on the SRB not only provides valuable resources from a research point of view, it also lends a pragmatic, real-world perspective to the NSUF's work. For example, one of the major concerns private industry has is that the research and work at the NSUF not only be relevant to commercial needs, but that it's done on budget and on schedule.

"In the nuclear science field, the interests of private industry and academia are closely intertwined," Klein said. "For example, one of the things our industry partner has stressed is balancing the costs and effectiveness of the technical support provided during experiments. One of the ways we've sought that balance is by optimizing our research processes on projects that begin at one facility and then must be moved to another when substantial demands on time exist at both facilities."

"The SRB has helped establish lines of communication through which industry or universities can say to the NSUF, 'We have a need. Can someone do a project on this?'" O'Kelly said. "So, now, you have university professors working on problems private industry has identified that, before, no one at the national labs or universities knew were out there. Now we do."

The SRB hopes that the NSUF can attract more members from the private sector, as well as from the international community. This seeming expansion of the NSUF's mission has led to a subtle yet dramatic shift in how it presents itself.

What's in a name? Everything.

If you didn't notice it on the cover of this annual report, the name of Advanced Test Reactor National Scientific User Facility has been changed to the Nuclear Science User Facilities. As we said, it's not a major shift. It's hardly noticeable, in fact.

Perhaps more apparent is the logo. The graphic of the United States with circles emanating from the ATR NSUF in southern Idaho has been replaced by a world globe.

"Both the new name and new logo signify the logical transition from our original focus on the Advanced Test Reactor to what we have grown into," says Sackett, pointing out that the reference to ATR has been removed. "We are an extended family of facilities providing research and technical expertise to a broad spectrum of scientific needs that go beyond just the materials and fuels sciences, but embrace other fields as well."

"The name change better represents the broad—and growing—set of capabilities available to researchers," Klein said.

"It's a reflection of the value not only of the NSUF but the SRB itself," O'Kelly said. "Board members want the NSUF to achieve its highest potential. They want it to grow, to become significantly larger than it is today, and to provide broad researcher access to the national nuclear research infrastructure, but they're not biased about how it gets there, they want to do it right. And the new name shows that we're heading in the right direction."

INTERVIEW WITH THE DIRECTOR

J. Rory Kennedy
Director
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An Interview with J. Rory Kennedy, Ph.D.

Dr. Rory Kennedy was appointed director of the Advanced Test Reactor National Scientific User Facility in January 2014, replacing Jeff Terry, who returned to his faculty position at the Illinois Institute of Technology. He is the first full-time director in the ATR NSUF's eight-year history, an indication of how the organization and the job itself have grown. Recently, Kennedy sat down with Sarah Robertson, NSUF's communications specialist, to talk about where the NSUF is now, where it's going and how it is re-emphasizing the resources, skills and strengths available through the NSUF's nuclear research partnerships.

Q: *First of all, can you briefly give us a little background on yourself? What did you do before you became director of the user facility?*

A: Before I took this position, I was the national technical lead for Metallic Fuel Technology Development within the Advanced Fuels Campaign of DOE's Fuel Cycle Research and Development Program.

Q: *You've been director for just over a year now. What have you learned in that time?*

A: I've learned how important it is to maintain a good relationship with the DOE, to work very closely with them, and to communicate with them on a regular basis. They act as our primary sponsor and interface with federal government policy makers. So that was a big learning experience since I didn't have quite the degree of contact with the DOE before as I do now.

I also learned how useful and valuable the user facility as a whole is to the advancement of nuclear research. Before, I concentrated on the individual program I was working. I had a very directed scope that commanded my full attention. Now, as director of the user facility, I am responsible for a much broader scope of research. There are many different types of investigations being conducted through the ATR NSUF and I have to have a significant degree of understanding of each of them. It's given me a greater appreciation for the set of diverse talents required to reach our common goal.

It's also made me more aware of the gaps in the data and the fundamental knowledge that I didn't realize existed in the field of nuclear fuels and materials that need to be addressed.

Q: *What sort of gaps?*

A: Well, let's take cladding as an example. There are cladding materials that have undergone systematic studies over the last 60-plus years, so you'd expect to be able to find data on the irradiation behavior of these materials at pretty much any temperature, neutron flux, and time in reactor. But in many cases, the studies that would provide the specific data you're looking for have never been performed. I find that

rather surprising, because over that period of time, I would have assumed that all that information would have been collected. The answer likely is that the scientists involved limited their research to fairly constrained sets of reactor conditions. As we move forward with advanced reactor designs, we need to understand the effects of irradiation over a broader range of conditions—filling in the gaps in the needed data, if you will—to really gain a better understanding of how these types of materials can be used in future reactors.

Q: *I know we're in the process of changing the name of the user facility. Can you tell us something about that?*

A: This was under consideration before I became director and I think it's a good idea, because if you look at the user facility and how it has grown since 2007, the current name does not adequately reflect that growth.

Of course, the original concept of the user facility centered around INL and its Advanced Test Reactor along with our PIE capabilities and the instrumentation that we were developing. But then its project load started expanding fairly quickly as the interest in the ATR and INL PIE facilities increased so that we couldn't handle it all and we started adding other existing facilities that could assume some of the workload as partners. Now, along with ATR, we can assign projects to the High Flux Isotope Reactor (HFIR) at Oak Ridge, the MIT reactor at Massachusetts Institute of Technology, or the PULSTAR reactor at North Carolina State University. And, as we get more projects, other reactors

of value might be added as part of our user facilities. The same can be said about the PIE facilities

So we have evolved. Rather than just one single entity user facility, we are now a network of facilities stretching from one coast to the other, all dedicated to the study and advancement of nuclear science.

And so, because we're expanding our resources, our capabilities and the community we engage, we started the conversation about changing our name to reflect this growing organization that had taken on a national scope.

After a great deal of discussion, we decided to change the name from the ATR National Scientific User Facility to the Nuclear Science User Facilities, which expresses more accurately what we really are: a broader national program that encompasses partner facilities around the country. And that's what we want to reflect. This also keeps the NSUF abbreviation that everyone is accustomed to.

Q: *Another significant change in policy is project-forward funding. Can you tell us about that?*

A: Yes. This was a very good suggestion from DOE because it alleviates the potential for fluctuations in funding from one year to the next. Up until now, each project was funded one year at a time. So, the possibility existed—and it has happened—that a project would have to be interrupted because there was not enough funding for it in the budget for that year.

Now, we get a cost estimate for the full scope of the project and the full cost of the project comes out of the funding for the year the project is approved. That money is then distributed as needed over the life of the project. So, instead of going year-to-year with the funding, the entire amount of support is there to carry the project to its completion.

Not surprisingly, the facilities look very favorably on this since it allows them to plan their facilities usage and budgets better, sometimes years in advance.

Another thing we've done that affects forward-funding is we've expanded the maximum length of time we'll support large irradiation and PIE projects up to seven years. Here we envision this schedule to cover one year for design and fabrication, three years irradiation and three years PIE.

This year is our first experience with forward funding and we're learning a lot. But we have a schedule, we have the funding, we have the people, and we have the facilities. So, even with its challenges, we're excited about the process improvements forward-funding can provide us.

Q: *Can you tell me about some other changes that you're working on?*

A: We're learning and we're instituting other practices that will help us improve our processes even more. For example, this year we've set up a number of metrics that will help us evaluate the effectiveness of these new processes and how the partnership program as a whole is progressing.

Another thing we changed in 2014 was designating who can lead a project. Previously, the policy was that only a university could submit an application to the NSUF. This year, we've opened the submission process up to everyone. So now, university, national laboratory, private industry and small business can all apply as principal investigator. And this is true not only for entities in the U.S., but foreign researchers as well. The only stipulation is that they have a U.S. entity as a co-lead. We want to make the benefits of the partnership program available to the broadest possible number of users and the broadest possible number of ideas. We want to encourage as many good ideas as we can, no matter where they come from.

Q: *It sounds like you're looking to expand the partnership program.*

A: Yes we are, and on two fronts. We've always been open to increasing the number of partners that offer unique capabilities but we're also trying to increase our use of the partners to engage them more and to allow them to be more involved as significant contributors to the user community.

It's all part of why we're changing the name to be more inclusive. We want the user facility as an organization to support the partners and we want the partners to support the user facility as an organization.

Q: *In your letter at the beginning of this annual report, you mention two resources you're developing to help researchers in their work. Can you talk about those?*

A: We're creating a database of all the capabilities available to DOE NE-supported researchers through this effort. There are a whole slew of analyses we'll be doing with this database: cost to maintain facilities, cost to replace equipment, facilities utilization, cost of utilization, equipment condition, anticipated remaining life and lots more.

Connected to that is something we're calling a gap analysis, which is basically compiling a list of the capabilities we may want to invest in according to

a cost-benefit analysis. For example, if we identify a required or desirable post-irradiation examination technology that's not currently accessible in the U.S., we do an assessment of how we might establish it in the U.S. and where.

The other side of the gap analysis is to identify if there are capabilities with a high degree of redundancy or under utilization. Is the same PIE technique duplicated at several facilities, but is only being used 20 percent of the time? So if a facility asks to add the same technology, we may suggest that the institution consider using the already established yet under-utilized capability. It's a big job to create this database, manage it and then employ it to help DOE use its capabilities more cost-efficiently.

This capability database will be made available to the public so users can come in and determine for themselves if the technology or area of expertise they need is already available, either in the U.S. or internationally, or if it has potential as an area of opportunity for their own institution. Also, individual researchers can use the database to help develop their research projects and proposals.

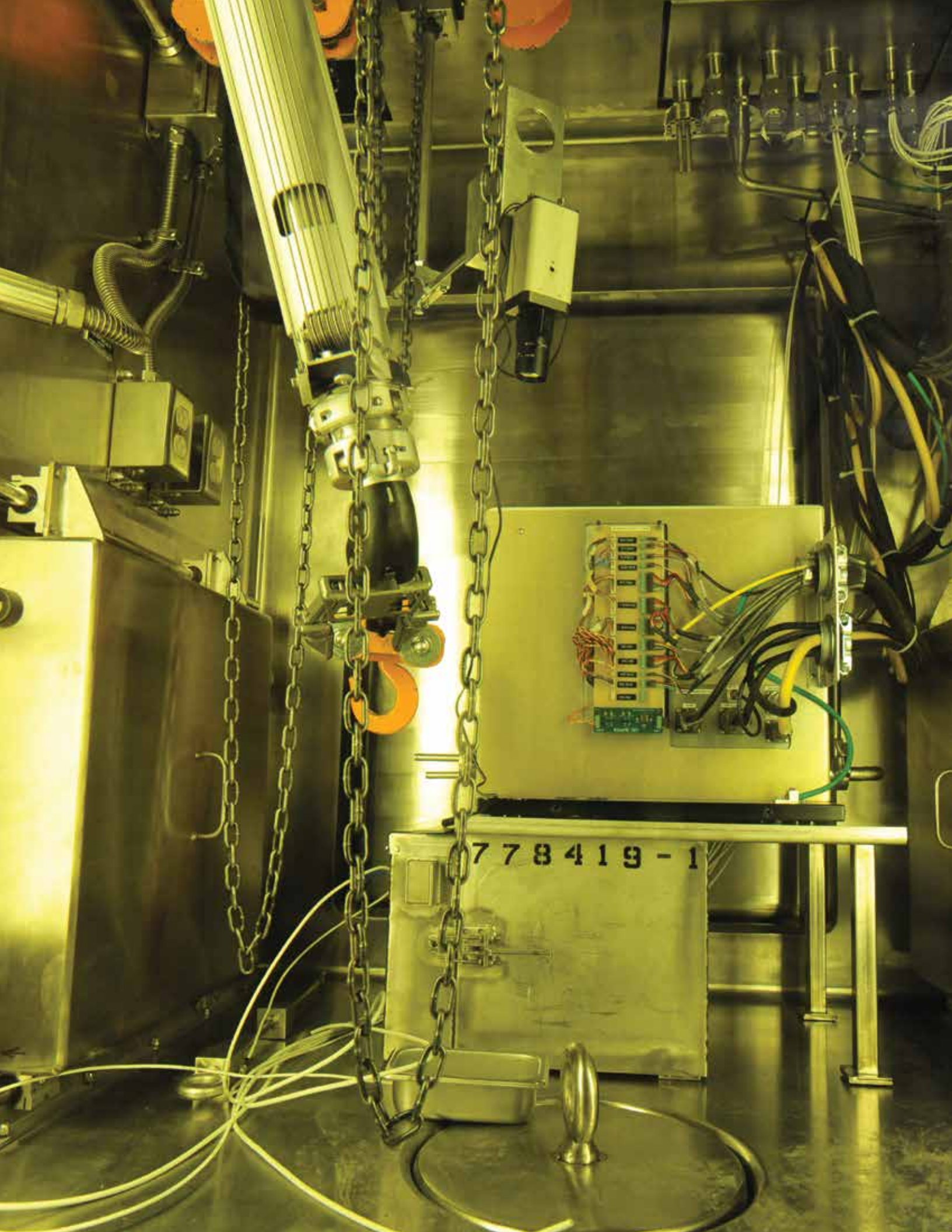
The second resource we're continuing to develop is the NSUF sample library. It has proven to be very valuable from cost-efficiency and time-saving perspectives and so we are actively moving to enhance it. There are a lot of irradiated materials that have been sitting around for quite some time. There's no reason for us to do an irradiation test that could cost upwards of a million dollars if the irradiated samples already exist. And there may already be a knowledge base associated with those samples that we haven't identified. It is very important, though, that the samples have a very good pedigree documented.

As we go forward, we will continue to add to the sample library with new and unique samples as we conduct additional irradiation tests. In addition, we are engaging the international community in the hopes that there could eventually be an international sample library of materials available to nuclear researchers.

DOE is very interested in pursuing this kind of cooperative effort and the NSUF is more than willing to be the tool to facilitate it. In fact, we're trying to establish a collaboration with the United Kingdom, which just recently established its own user facility. One of the conversations we've had with them dealt with the sample library. They have a large number of archived samples and agree that we should work toward collaborating in some way. We're hoping that once the momentum is achieved with the UK, we can use it to forge relationships with other countries as well.

Q: Finally, what's your view of the future of nuclear energy, both in the U.S. and abroad?

A: I'm very optimistic both on the domestic and international fronts. The fact is nuclear energy has a very high value as an inexpensive source of energy over the long term. Technologies are being investigated that can allow us to build safer, more efficient reactors that can last perhaps 100 years or more. So while their initial construction costs can be high, the cost of the power it produces over the life of that reactor is actually very low. At the same time, nuclear power helps reduce the carbon emissions produced by fossil-fuel power generation. It's so gratifying that the NSUF and the researchers and staff members at our various partner facilities are making a significant contribution to making nuclear energy a safer, more efficient, more environmentally responsible energy resource for the entire world.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PROGRAM OVERVIEW

NSUF: A Model for Collaboration

NSUF and its partner facilities represent a prototype laboratory for the future. This unique model utilizes a distributed partnership with each facility bringing exceptional capabilities to the relationship including reactors, beamlines, state-of-the-art instruments, hot cells, and most importantly, expert mentors. Together these capabilities and people create a nationwide infrastructure that allows the best ideas to be proven using the most advanced capabilities. Through NSUF, university researchers and their collaborators are building on current knowledge to better understand the complex behavior of materials and fuels in a nuclear reactor.

In 2014, NSUF's partnership program had eight universities, two national laboratories, and added one industry partner. The partner facility capabilities greatly expand the types of research offered to users. The avenues opened through these partnerships facilitate cooperative research across the country, matching people with capabilities and students with mentors. In 2014, NSUF included INL and the following institutions:

- Illinois Institute of Technology
- Massachusetts Institute of Technology
- North Carolina State University
- Oak Ridge National Laboratory
- Pacific Northwest National Laboratory
- Purdue University
- University of California, Berkeley
- University of Michigan
- University of Nevada, Las Vegas
- University of Wisconsin
- Westinghouse

The pages that follow contain specific details on the capabilities of NSUF, its partners, and how to access these capabilities through the calls for proposals. There is also information on the Users Meeting, a yearly event hosted by NSUF designed to instruct and inform. This event is free of charge to interested persons and a number of scholarships for travel and hotel are offered to students and faculty. Take time to familiarize yourself with the many opportunities offered by NSUF and consider submitting a proposal or two.

NSUF Research Supports DOE-NE Missions

The U.S. DOE-NE organizes its research and development activities based on four main objectives that address challenges to expanding the use of nuclear power:

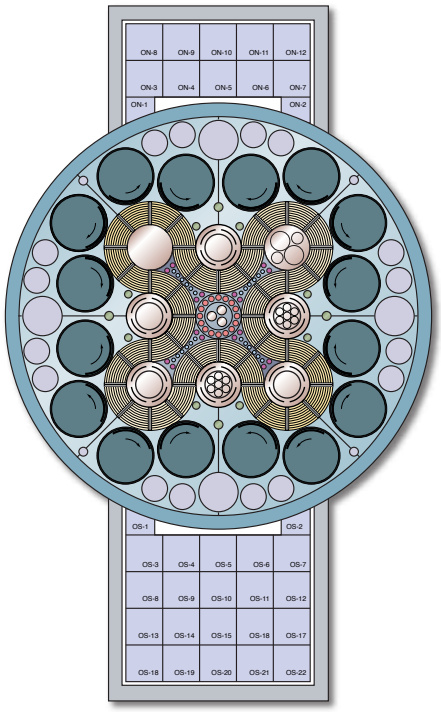
- Develop technologies and other solutions that can improve the reliability, sustain the safety, and extend the life of current reactors.
- Develop improvements in the affordability of new reactors to enable nuclear energy to help meet the administration's energy security and climate change goals.
- Develop sustainable nuclear fuel cycles.
- Understand and minimize the risks of nuclear proliferation and terrorism.

NSUF research addresses a number of these mission needs. Most of the research contained in this report looks at either understanding the mechanisms of radiation on materials and fuels to address the challenges of the aging current fleet or looks at materials and fuels for the next generation of reactors. To be eligible as an NSUF research project, the research must support at least one of the DOE-NE missions. For specifc information on DOE missions, go to http://www.energy.gov/ne/mission.

To learn more about proposing a research project, visit the NSUF website: http://nsuf.inl.gov.

U.S. DEPARTMENT OF ENERGY
Nuclear Energy

REACTOR Capabilities



ATR's serpentine design allows a variety of experiment configurations.

ATR NSUF offers access to a number of reactors. ATR is located at the ATR Complex (ATR) on the INL Site and has been operating continuously since 1967. In recent years, the reactor has been used for a wide variety of government and privately sponsored research. The ATRC reactor is low-power version of ATR.

The MIT reactor is a 5-MW reactor with positions for in-core fuels and materials experiments. Oak Ridge National Laboratory's (ORNL) HFIR is an 85-MW reactor offering steady-state neutron flux and a variety of experiment positions. The PULSTAR reactor at North Carolina State University is a pool-type reactor that offers response characteristics similar to commercial light water power reactors.

Idaho National Laboratory Advanced Test Reactor

ATR is a water-cooled, high-flux test reactor, with a unique serpentine design that allows large power variations among its flux traps. The reactor's curved fuel arrangement places fuel closer on all sides of the flux trap positions than is possible in a rectangular grid. The reactor has nine of these high-intensity neutron flux traps and 68 additional irradiation positions inside the reactor core reflector tank, each of which can contain multiple experiments. Experiment positions vary in size from 0.5 to 5 inches in diameter (1.27 to 12.7 centimeters) and all are 48 inches (121.92 centimeters) long. The peak thermal flux is 1X1015 n/cm^2-sec and fast flux is 5X1014 n/cm -sec when operating at full power of 250 MW. There is a hydraulic shuttle irradiation system, which allows experiments to be inserted and removed during reactor operation, and pressurized water reactor (PWR) loops, which enable tests to be performed at prototypical PWR operating conditions.

Idaho National Laboratory Advanced Test Reactor Critical Facility

ATRC is a low-power version (same size and geometry) of the higher-powered ATR core. It is operated at power levels less than 5 KW with typical operating power levels of 600 W or less. ATRC is primarily used to provide data for the design and safe operation of experiments for ATR. ATRC is also used to supply core performance data for the restart of ATR after periodic core internals replacement. Occasionally ATRC is used to perform low-power irradiation of experiments.

Oak Ridge National Laboratory High Flux Isotope Reactor

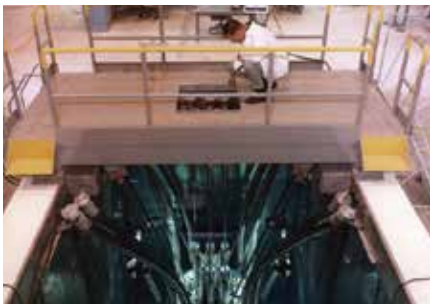
HFIR is a versatile 85-MW research reactor offering the highest steady-state neutron flux in the western world. With a peak thermal flux of 2.5×10^{15} n/cm^2-s and a peak fast flux of 1.1×10^{15} n/cm^2-s, HFIR is able to quickly generate isotopes that require multiple neutron captures and perform materials irradiations that simulate lifetimes of power reactor use in a fraction of the time. HFIR typically operates seven cycles per year, each cycle lasting between 23 and 26 days. Associated irradiation processing facilities include the Hydraulic Tube Facility, Pneumatic Tube Facilities for Neutron Activation Analysis (NAA) and Gamma Irradiation Facility.

Massachusetts Institute of Technology Reactor

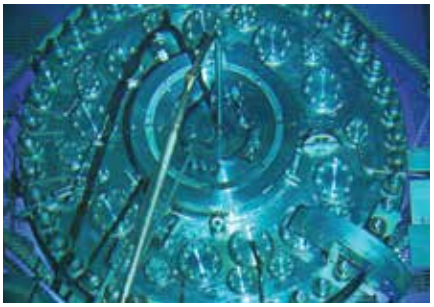
Massachusetts Institute of Technology (MITR) is a 5-MW tank-type research reactor. It has three positions available for in-core fuel and materials experiments over a wide range of conditions. Water loops at pressurized water reactor/boiling water reactor (PWR/BWR) conditions, high-temperature gas reactor environments at temperatures up to 1400°C and fuel tests at light water LWR temperatures have been operated and custom conditions can also be provided. A variety of instrumentation and support facilities are available. Fast and thermal neutron fluxes are up to 1014 and 5X1014 n/cm^2-s. MITR has received approval from the Nuclear Regulatory Commission for a power increase to 6 MW, which will enhance the neutron fluxes by 20 percent.

North Carolina State University PULSTAR Reactor

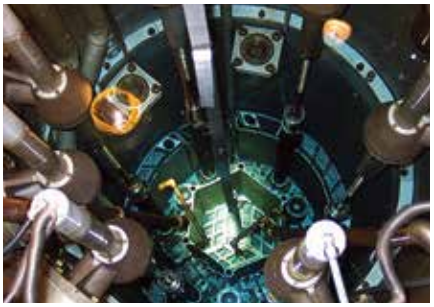
The PULSTAR reactor is a 1-MW pool-type nuclear research reactor located in North Carolina State University's (NCSU) Burlington Engineering Laboratories. The reactor, one of two PULSTAR reactors built and the only one still in operation, uses 4 percent enriched, pin-type fuel consisting of uranium dioxide pellets in zircaloy cladding. The fuel provides response characteristics that are very similar to commercial light water power reactors. These characteristics allow teaching experiments to measure moderator temperature and power reactivity coefficients including Doppler feedback. In 2007, the PULSTAR reactor produced the most intense low-energy positron beam with the highest positron rate of any comparable facility worldwide.



Aerial view of the ATRC reactor core and bridge.



Top of the HFIR reactor.



Annular fuel rig in the MITR core.

Downward view of the PULSTAR reactor pool.

POST-IRRADIATION EXAMINATION Capabilities

Hot Fuel Examination Facility, located at the Materials and Fuels Complex at DOE's INL Site in Idaho.

ATR NSUF offers researchers access to a broad range of PIE facilities.

These include capabilities at INL's MFC; the Microscopy and Characterization Suite (MaCS) at the Center for Advanced Energy Studies; the Nuclear Services Laboratories at North Carolina State University; hot cells, radiological laboratories and the Low Activation Materials Development and Analysis (LAMDA) facility at Oak Ridge National Laboratory; the Radiochemistry and Materials Science and Technology Laboratories at Pacific Northwest National Laboratory; the Interaction of Materials with Particles and Components Testing (IMPACT) facility at Purdue University; several instruments from the Nuclear Materials Laboratory at University of California, Berkeley; the Irradiated Materials Complex at the University of Michigan; the Harry Reid Center Radiochemistry Laboratories at the University of Nevada, Las Vegas; and the Characterization Laboratory for Irradiated Materials at the University of Wisconsin.

Transmission electron microscope, one of many PIE capabilities in the Microscopy & Characterization Suite (MaCS) at the Center for Advanced Energy Studies in Idaho Falls, Idaho.

Idaho National Laboratory: Hot Fuel Examination Facility Analytical Laboratory, Electron Microscopy Laboratory

Hot Fuel Examination Facility (HFEF) is a large alpha-gamma hot cell facility dedicated to remote examination of highly irradiated fuel and structural materials. Its capabilities include nondestructive and destructive examinations. The facility also offers a 250-kWth Training Research Isotope General Atomics (TRIGA) reactor used for neutron radiography to examine internal features of fuel elements and assemblies.

The Analytical Laboratory is dedicated to analytical chemistry of irradiated and radioactive materials. It offers NIST-traceable chemical and isotopic analysis of irradiated fuel and material via a wide range of spectrometric techniques.

The Positronium Annihilation Lifetime Spectrometer, located in the PULSTAR reactor facility on the NC State North Campus in Raleigh, N.C.

The Scanning Electron Microscope in Oak Ridge National Laboratory's LAMDA facility.

A hot cell in the Radiochemistry Processing Laboratory at Pacific Northwest National Laboratory.

The Electron Microscopy Laboratory (EML) is dedicated to materials characterization, primarily using transmission electron, scanning electron and optical microscopy. The EML also houses a dual-beam FIB that allows examination and small-sample preparation of radioactive materials.

Center for Advanced Energy Studies Microscopy and Characterization Suite

The MaCS is equipped to handle low-level radiological samples as well as non-radiological samples. MaCS offers several high-end pieces of equipment, including a local electrode atom probe (LEAP), automated hardness tester, scanning electron microscope (SEM), nano indenter and atomic force microscope, TEM and focused ion beam.

North Carolina State University Nuclear Services Laboratories

Post-irradiation examination capabilities at NCSU's Nuclear Services Laboratories include neutron activation analysis, radiography and imaging capabilities, and positron spectrometry.

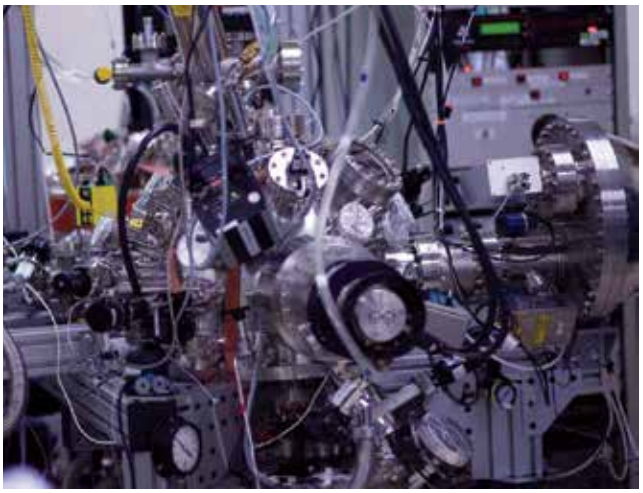
Oak Ridge National Laboratory Hot Cells, Radiological Laboratories, LAMDA Facility

ORNL hot cells and radiological laboratories offer a wide variety of research and development and production capabilities from radiochemistry and isotope packaging to materials testing to irradiated fuels examination. Facilities include the Irradiated Materials Examination and Testing (IMET) facility, Irradiated Fuels Examination Laboratory (IFEL) and Radiochemical Engineering Development Center (REDC).

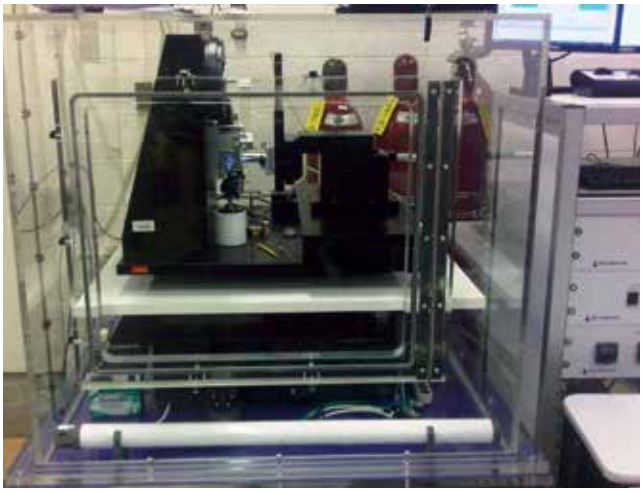
The Low Activation Materials Development and Analysis (LAMDA) Laboratory added in 2012 offers post-irradiation examination capabilities including refractory element test stands for tensile testing, optical and scanning electron microscopes, and thermal diffusivity and density measurement equipment.

Pacific Northwest National Laboratory Radiochemistry Processing Laboratory, Materials Science and Technology Laboratory

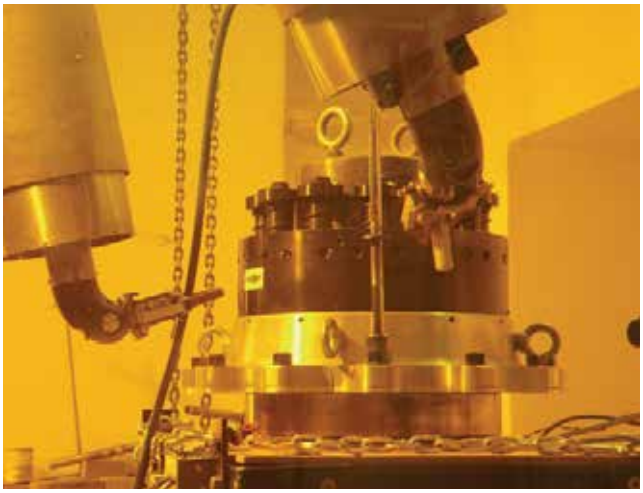
The RPL and the Materials Science and Technology Laboratory (MSTL) offer a wide range of specialized equipment for handling and testing fuels and materials. Capabilities include experiment hardware design, fabrication and assembly, testing facilities for both nonradioactive and radioactive structural materials, and the advanced characterization of unirradiated and irradiated fuels and materials using instruments including TEM, SEM and optical microscopy.



The IMPACT facility at Purdue University.



UC Berkeley nano-indentation system.



Capability at the Irradiated Materials Complex on the UM campus at Ann Arbor, Michigan.

Purdue University IMPACT Facility

The IMPACT facility offers a wide range of spectroscopy techniques to study the surface of materials. The IMPACT facility houses a variety of examination instruments including low-energy scattering spectroscopy (LEISS), X-ray photoelectron spectroscopy (XPS), auger electron spectroscopy (AES), extreme ultraviolet reflectometry (EUVR), extreme ultraviolet (EUV) photoelectron spectroscopy and mass spectrometry.

University of California, Berkeley Nuclear Materials Laboratory

The Nuclear Materials Laboratory provides several capabilities for examining irradiated material samples including a nano-indentation system for nano and microscale hardness testing at ambient and elevated temperature and inert environments, positron annihilation spectroscopy, and warm sample preparation (polishing, cutting, grinding and mounting).

University of Michigan Irradiated Materials Complex

The Irradiated Materials Complex provides laboratories and hot cells with capabilities for conducting high-temperature mechanical properties, and corrosion and stress corrosion cracking experiments on neutron-irradiated materials in an aqueous environment, including supercritical water, and for characterizing the fracture surfaces after failure.

University of Nevada, Las Vegas Harry Reid Center Radiochemistry Laboratories

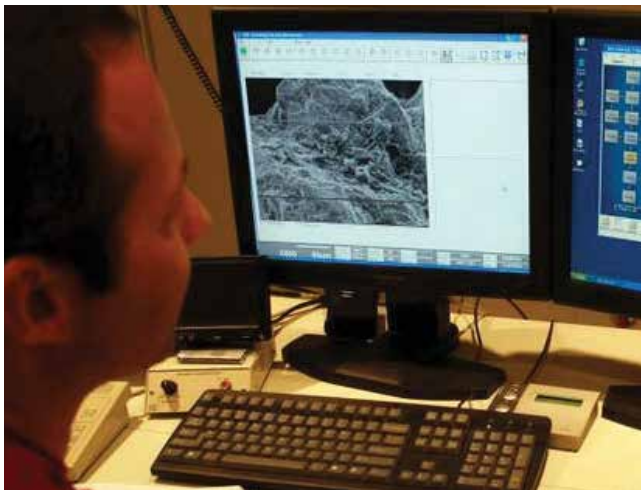
Post-irradiation examination capabilities at the Radiochemistry Laboratories include metallographic microscopy, X-ray powder diffraction, Rietveld analysis, SEM and TEM, electron probe microanalysis, and X-ray fluorescence spectrometry.

University of Wisconsin Characterization Laboratory for Irradiated Materials

The Characterization Laboratory for Irradiated Materials offers PIE capabilities including SEM and TEM on neutron-irradiated materials.

Westinghouse Materials Center of Excellence Laboratories

Westinghouse offers its Materials Center of Excellence Laboratories (MCOE) Hot Cell Facility and accompanying laboratories to provide experimental support to ATR-related nuclear energy materials research programs. The Westinghouse facilities in Churchill, Pennsylvania, are housed in four cells that provide a broad range of testing, evaluation and characterization capabilities for both unirradiated and irradiated materials. In-place capabilities include the ability to test under a variety of environments, an extensive mechanical testing laboratory, a specialized corrosion and stress corrosion cracking lab, and materials microstructure and chemical characterization instruments. Specialized facilities are also available to measure the radioactivity properties of materials under investigation as well as neutron and gamma sources facilities, which can be employed to assess materials' response to in-situ radiation.



PIE capabilities at the Harry Reid Center Radiochemistry Laboratories, located on the UNLV campus in Las Vegas, Nevada.

A JEOL 200CX TEM equipped with EDS and scanning system, and an electro-polisher and dimpler at the Characterization Laboratory for Irradiated Materials, located on the UW campus in Madison, Wisconsin.

Operators use manipulators to perform work at the Westinghouse Hot Cell Lab.

BEAMLINE Capabilities

Aerial view of the Advanced Photon Source at Argonne National Laboratory, located in Argonne, Illinois.

ATR NSUF offers researchers access to a broad range of facilities with beamlines, including accelerator facilities for radiation damage experiments, synchrotron radiation studies, neutron diffraction and imaging, as well as positron and neutron activation analysis.

In 2014, the ATR NSUF program offered researchers access to four university partner beamline facilities. These include the Illinois Institute of Technology Materials Research Collaborative Access Team (MRCAT) beamline at Argonne's Advanced Photon Source, the PULSTAR reactor facility at North Carolina State University, the University of Michigan Ion Beam Laboratory, and the University of Wisconsin Tandem Accelerator Ion Beam.

Illinois Institute of Technology (IIT) MRCAT at Argonne National Laboratory's Advanced Photon Source

The MRCAT beamline offers a wide array of synchrotron radiation experiment capabilities, including X-ray diffraction, X-ray absorption, X-ray fluorescence and 5-μm-spot size fluorescence microscopy.

North Carolina State University PULSTAR Reactor Facility

The PULSTAR reactor facility offers a selection of dedicated irradiation beam port facilities—neutron powder diffraction, neutron imaging, intense positron source and ultra-cold neutron source. An intense positron source has been developed to supply a high-rate positron beam to two different positron/positronium annihilation lifetime spectrometers.

University of Michigan Michigan Ion Beam Laboratory

The 1.7-MV Tandetron accelerator in the Michigan Ion Beam Laboratory offers controlled temperature proton irradiation capabilities with energies up to 3.4 MeV as well as heavy ion irradiation.

University of Wisconsin Tandem Accelerator Ion Beam

A 1.7-MV terminal voltage tandem ion accelerator (Model 5SDH-4, National Electrostatics Corporation Pelletron accelerator) installed at UW features dual ion sources for producing negative ions with a sputtering source or using a radio frequency (RF) plasma source. The analysis beamline is capable of elastic recoil detection and nuclear reaction analysis.

Positron beam cave containing magnetic switchyards and transport solenoids, located in the PULSTAR reactor facility on the NC State North Campus in Raleigh, NC.

Michigan Ion Beam Laboratory for Surface Modification and Analysis, located on the UM campus in Ann Arbor, Michigan.

Tandem Ion Beam Accelerator, located on the UW campus in Madison, Wisconsin.

CALL FOR PROPOSALS

Jeff Benson
Program Administrator

The NSUF mission is to provide nuclear energy researchers access to world-class capabilities to facilitate the advancement of nuclear science and technology. This mission is supported by providing cost-free access to state-of-the-art experimental irradiation testing and PIE facilities as well as technical assistance in design and analysis of reactor experiments. Access is granted through a competitive proposal process.

NSUF offers research proposal options through an online submittal system that helps prospective researchers develop, edit, review and submit their proposals. NSUF staff is available to help any researcher who desires to submit a proposal.

Submitted proposals should be consistent with the DOE-NE mission and its programmatic interests. These include the Light Water Reactor Sustainability, Fuel Cycle Research and Development, Advanced Modeling and Simulation, Next Generation Nuclear Plant and the Generation IV Nuclear Energy Systems Initiative programs.

All proposals are subject to a peer-review process before selection. All NSUF research must be non-proprietary and results are expected to be published. Collaborations with other national laboratories, federal agencies, non-U.S. universities and industries are encouraged. Any U.S.-based entities, including universities, national laboratories, and industry can propose research that would utilize the MRCAT beamline at the Advanced Photon Source or would be conducted as a rapid turnaround experiment.

Calls for Irradiation, Post-irradiation Examination and Synchrotron Radiation Experiments

Applications are submitted annually through the Consolidated Innovative Nuclear Research Funding Opportunity Announcement. More information is available on the NEUP website, www.neup.gov.

While priority will be given to proposals that further the direction of DOE's nuclear energy research programs, the NSUF will consider all technical feasible proposals for scientific merit and selection.

- Irradiation/post-irradiation examination of materials or fuels.
- PIE of previously irradiated materials or fuels from the NSUF sample library.
- Research that requires the unique capabilities of the Advanced Photon Source through the MRCAT beamline, operated by the Illinois Institute of Technology.

All proposals submitted to the open calls undergo thorough reviews for feasibility, technical merit, relevance

to the DOE-NE missions and cost. The results are compiled and provided to a panel committee who performs a final review and ranks the proposals. The ranking is given to the NSUF director. Awards are announced within two to three months of the call's closing date, generally in January and June. Awards allow users cost-free access to specific NSUF and partner capabilities as determined by the program.

Calls for Rapid Turnaround Experiments

Rapid turnaround experiments are experiments that can be performed quickly—typically in two months or less—and include, but are not limited to, PIE requiring use of an instrument (FIB, TEM, SEM, etc.), irradiations in the PULSTAR reactor, ion beam irradiation and neutron scattering experiments. Proposals for rapid turnaround experiments are reviewed on a quarterly basis in January, April, July and October and awarded based on the following rankings:

- High Priority—Proposal is awarded immediately upon review if funding is available.
- Recommended—Proposal is placed in a queue from which awards are made approximately every other month if funding is available.
- Not Recommended—Proposal is not awarded, but the project investigators are offered an opportunity to read the review comments and then resubmit the proposal for the next call.

For more information, visit the NSUF website, www.nsuf.inl.gov

NSUF Sample Library

NSUF has established a sample library as an additional pathway for research. The library contains irradiated and unirradiated samples in a wide range of material types, from steel samples irradiated in fast reactors to ceramic materials irradiated in the Advanced Test Reactor. Many samples are from previous DOE-funded material and fuel development programs. University researchers can propose to analyze these samples in a PIE-only experiment. Samples from the library may be used for proposals for open calls and rapid turnaround experiments.

As the NSUF program continues to grow, so will the sample library. To review an online list of available specimens, visit the NSUF electronic system at the address above.

USERS MEETING

The annual ATR NSUF Users Meeting offers researchers days of workshops, tours, discussions and classes. The focus is on providing an understanding of key nuclear technology gaps, capabilities required for addressing those gaps, recent or emerging advances and techniques for conducting reactor experiments and PIE.

Users Meeting is not just a way to learn more about ATR NSUF, its capabilities and ongoing research; it is also a great opportunity to meet other students, scientists and engineers who are interested in responding to ATR NSUF's call for proposals. Users Meeting supports ATR NSUF as a model for the laboratory of the future, where collaborative research and shared resources among universities and national laboratories will help prepare a new generation of nuclear energy professionals.

The events are free of charge for students, faculty and post docs as well as researchers from industry and national laboratories who are interested in materials, fuels, PIE and reactor-based technology development. In the six years since its inception, ATR NSUF Users Meeting has hosted 633 participants from 30 countries and 37 U.S. universities.

Support to help defray travel, hotel and meal expenses is offered to university faculty and students on a competitive basis.

What to expect at Users Meeting

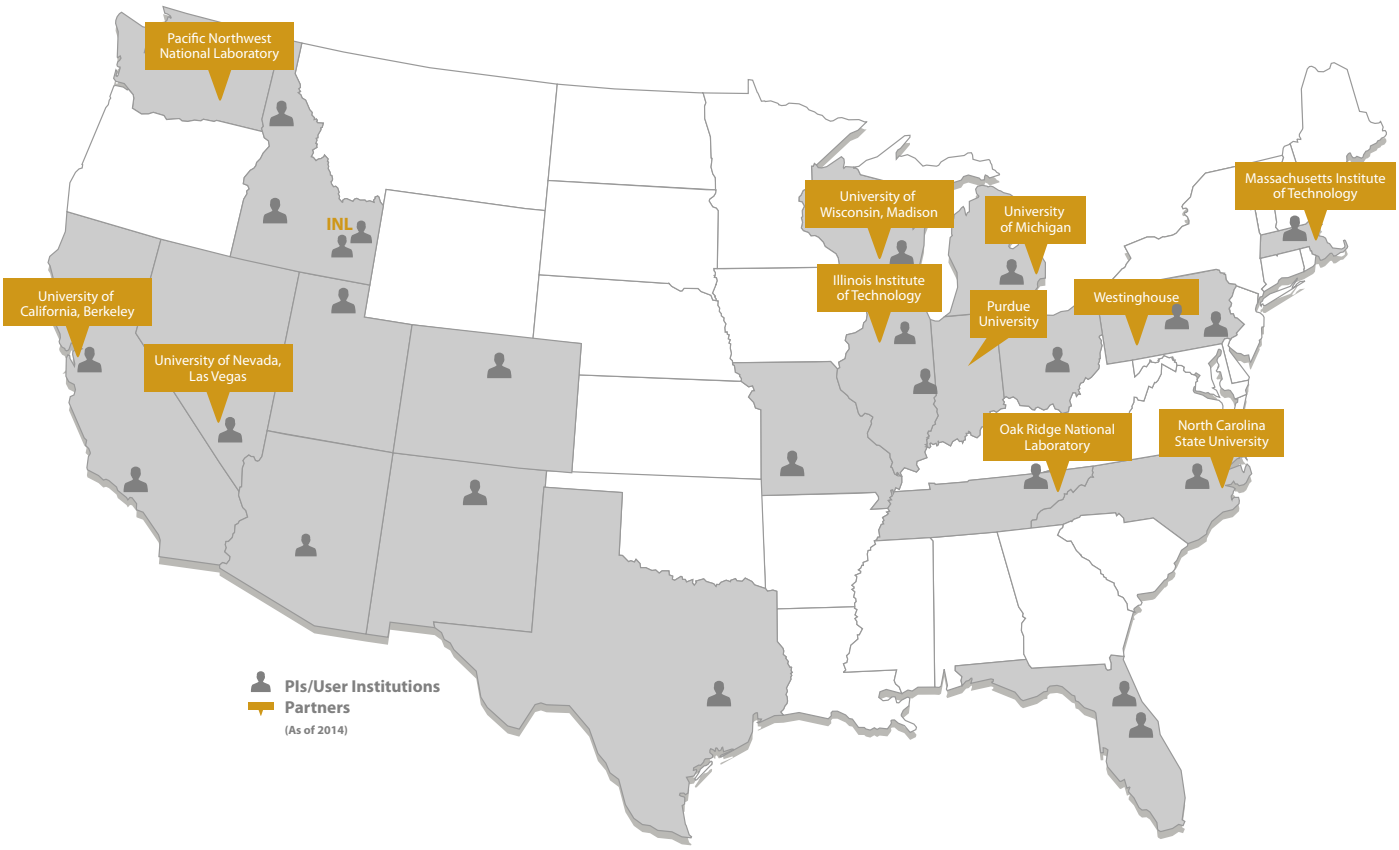
Users Meeting kicks off with an introductory workshop to ATR NSUF, which includes a description of current and upcoming research capabilities offered by INL and its university partners, a briefing on the solicitation process and a welcome from DOE.

Each year, Users Week offers a number of workshops and courses for students to participate. These may vary from year to year, but courses generally focus on a variety of topic-specific areas, such as in-reactor instrumentation, fuels and materials, or how to conduct radiation experiments.

Participants are always offered an opportunity to tour ATR as well as INL's MFC, where many PIE facilities are housed.

Users who are not able to attend the Users Meeting in person now have the ability to participate in the meeting online. For more information about the Users Meeting, visit our website, nsuf.inl.gov.

DISTRIBUTED PARTNERSHIP



GENERAL ATOMICS
ELECTRONIC SYSTEMS
VEXTA

NSUF AWARDED PROJECTS

Awarded Reports 48-135

Industry Program Reports 136-141

Irradiation Effects on Thermophysical Properties of Hafnium-Aluminide Composite: *A Concept for Fast Neutron Testing at ATR*

Heng Ban – Utah State University (USU) – heng.ban@usu.edu

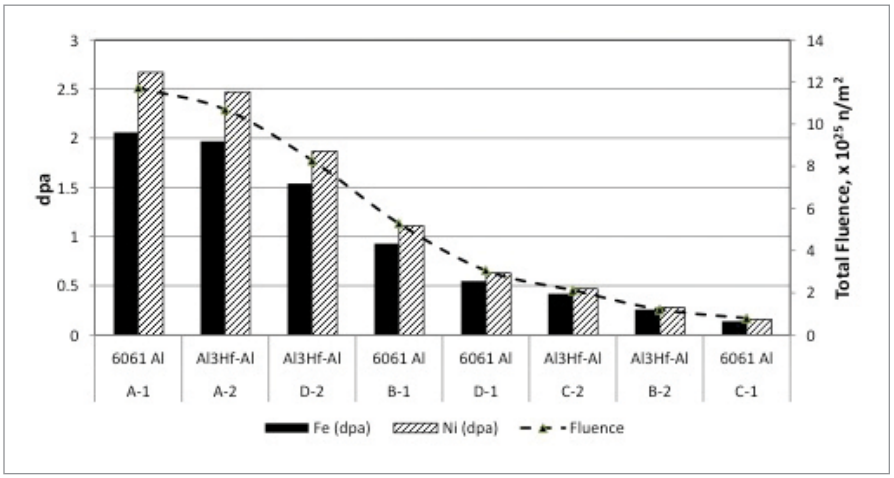


Figure 1. Calculated dpa values and total fluence for experiment.

The ability to conduct fast neutron irradiation tests is essential to meeting fuels and materials development requirements for future nuclear reactors. At the same time, the lack of domestic fast neutron testing capabilities hinders the development of advanced reactors.

The concept behind this project is to equip one of the corner lobes of the Advanced Test Reactor (ATR) with a thermal neutron filter. This material, comprised of hafnium-aluminide (Al_3Hf) particles (~23% by volume) in an aluminum matrix (Al_3Hf-Al), will absorb thermal neutrons and booster fuel, augmenting the neutron flux and heat transfer from the experiment to pressurized water cooling channels.

Thermal analyses conducted on a candidate configuration confirmed that the design of the water-cooled Al_3Hf-Al absorber block is capable of keeping the temperature of all system components below their maximum allowable limits. However, the thermophysical properties of Al_3Hf have never been measured, nor have the effects of irradiation on these properties ever been determined. It is essential, therefore, to obtain data on the effects of irradiation, including corrosion behavior and radioactive decay products, on the thermophysical and mechanical properties of both Al_3Hf intermetallic and Al_3Hf-Al composite before we can proceed with the design and optimization of the filter.

Project Description

The purpose of this project is to evaluate the properties and behavior of this new material. Specific objectives are to determine:

1. The thermophysical and mechanical properties of Al_3Hf intermetallic and Al_3Hf-Al composite at different temperatures.

2. The effects of irradiation on the thermophysical and material properties of Al_3Hf intermetallic and Al_3Hf-Al composite, and physical/morphological, metallurgical, and microstructural changes of Al_3Hf-Al composite after different cycles of irradiation.
3. The decay products of hafnium (Hf-179m1 vs. Hf-179m2) and corrosion behavior of the Al_3 composite.

Successful completion of the project will:

1. Provide the necessary data for the development of fast neutron test capabilities at ATR.
2. Fill a knowledge gap on the basic properties of the Al_3Hf intermetallic and Al_3Hf-Al composite.
3. Advance the scientific understanding of the irradiation effects on these materials.

The end result, in terms of the data and fundamental understanding obtained, will directly support DOE's mission and benefit the science community in general.

Accomplishments

During FY 2014 Zilong Hua, Utah State University (USU), performed 3D microstructural reconstruction of unirradiated specimens of the $HfAl_3$-Al metal matrix composite material developed for this project. Focused ion beam (FIB) milling and electron backscatter diffraction (EBSD) was performed using the FEI Quanta 3D field emission gun (FEG) located at CAES. The gallium ions from the FIB were found to be very damaging to the $HFAl_3$-Al, so a new procedure was developed to enable the acquisition of acceptable Kikuchi patterns. The serial scans were reconstructed using Dream.3D software and visualized using ParaView. This work is pioneering in that 3D microstructural reconstruction has never before been attempted on this material. The procedure was developed using an unirradiated specimen, in preparation for work with an irradiated specimen.

Completed post-irradiation examination (PIE) of specimens irradiated in the ATR includes:

1. Gamma scans of 18 specimens
2. X-ray diffraction (XRD) on four specimens
3. Scanning electron microscopy (SEM) on one specimen
4. Differential scanning calorimeter (DSC) testing on nine specimens
5. Density measurements on nine specimens

DSC results show a marked exotherm on the first heating cycle, a manifestation of radiation damage in the material. DSC was performed on irradiated specimens with 20.0, 28.4 and 36.5 vol% $HfAl_3$ and compared to similar measurements for the unirradiated material. The specific heat of the irradiated material was more than 50% higher than that of the unirradiated material.

Results of the flux monitor analysis were interpreted and published. The performance of the material was evaluated by placing neutron fluence monitors within shrouded and unshrouded holders and irradiating them in the ATR for up to four cycles. The irradiation assembly consisted of eight capsules containing flux monitors placed in holders fabricated from this new material (referred to as "shrouded") or in 6061 aluminum alloy holders (referred to as "unshrouded"). The adjusted neutron fluences were calculated and grouped into three bins—thermal, epithermal, and fast—to evaluate the spectral shift created by the new material. A comparison of shrouded vs. unshrouded fluence monitors showed a thermal fluence decrease of approximately 11% for the shrouded monitors.

For all capsules, the fast-to-thermal neutron ratio was higher for the flux monitors shrouded with the $HfAl_3$-Al composite material, whereas the ratio is nearly uniform for the wires shrouded by the 6061 aluminum material. The fast-to-thermal ratio appears to be fairly consistent for the unshrouded flux wires, regardless of irradiation position (e.g., height in the reactor) or total fluence (e.g., MWd).

"The opportunity to use state-of-the-art instruments and work with top level researchers in the CAES Microscopy Suite is a great experience that has provided me with valuable skills to kickstart my career."

— Zilong Hua, USU
Postdoctoral Researcher

Future Activities

Research yet to be completed on the irradiated materials includes:

1. Measurements of thermal conductivity using the laser flash method on nine specimens
2. Thermal expansion measurements on three specimens
3. 3D EBSD of one specimen
4. Transmission electron microscopy (TEM) utilizing local electrode atom probe (LEAP) of one specimen
5. Tensile and hardness tests on 11 specimens

Zilong Hua and Donna Guillen of INL are developing a model of the thermal conductivity in both the irradiated and unirradiated materials using the EBSD data that has been reconstructed to 3D. Using INL's MARMOT code, they will further investigate the effects of radiation by comparing the response of irradiated vs. unirradiated material to a heat source applied at the boundaries of the material. Thermal conductivity is a key parameter because the material is being developed as a conduction-cooled neutron absorber block.

Publications and Presentations*

1. D.P. Guillen, L.R. Greenwood, and J.R. Parry, 2014, "High Conduction Neutron Absorber to Simulate Fast Reactor Environment in an Existing Test Reactor," *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 302, No. 1, pp. 413–424.
2. D.P. Guillen, Z. Hua and H. Ban, 2014, "Procedure For 3D Microstructure Reconstruction of a Heterogeneous Metal Matrix Composite Material," 3D Materials Science 2014, *Annecy, France, June 29–July 2, 2014.*
3. D.P. Guillen, J. Burns, Z. Hua and H. Ban, 2014, "*Microstructure of Aluminum Matrix in Composite Absorber Block Material,*" *2014 TMS Meeting, San Diego, CA, Feb. 16-20, 2014.*

**See additional publications from other years in the Media Library on the NSUF website.*

Distributed Partnership at a Glance

ATR NSUF and Partners	Facilities and Capabilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Idaho National Laboratory	Advanced Test Reactor
Collaborators	
Idaho National Laboratory	Donna Post Guillen (principal investigator)
University of Nevada, Las Vegas	Thomas Hartmann (co-principal investigator)
Utah State University	Heng Ban (principal investigator), Zilong Hua (postdoctoral researcher), Adam Zabriskie (graduate student), Kurt Harris (undergraduate student), Heather Wampler (undergraduate student)

This experiment will provide the necessary data for the development of a new material that will enable a robust fast neutron test capability at ATR.

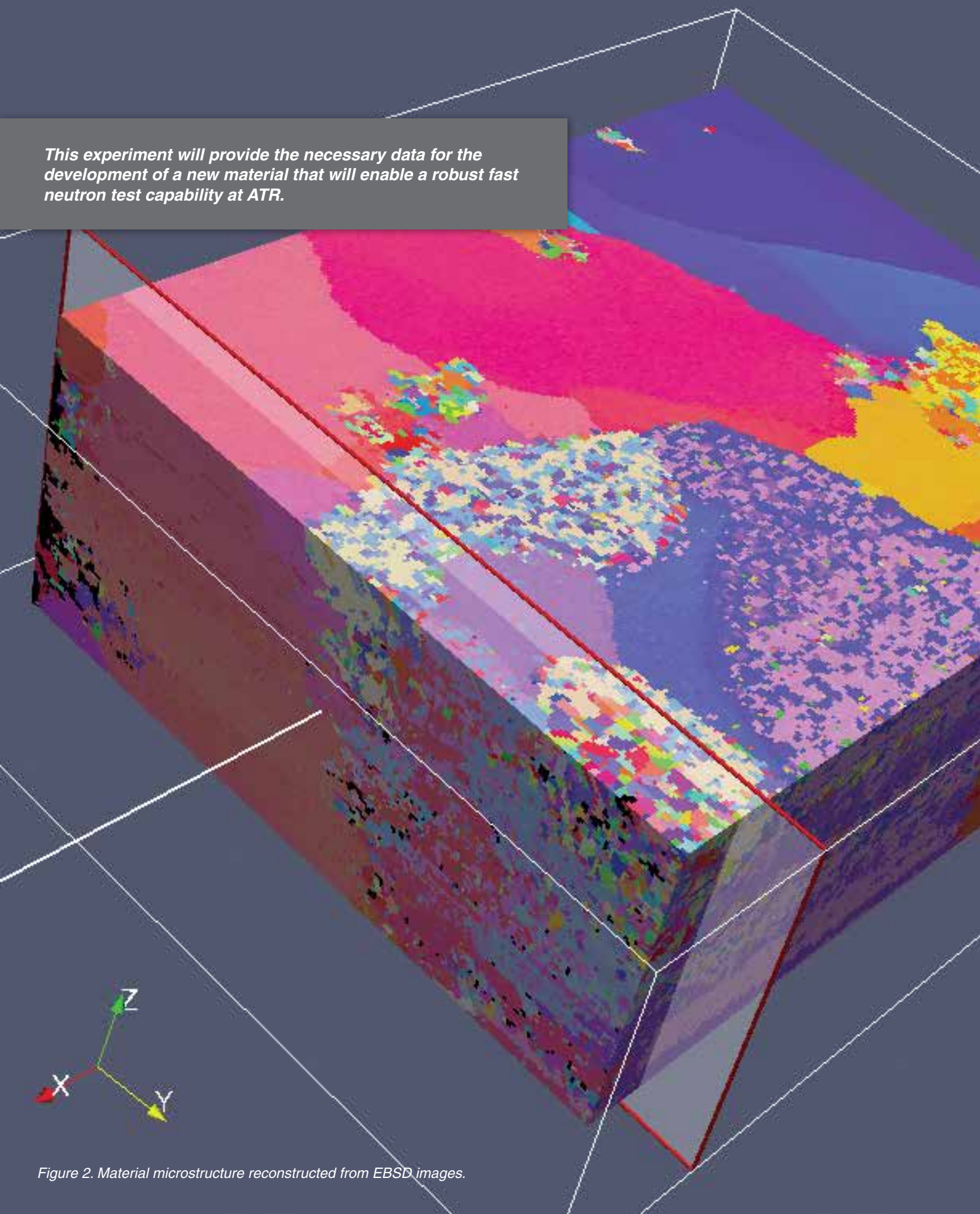


Figure 2. Material microstructure reconstructed from EBSD images.

Measurement of Actinide Neutronic Transmutation Rates with Accelerator Mass Spectroscopy (MANTRA)

George Imel – Idaho State University – gimel@isu.edu

FIGURE 1. OM: R&D technician Crystal Poole prepares a non-rad test sample to run through the multicollector (left). INL chemist Jeffrey Berg presses test samples (right).

The critical need for accurate nuclear data has been pointed out in recent studies devoted to Generation IV systems. The very high mass actinides can play a significant role in the feasibility assessment of innovative fuel cycles. For example, the build-up of 252 californium (Cf) when recycling all transuranic waste (TRU) in a light water reactor (LWR) leads to increased neutron emissions that could impact the fuel fabrication process. This experiment will generate enough data to validate models and provide better neutron cross-sections for advanced reactor designers for years to come. As a consequence, nuclear data of higher mass transuranics should be significantly improved.

Project Description

The principal goal of this experiment is to irradiate very pure actinide samples in ATR and determine the amounts of resulting transmutation products. Determining nuclide densities before and after neutron irradiation will allow the inference of energy-integrated neutron cross-sections. This type of information, together with neutron cross-section differential measurements, is ultimately used by physicists in charge of nuclear data evaluations for the Evaluated Nuclear Data File (ENDF).

In order to obtain effective neutron capture cross-sections corresponding to different neutron spectra, ranging from fast to epithermal, three sets of actinide samples were irradiated. The first one was filtered with cadmium (Cd), and the other two were filtered with 5-mm and 10-mm thicknesses of boron (B). The determination of atom densities before and after irradiation will be carried out using inductively-coupled plasma mass spectrometry (ICPMS) at the INL's Analytical Lab and the accelerator mass spectrometry (AMS) at the Argonne Tandem Linac Accelerator System (ATLAS) facility.

Creating two different sets of independent measurements will increase confidence in the results.

This project has been funded in part by the DOE Office of Science, in addition to the ATR NSUF, and has been given the name MANTRA. It became an official ATR NSUF project in January 2010.

Accomplishments

The first two irradiations were completed in January 2013. The third and last sample irradiation was completed in January 2014, after two cycles in ATR. Under neutron irradiation, these isotopes transmute into other isotopes, and even though the number of transmutation products at the end of the irradiation is relatively small, it is sufficient to infer the neutron capture cross-section if the measurements are precise enough.

Measurements of isotopic ratios in most of the samples were finalized in 2014 using the Multi-Collector Inductively Coupled Plasma Mass Spectrometer (MS-ICP-MS). This project was very successful, with about 65 samples (initial + irradiated) characterized in a relatively short period of time. All the isotopic ratios of interest have an associated 2-sigma uncertainty of less than 1%.

The measurements showed that after irradiation less than 1% of the initial material was transmuted when the 5-mm B filter was used, whereas up to 25% was transmuted when the Cd filter was used. This was expected, as the capture reaction rates in the resonance region seen by the Cd-filtered samples are higher than those in the faster neutron region seen by the 5-mm B-filtered samples. The detailed ATR as-run calculations using the Monte Carlo N-Particle (MCNP) code proved more challenging than originally thought; however, significant progress was made in 2014. In particular, Jim Sterbentz, the analyst in charge, as well as Idaho State University (ISU) Ph.D. student Jyothier Kumar Nimmagadda, showed that a detailed modeling of the neutron self-shielding found in some of the samples is crucial to reproducing the physical phenomena during irradiation. This will complicate the calculations and will require a more detailed model than was originally planned.

These analyses will be completed in 2015 and will allow us to start interacting with the nuclear data community and, in particular, the evaluators in charge of the nuclear data files. In April 2014, based on the recommendations of INL Fellow Pino Palmiotti, this work was presented to the Organisaton for Economic Co-operation and Development's Nuclear Energy Agency (OECD/NEA) Expert Group on Improvement of Integral Experiment Data for Minor

"This experiment is unique in the sense that it will provide a consistent set of neutron cross-sections in fast and epithermal neutron spectra for most isotopes of interest to reactor physics."

— Gilles Youinou, INL Principal Investigator

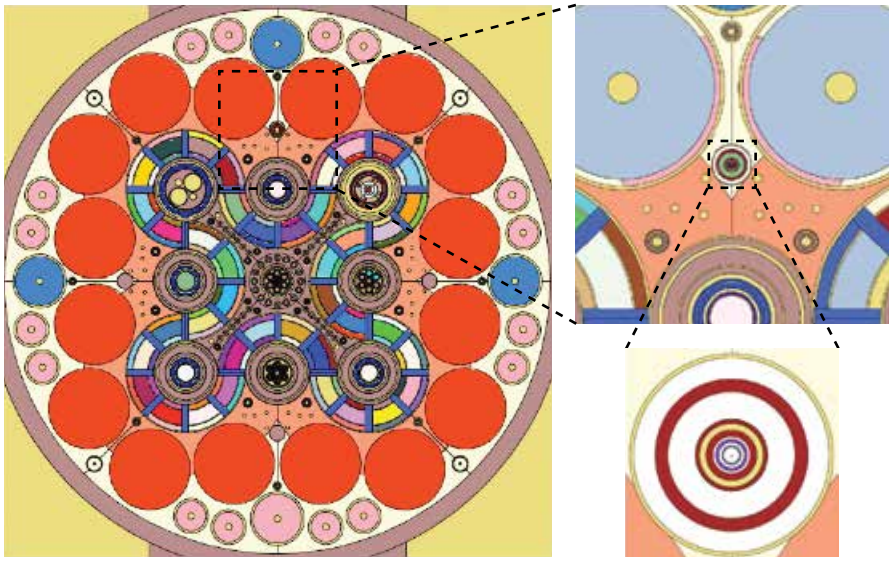


FIGURE 2. Details of the ATR MCNP model.

Actinides Management, where it received very positive feedback. More information was provided at another OECD/NEA meeting in October 2014. A status-of-the-experiment report was also presented at the ATR NSUF Users Meeting in June 2014.

Future Activities

Goals for 2015 include (1) finalizing the MC-ICP-MS measurements, (2) finalizing ATR as-run calculations with MCNP, and (3) preparing samples for researchers to run complementary measurements using the AMS at the ATLAS facility.

Publications and Presentations*

**See additional publications from other years in the Media Library on the NSUF website.*

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
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Collaborators	
Idaho State University	George Imel (principal investigator)
Idaho National Laboratory	Gilles Youinou (principal investigator), Giuseppe "Pino" Palmiotti (collaborator), Tom Maddock (collaborator), Jeff Giglio (collaborator), Theresa Giglio (collaborator), Jeff Berg (collaborator), Jim Sterbentz (collaborator)

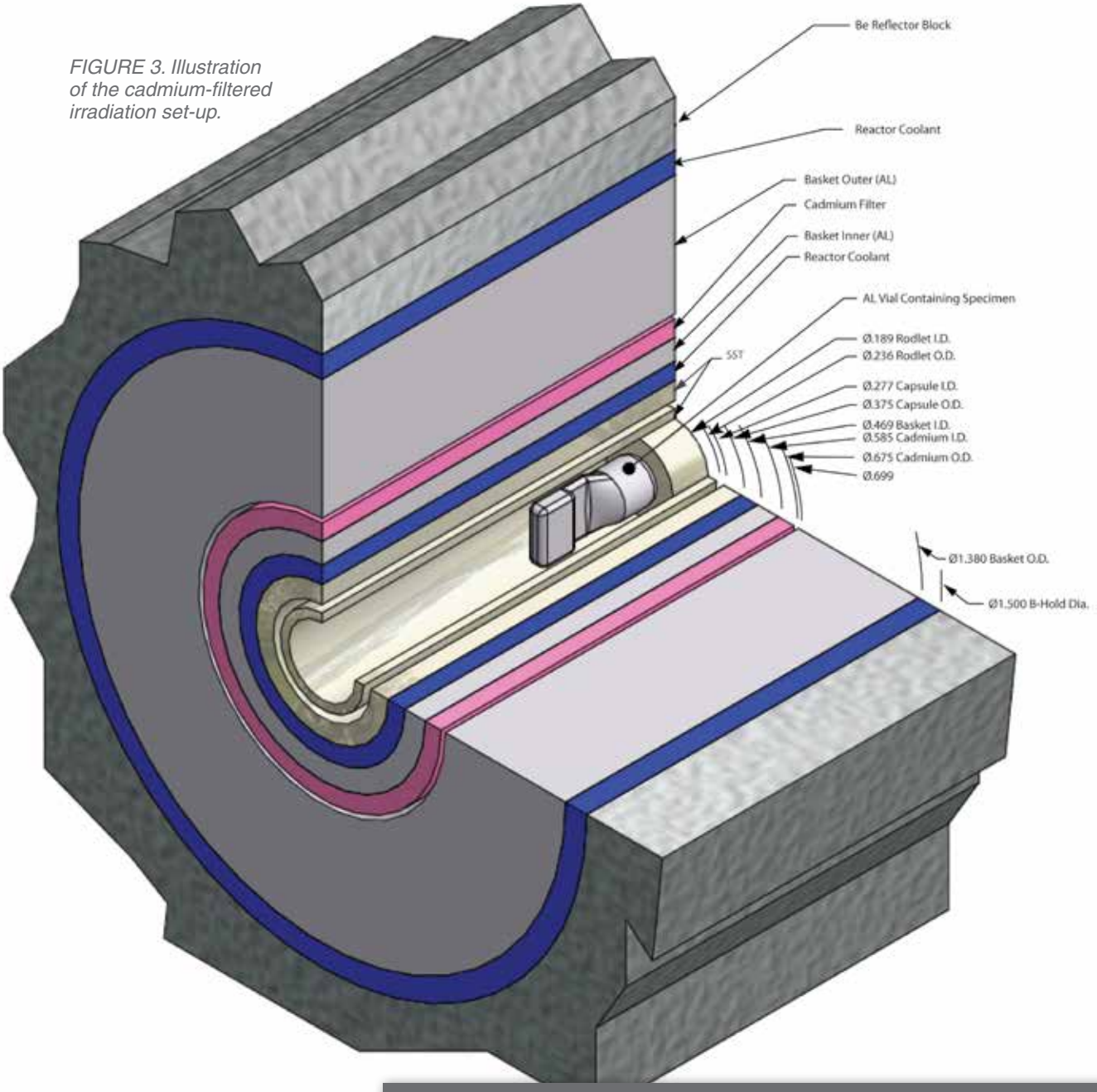


FIGURE 3. Illustration of the cadmium-filtered irradiation set-up.

This experiment will generate enough data to validate models and provide better neutron cross-sections for advanced reactor designers for years to come.

Advanced Damage-Tolerant Ceramics:
Candidates for Nuclear Structural Applications

Michel W. Barsoum – Drexel University – barsoumw@drexel.edu

Robust materials are critical to meet evolving advanced reactor and fuel designs. These materials need to operate in extreme environments of elevated temperatures, corrosive media, and high radiation fluences, with lifetime expectation of greater than 60 years. Full understanding of a material's response to irradiation is paramount to long-term, reliable service. The layered ternary carbides and nitrides known as MAX phases have the potential to be used in the next-generation nuclear reactors. All MAX phases are fully machinable even though some of them, such as Ti_3SiC_2 and Ti_2AlC_2, are similar to titanium metal in density but are three times as stiff. The thermal and electrical conductivities are high and metal-like. They have relatively high fracture toughness values and some are chemically stable in corrosive environments. They also have shown irradiation damage tolerance in heavy ion studies.

The aim of this project is to investigate the damage in Ti_3SiC_2, Ti_3AlC_2 and chemical vapor deposition CVD SiC (for comparison) after exposure to a spectrum of neutron irradiations consistent with conditions found in light water nuclear reactors.

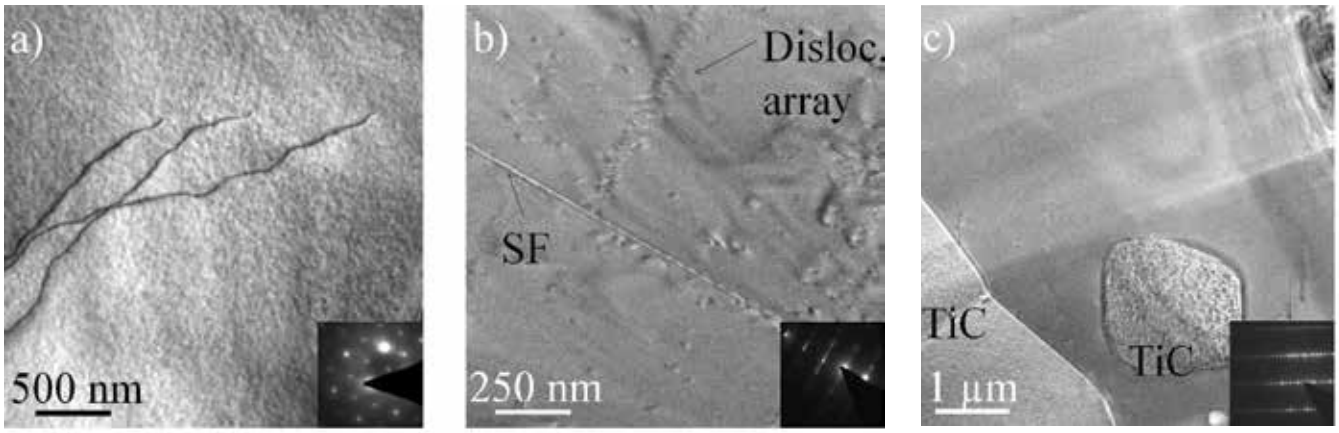


Figure 1. a) Brightfield TEM images taken near the (0001) zone axis of the ATR-Ti_3SiC_2 sample irradiated to 0.1 dpa at 100°C reveals perturbation of the surface and long basal plane dislocations. The mottled surface was likely due to improper FIB cleaning. b) TEM micrograph of ATR-Ti_3SiC_2 sample irradiated to 0.1 dpa at 500°C taken in 2 beam condition near the (11-20) zone axis shows dislocation arrays parallel to the basal plane and stacking faults. Small defects can be seen throughout, but were not confirmed as loops. c) Brightfield TEM micrograph taken on the (11-20) zone of the ATR-Ti_3SiC_2 sample irradiated to 0.1 dpa at 1000°C showing a preexisting TiC particle that was highly damaged - with dislocation loops and black spots - not present in the surrounding MAX matrix, which remains clear of irradiation induced defects.

The carbides are exposed to a series of neutron fluence levels (0.1, 1, and 9 dpa) at moderate to high irradiation temperatures (100, 500, and 1000°C) in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL). The damage to the microstructures and the effects of the radiation on the mechanical and electrical properties of the materials will be characterized during post-irradiation examinations. The results will provide an initial database that can be used to assess the microstructural responses and mechanical performances of these ternaries.

Accomplishments

This project is a collaborative effort between the INL, Savannah River National Lab and Drexel University, and was initiated in 2009. As noted above, the team seeks to characterize the effect of neutron irradiation of select MAX phases for use in nuclear reactor applications. During the program's second year, all irradiated samples were shipped to the PIE facility at INL. Unfortunately, unavoidable delays within INL throughout the FY 2013 continued to delay the project. The receipt, cask unloading, experiment disassembly and cataloging of specimens have finally commenced, led by Collin Knight. Upon opening the capsules, it was observed that in several instances the samples had either fused together, or to the capsule making retrieval difficult. The most troublesome capsules were the ones held at 1000°C for the longest times. What is believed to have happened is that the materials swelled more than anticipated for the designed capsule volume. As many samples were recovered as possible, and all samples have been separated and organized into storage vessels (KGTs).

Throughout FY 2014, Darin Tallman scheduled several trips to INL for PIE characterization using equipment at CAES. Most capsules remain at HFEF, where they are stored and await characterization. PIE characterization commenced in FY 2014 with capsules KGT1367, KGT1369 and KGT1371 corresponding to resistivity bars irradiated to 0.1 dpa at 100, 500 and 1000°C, respectively. These were the most readily available capsules for decontamination and preparation, as they had already been sent to EML for preliminary analysis. Samples were expertly cleaned and prepared by Karen Wright and Collin Knight at EML for shipment to CAES. Due to their high activity readings, the resistivity bars were mounted in

The facilities and capabilities available to me at CAES have been incredibly helpful in advancing my research, and I am looking forward to my future visits.

— Darin J. Tallman, Ph.D. Candidate

The MAX phases, a class of machinable, layered, ternary carbides and nitrides, have great promise for use in the next-generation of nuclear reactors. This is the first time the MAX phases have been neutron irradiated at temperatures as high as those carried out here.

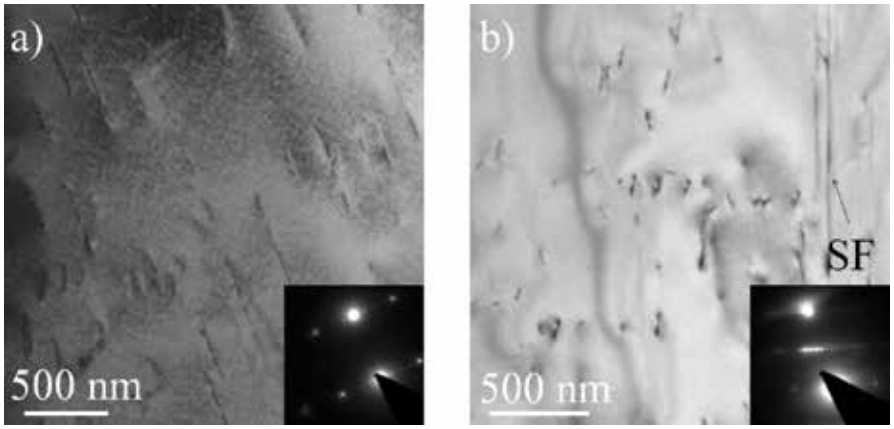


Figure 2. a) Brightfield TEM image near the (11-20) zone axis of ATR-Ti_3AlC_2 sample irradiated to 0.1 dpa at 100°C reveals basal dislocations throughout the sample, some of which may possibly be short dislocation loops. b) TEM micrograph of same sample irradiated to 0.1 dpa at 500°C taken in 2 beam condition tilted away from the (11-20) zone axis shows dislocation arrays parallel to the basal planes, as well as, stacking faults. Several of the dislocations do not appear straight due to the tilt of the sample, they are most likely curved within the basal planes. Further TEM investigation is needed to confirm the presence of dislocation loops or other irradiation induced defects within these materials.

epoxy to limit worker exposure, and the resistivity jig was redesigned. During three-week trips in June and September, Darin collected preliminary TEM results (Figures 1 and 2) and resistivity measurements (Figure 3) for Ti_3SiC_2 and Ti_3AlC_2 from these three capsules. Ti_3AlC_2 irradiated at 1000°C did not survive extraction, and is absent from these results.

FIB prep for these initial samples was poorly executed, and Darin has since been improving on his techniques. TEM micrographs of Ti_3SiC_2 reveal (1) black spots within the basal plane at 100°C, (Fig. 1a), (2) dislocation arrays and stacking faults at 500°C (Fig 1b), and (3) highly damaged preexisting TiC grains next to mostly clean Ti_3SiC_2 grains at 1000°C (Figure 1c). From Fig. 1c alone it is clear that the Si present in Ti_3SiC_2 provides significant irradiation resistance compared to its binary counterpart TiC. TEM micrographs of the Ti_3AlC_2 samples revealed stacking faults and possible dislocation loops at both 100 (Figure 2a) and 500°C, (Figure 2b). More extensive TEM work is necessary for these samples to collect high-resolution TEM micrographs of the defect microstructures, and confirm the presence of loops.

Resistivity measurements were collected using a 4-pt probe technique while applying a constant current of 100 mA. Voltages were collected every 5 seconds for 10 minutes to reach steady state. The resistivity values show more than an order of magnitude increase after irradiation at 100°C, but

after irradiation at 500°C, the resistivity values recovered to values closer to their values for the pristine samples (Fig. 3). These results are consistent with those obtained from our Drexel-MITR NEUP project, recently published in Acta Materialia.

The 9 dpa samples were deemed as high-priority samples to see signs of irradiation damage. However, unrelated laboratory shutdowns led to further delays throughout FY 2014, including limitations on sample shipping within the INL complex. The 9 dpa samples were also mostly broken during retrieval, and were unable to be separated and identified easily. Resources and funding were directed toward the readily available capsules. These issues prevented the high-dose samples from being available for characterization at CAES in 2014.

With the extensive work stoppages preventing ATR sample access at CAES, several TEM samples from our parallel Drexel-MITR NEUP irradiation project were shipped to INL for PIE utilizing ATR funds while Darin was onsite. With the excellent assistance of Lingfeng He, a recently hired research scientist at INL, MITR samples of fine-grained Ti_3SiC_2 and Ti_2AlC were characterized. Results from this work are being prepared for publication in Acta Materialia, which revealed the formation of black spots and defect clusters in Ti_2AlC after irradiation to 0.1 and 0.4 dpa at 360°C (Figs. 4a and b), and dislocation loops and stacking fault formation in both Ti_3SiC_2 and Ti_2AlC

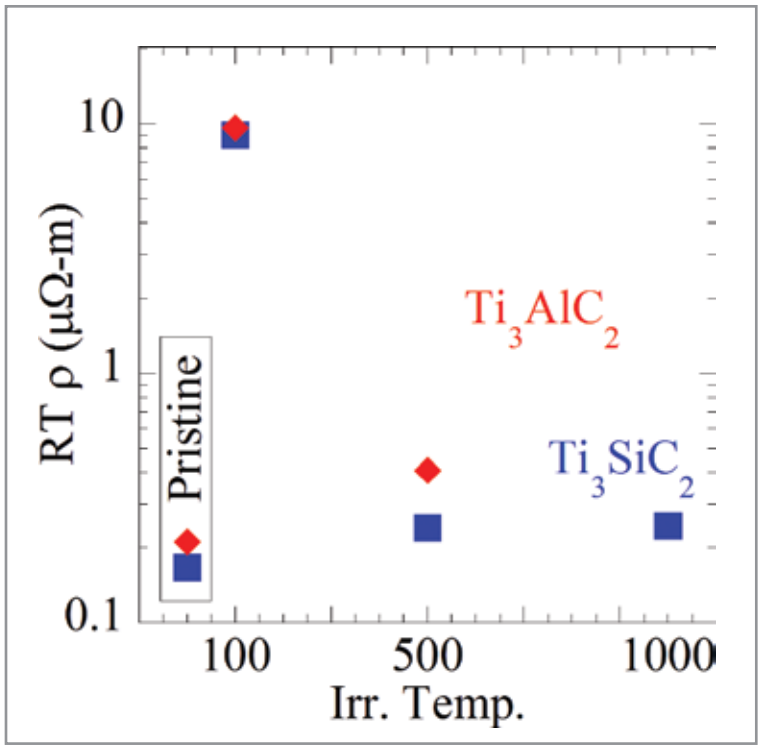


Figure 3. a) Room temperature resistivity plotted on a log scale as a function of neutron irradiation temperature at 0.1 dpa. After irradiation at 100°C, the resistivities of both Ti_3SiC_2 and Ti_3AlC_2 increase more than an order of magnitude. After irradiation at 500 and 1000°C, the resistivity of the Ti_3SiC_2 recovered to values near those of the pristine values. Some recovery is observed in Ti_3AlC_2, with only a 2-fold increase after irradiation at 500°C.

after irradiation up to 0.1 dpa at 700°C, (Fig. 4c and d, respectively). Characterization of both sample sets is ongoing.

Research to be completed

Based on the results obtained thus far with the MAX phase samples irradiated at the MIT reactor, it is anticipated that most of the irradiation damage would have been rapidly annealed out at 1000°C. Comparison with other temperatures is needed to explore the irradiation behavior of these materials. Results to date suggest that Ti_3SiC_2 remains a strong candidate for irradiation application. Darin Tallman is collaborating with Lingfeng He at INL to continue the TEM characterization of both the ATR and MITR sample sets. Darin anticipates graduation in the summer of 2015, thus remaining PIE trips are limited. The high dose, 9 dpa, ATR samples are anticipated to be available by April of 2015, and will be the focus of future characterization. XRD diffractograms of the samples will be obtained once the remaining samples are removed from the capsules, and shipping of samples is allowed within INL.

Distributed Partnership at a Glance

ATR NSUF and Partners	Facilities and Capabilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Idaho National Laboratory	Advanced Test Reactor, Hot Fuel Examination Facility, Analytical Laboratory, or Electron Microscopy Laboratory, Irradiation Assisted Stress Corrosion Cracking Facility
Massachusetts Institute of Technology	Reactor
Collaborators	
Drexel University	Darin J. Tallman (collaborator)
Idaho National Laboratory	Lingfeng He (collaborator)
Savannah River National Laboratory	Elizabeth N. Hoffman (collaborator), Brenda L. Garcia-Diaz (collaborator)

Publications and Presentations

1. D. J. Tallman, E. N. Hoffman, E. N. Caspi, B. L. Garcia-Diaz, G. Kohse, R. L. Sindelar, M. W. Barsoum. 2014, "Effect of Neutron Irradiation on Mn+1AXn Phases," *ICACC14, Daytona Beach, FL, January 29, 2014.*
2. D. J. Tallman, E. N. Hoffman, E. N. Caspi, B. L. Garcia-Diaz, G. Kohse, R. L. Sindelar, M. W. Barsoum, 2014, "Effect of Neutron Irradiation on Mn+1AXn phases," *TMS2014, San Diego, CA, February 17, 2014.*
3. D. J. Tallman, E. N. Hoffman, E. N. Caspi, B. L. Garcia-Diaz, G. Kohse, R. L. Sindelar, M. W. Barsoum, 2014, "Effect of Neutron Irradiation on Select Mn+1AXn phases," *CIMTEC, Montecatini Terme, Italy, June 11, 2014.*
4. D. J. Tallman, E. N. Hoffman, E. N. Caspi, B. L. Garcia-Diaz, G. Kohse, R. L. Sindelar, M. W. Barsoum, 2015, "Effect of neutron irradiation on select MAX phases," *Acta Materialia Vol. 85, pp. 132–143.*

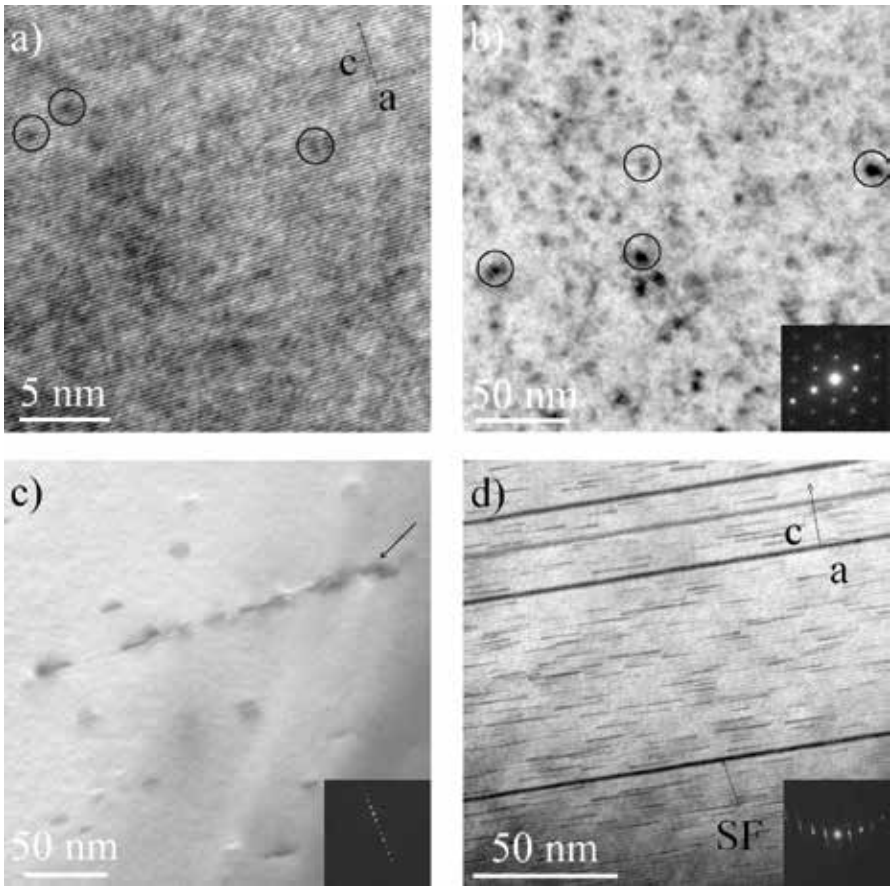


Figure 4: a) Brightfield high resolution transmission electron microscopy (HRTEM) micrograph of an edge on grain of a MITR-Ti_2AlC sample irradiated to 0.1 dpa at 360°C reveals perturbation of the basal planes and the presence of defect clusters and/or black spots (circled). b) TEM micrograph of a MITR-Ti_2AlC sample irradiated to 0.4 dpa at 360°C taken in 2 beam condition near the (0001) zone axis shows a higher density of small black spots (circled). c) Brightfield TEM micrograph of a fine-grained MITR-Ti_3SiC_2 sample irradiated to 0.1 dpa at 695°C shows dislocation loops imaged near the (11 -20) zone axis, parallel to the basal plane, agglomerating near a stacking fault (black arrow). The loops are seen as black and white lobes due to strain induced in the surrounding lattice, with an average defect density of 3.94 x 1021 loops/m_3. d) Brightfield TEM micrographs of MITR-Ti_2AlC irradiated to 0.1 dpa at 695°C taken near the (11 -20) zone axis reveal dislocation loops edge on within the basal planes, with a higher average density of 1.1 x 1023 loops/m^3.

Low Fluence Behavior of Metallic Fuels

Yongho Sohn – University of Central Florida (UCF) – yongho.sohn@ucf.edu

Low fluence experiments in metallic fuels, specifically uranium-zirconium (U-Zr) and uranium-molybdenum (U-Mo) types, have a relevance to both the Advanced Fuel Cycle Initiative (AFCI) and the Reduced Enrichment for Research and Test Reactors (RERTR) program. Quantified findings for the low fluence behavior of metallic fuels will help us understand fuel performance in thermal and irradiation fields.

Project Description

The objectives of this program are to understand (1) the microstructural evolution of these fuels as a function of temperature, fluence and composition; and (2) diffusion-related phenomena in the fuels and cladding, also as a function of temperature, fluence and composition. Findings from these experiments will explain early microstructural development and mechanisms in detail, as well as provide critical data for models under development in both programs.

Near-term, critical results from this project will support AFCI modeling work on constituent redistribution in irradiated uranium-prasecoymium-zirconium (U-Pr-Zr) fuels that is currently being undertaken by collaborators on this team. These results will also be used to improve the accuracy of computer models in the RERTR program that predict the overall swelling behavior of the fuel.

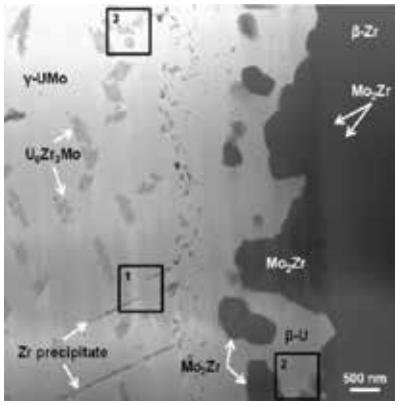
Accomplishments

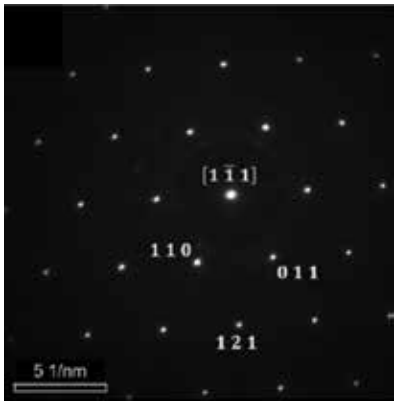
The design of these experiments, along with the necessary quality control documents from University of Central Florida (UCF) researchers and INL scientists, was finalized at a meeting at UCF during FY 2014. Meanwhile, UCF researchers have continued to pave the way to a better understanding of thermal behavior without irradiation (see graphics). Published technical accomplishments from this independent work are listed below in Publications and Presentations.

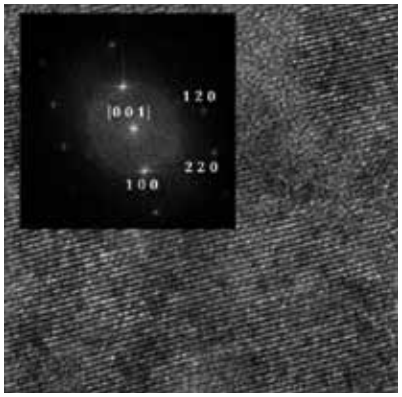
Future Activities

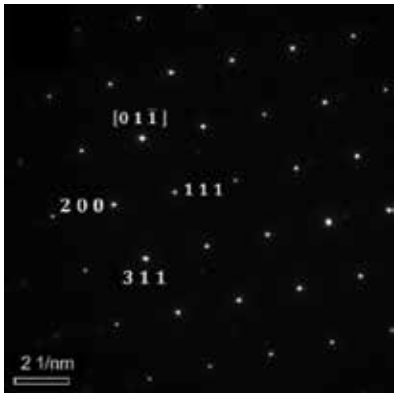
During 2015, work on specimen preparation with alloy casting is planned at INL. Upon receiving the alloys, UCF will produce the samples to be inserted into the ATR. UCF will also continue to document the thermal behavior of alloys and diffusion couples so that, upon completion of ATR experiments, the effects of radiation can be elucidated.

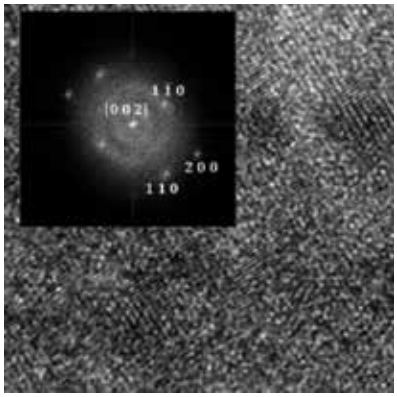
Quantified findings of low fluence behavior in metallic fuels will help us understand fuel performance under thermal and irradiation fields.











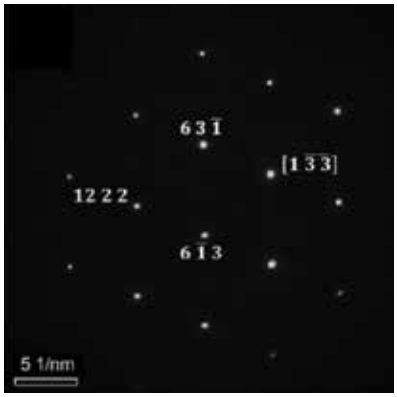


Figure 1. High angle annular dark field STEM micrographs from the water-quenched U-10 wt.% Mo vs. Zr diffusion couple annealed at 650°C for 720 hours. The three squares indicate sampling positions of the (1) α-Zr, (2) Mo_2Zr, β-U, and β-Zr, and (3) U_6Zr_3Mo by selected area electron diffraction and high-resolution TEM with fast Fourier transformation analyses.

"My interaction has been truly rewarding, giving me an opportunity to strengthen my fundamentals of science, hone my hands-on laboratory skills, and gain greater perspective on energy research with respect to application and societal impact. It is a great program in which graduate students can cover the entire spectrum of R&D."

— Ashley Paz y Puente (formerly Ashley Ewh), graduate student, University of Central Florida

Publications and Presentations*

1. Y. Park, D.D. Keiser, Jr., Y.H. Sohn, 2015, "Interdiffusion and reaction between U–Mo And Zr at 650°C as a function of time," *Journal of Nuclear Materials*, Vol. 456, pp. 351-358.
2. Y. Park, K. Huang, A. Paz y Puente, H.S. Lee, B.H. Sencer, J.R. Kennedy, Y.H. Sohn, 2015, "Diffusional Interaction Between U–10wt.%Zr and Fe at 903K, 923K and 953K (630°C, 650°C, and 680°C)," *Metallurgical and Materials Transactions A*, Vol. 46A, pp. 72–82.
3. K. Huang, Y. Park, L. Zhou, K.R. Coffey, Y.H. Sohn, B.H. Sencer, J.R. Kennedy, 2014, "Effects of Cr and Ni on interdiffusion and reaction between U and Fe–Cr–Ni alloys," *Journal of Nuclear Materials*, Vol. 451, pp. 372–378.
4. A. Paz y Puente, J. Dickson, D.D. Keiser, Jr., Y.H. Sohn, 2014, "Investigation of interdiffusion behavior in the Mo–Zr binary system via diffusion couple studies," *International Journal of Refractory Metals and Hard Materials*, Vol. 43, pp. 317–321.
5. J. Dickson, L. Zhou, A. Paz y Puente, M. Fu, D.D. Keiser, Jr., Y.H. Sohn, 2014, "Interdiffusion and reaction between Zr And Al, Al-2wt.%Si, Al-5wt.%Si, or 6061 from 425° to 625°C," *Intermetallics*, Vol. 49, pp. 154–162.
6. Y. Park, J. Yoo, K. Huang, D.D. Keiser, Jr., J.F. Jue, B. Rabin, G. Moore, Y.H. Sohn, 2014, "Growth kinetics and microstructural evolution during hot isostatic pressing of U-10wt.%Mo monolithic fuel plate in AA6061 cladding with Zr diffusion barrier," *Journal of Nuclear Materials*, Vol. 447, pp. 215–224.
7. K. Huang, C. Kammerer, D.D. Keiser, Jr., Y.H. Sohn, 2014, "Diffusion Barrier Selection from Refractory Metals (Zr, Mo and Nb) Via Interdiffusion Investigation For U-Mo RERTR Fuel Alloy," *Journal of Phase Equilibria and Diffusion*, Vol. 35, pp. 146–156.

**See additional publications from other years in the Media Library on the NSUF website.*

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	Advanced Test Reactor
Collaborators	
University of Central Florida	Yongho Sohn (principal investigator), Youngjoo Park (graduate student), Ryan Newell (graduate student), Esin Geller (graduate student), Nicholas Eriksson (graduate student), Felipe Betanco (undergraduate student)
Idaho National Laboratory	Maria Okuniewski (co-principal investigator), Dennis D. Keiser, Jr. (co-principal investigator)

Radiation-Induced Segregation/Depletion at Grain Boundaries in Neutron-Irradiated 304SS at Low Dose Rates

Emmanuelle Marquis – University of Michigan – emarq@umich.edu

Within the light water reactor susceptibility program, understanding the microstructures developing in austenitic stainless steels under very low dose rates is essential to ensuring reliable predictions.

Radiation-induced segregation/depletion and its deleterious impact on properties in austenitic stainless steels have been studied extensively, particularly at high dose rates (typically >10-4dpa/s). However, validation of life-extension plans for light water reactors and future applications in advanced fission and fusion reactors require input data on the effects of high fluence obtained at low dose rates on the microstructure and mechanical properties of these steels.

Project Description

Irradiating the 304 stainless steel (304SS) hex-blocks in the Experimental Breeder Reactor (EBR)-II fast reactor to relatively high doses at low dose rates provides an opportunity to investigate the irradiation-induced microstructures. This was performed using transmission electron microscopy (TEM) and atom probe tomography (APT) both at CAES and at the University of Michigan. The objectives of the work were to:

- Understand the synergy or competition between radiation-induced segregation, carbide formation, and swelling as function of dose rate, dose and temperature.
- Understand the microstructural changes induced near the sodium-wetted surfaces and their consequences on radiation-induced segregation at grain boundaries.
- Benchmark the APT measurements by comparing them to information obtained by analytical electron microscopy.
- Develop a mechanistic understanding for the observed changes.

Accomplishments

After a delayed start of over two years due to technical issues regarding materials selection, availability, and preparation, the project progressed significantly thanks to the involvement of Bulent Sencer at INL. The microstructures of two hex-blocks irradiated at different dose rates and doses were characterized in great detail, including voids, nickel (Ni)-silicon (Si) clusters, dislocation loop density, loop chemistry, phosphide precipitates, grain boundary chemistry, grain boundary carbides, and surface chemistry. Selected examples of these observations are illustrated in Figure 1.

Future Activities

All the objectives have now been addressed. University of Michigan graduate student Yan Dong will be presenting the results at The Minerals, Metals and Materials Science (TMS) conference in 2015, and two publications are in preparation for submission to the Journal of Nuclear Materials.

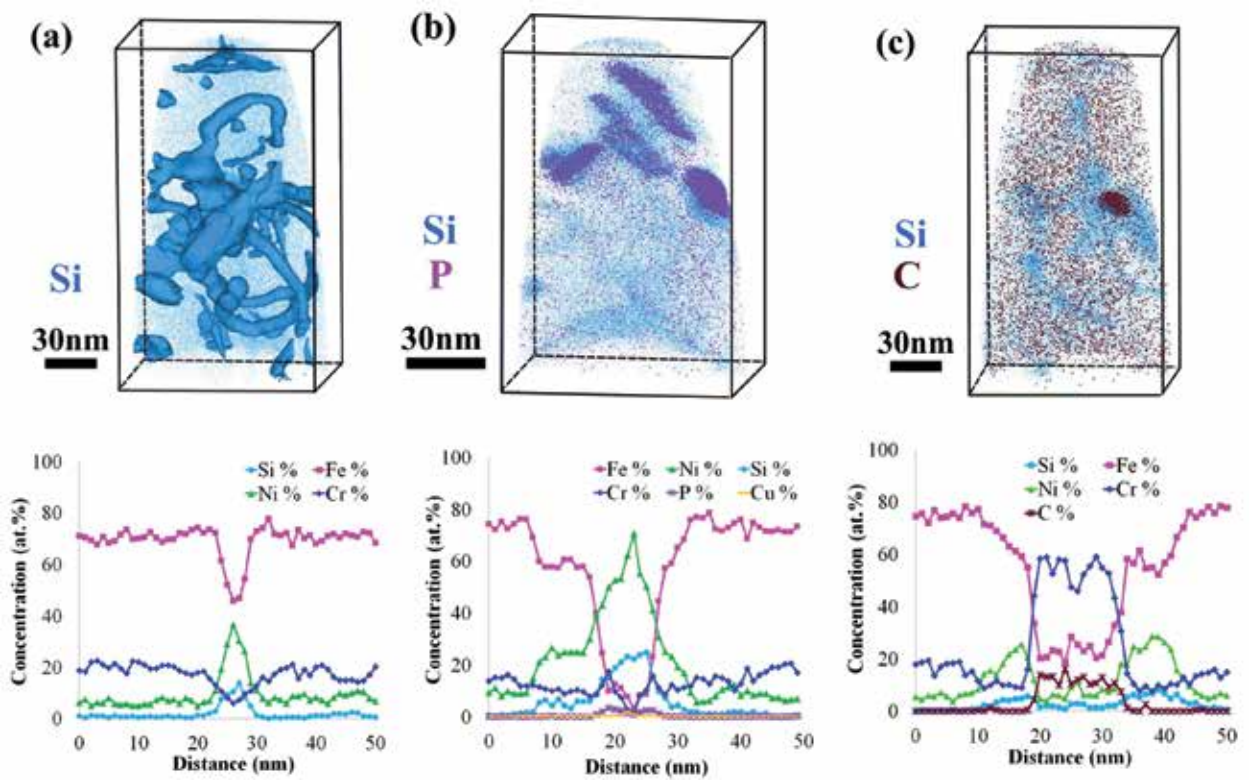


Figure 1. 3D reconstructions highlighting a) the network of dislocation loops segregated with Si and Ni as indicated by the concentration profile underneath. b) Phosphide plates with Ni and Si segregating to the plate interface, and c) small carbide and Ni_3Si precipitates with Ni and Si segregating to the carbide interface.

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
University of Michigan	Emmanuelle Marquis (principal investigator), Yan Dong (collaborator)
Idaho National Laboratory	Bulent Sencer (collaborator)
Consultant	Frank Garner

Multiscale Investigation of the Influence of Grain Boundary Character on Radiation-induced Segregation and Mechanical Behavior in Steels Used in Light Water Reactors

Mitra Taheri – Drexel University – mtaheri@coe.drexel.edu

The research provides direct quantification of the responses of grain boundary, character-dependent stainless steel under LWR irradiation.

Project Description

This project is centered on understanding the behavior of 304 and 316 stainless steels (304SS, 316SS) in light water reactors (LWR). The primary objective is to understand and quantify any microstructural evolution taking place related to long-term aging (thermal effects) and any irradiation-induced or enhanced solute segregation and precipitation occurring in-grain or at the grain boundaries.

A second objective is to understand the role of grain boundary character in low-stacking-fault, face-centered-cubic (fcc) stainless steels, including any dependence they exhibit on irradiation-enhanced or -induced grain boundary solute segregation, under long-term irradiation.

The research provides, through advanced electron and atom probe microscopy, direct quantification of grain boundary, character-dependent stainless steel irradiation responses during long-term aging in current LWRs.

Accomplishments

Drexel University's technical objective in the project was to investigate the role of grain boundary character in radiation-induced segregation and precipitation in fcc austenitic stainless steel. A 304SS from the Experimental Breeder Reactor II (EBR-II) outer-blanket assembly, Row 13, was previously irradiated to a peak fluence of ~4.5×10^{21} n/cm^2. High-quality, electro-polished bulk samples were prepared at INL's Hot Fuel Examination Facility (HFEF) for end-user examination at CAES using advanced microscopy techniques. These techniques included electron backscatter diffraction (EBSD), transmission electron microscopy – bright field (TEM-BF), scanning transmission electron microscopy – energy dispersive x-ray spectroscopy (STEM-EDS), and atom probe tomography (APT), all of which are capable of probing any grain boundary character dependence on multiple-length scales, from mesoscale grain boundary structure to atomic scale solute segregation.

The 304SS (U1302) sample was a hexagonal duct strip from the outer-blanket assembly irradiated

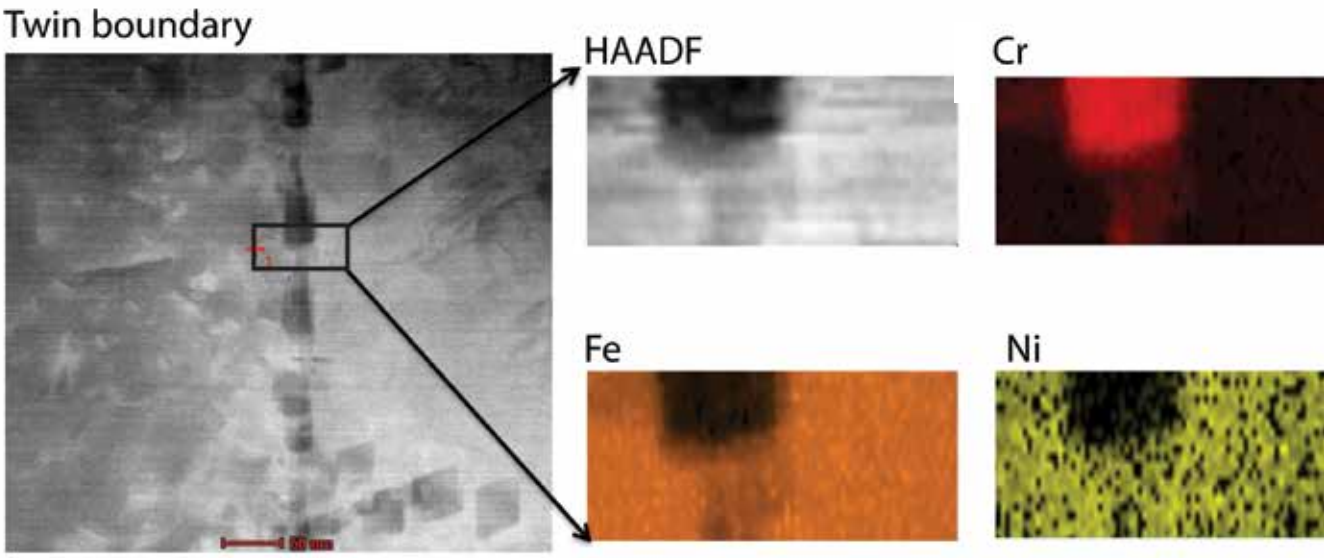


Figure 1. STEM and STEM-EDS results from a twin Σ3 grain boundary highlighting characteristic Cr rich $M_{23}C_6$ carbides along the examined GB length.

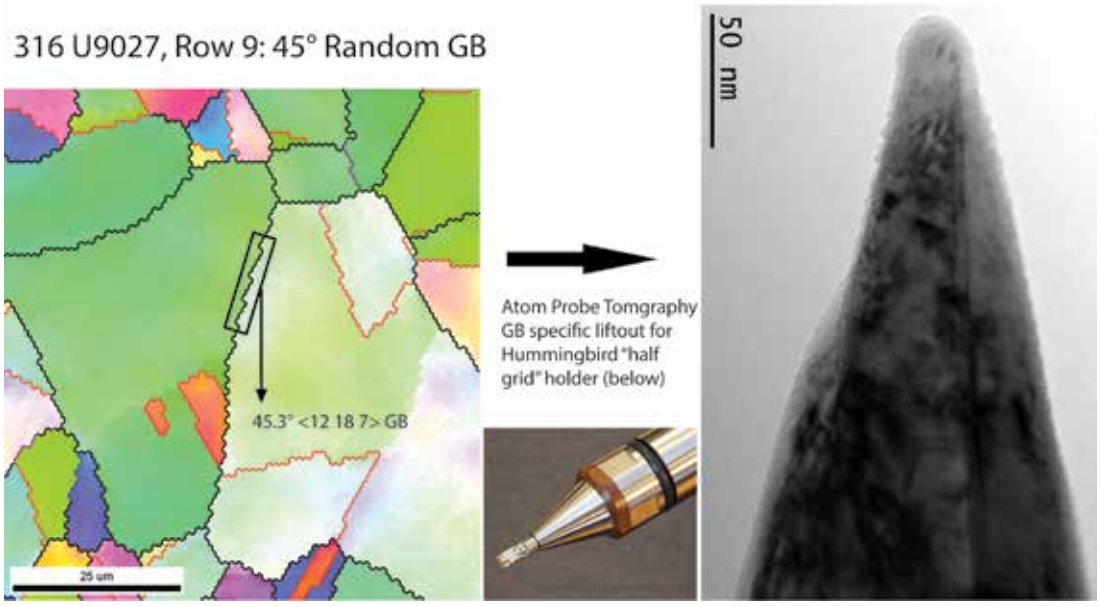


Figure 2. Highlight of EBSD map indicating a 45-degree grain boundary that is then thinned for a TEM and APT correlated study.

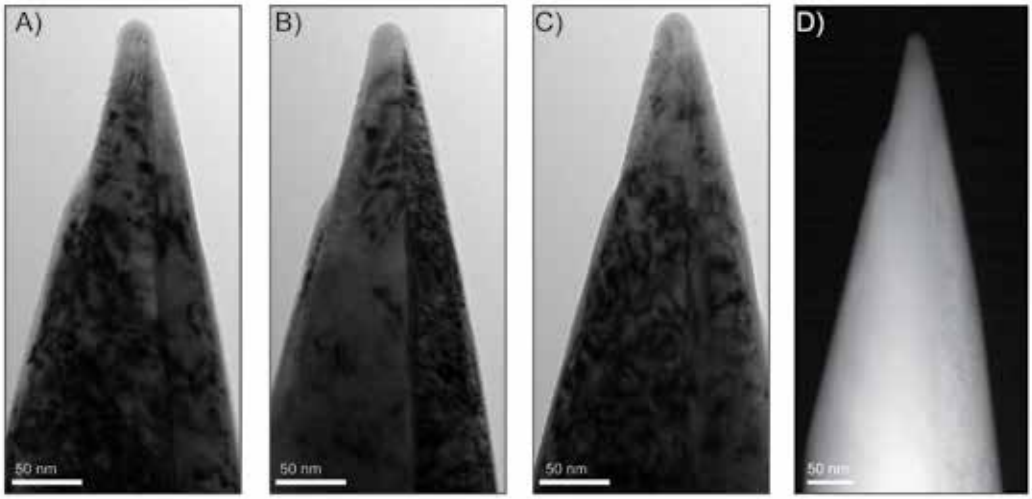


Figure 3. Sequence of three BF-TEM diffraction conditions (A-C) and STEM-HAADF (D) of the same GB structure shown in Figure 2.

in Row 13, Position 13D4. Over the course of the EBR-II irradiations (Run 1 to Run 170B), the 304SS was exposed to extended periods of high operating temperatures (450°–460°C). Researchers analyzed samples using a using multi-length scale, grain-boundary, site-specific process optimized to obtain detailed chemical and structural information. Specifically, they performed EBSD to determine particular grain boundary misorientations and used an FIB to extract specific grain boundaries. These samples were analyzed using both TEM and APT.

The study focused on the following grain boundary characters: coincidence site lattice (CSL) three coherent twin; three incoherent twin; and random, high-angle misorientation grain boundaries. Researchers focused on the three system due to the differences observed between the atomic structures of the coherent twins having a {111} symmetric tilt grain boundary plane and the incoherent twin having a {112} symmetric tilt grain boundary plane. This grain boundary examination system allows the careful study of the effects of the grain boundary plane inclination angle with respect to irradiation-induced precipitation and segregation.

The goal was accomplished, and all three grain boundary types were examined using both APT and STEM-EDS. The results indicated that each grain boundary type had extensive carbide ($M^{23}C^6$) precipitation. The presence of carbide nucleation and growth on the coherent twin grain boundary (Figure 1) highlights the fact that even low grain-boundary-energy and high atomic-fit grain boundary characters can be susceptible to carbide growth when subjected to extensive irradiation at relatively high temperatures (450°–460°C). The APT examination showed that depletion of Cr to less than 10 at.% existed in regions adjacent to the grain boundary carbide. These Cr-depleted zones are typical of grain boundary carbide formations and indicate regions that are potentially susceptible to increases in localized corrosion of aging stainless steel alloys.

The second sample condition the project focused on was a 316 hex-duct stainless steel from location U9027 (Row 9) in the EBR-II. Data collection on the exact reactor temperature and fluence is ongoing but they are estimated to be 370°–500°C and ~20–25 displacements per atom (dpa), respectively. Unlike the 304SS detailed above, APT revealed that the 316SS had extensive nickel-silicon (Ni-Si) clusters throughout the examined regions. The grain-boundary microchemistry, independent of grain-boundary character, showed areas of both carbides and extensive W-shaped Cr depletion. Figures 2-4 show the steps researchers performed on the grain-boundary structures at CAES to fully characterize a particular grain boundary:

EBSD → hummingbird half-grid TEM holder → STEM and TEM → atom probe.

Overall, like the 304SS, the 316SS has grain-boundary-depleted regions of Cr and other minor solute elements. Unlike the examined outer blanket assembly in 304SS, the 316SS has extensive Ni-Si clusters at levels consistent with the presence of Ni_3Si precipitates and/or possible Si enrichment in dislocation arrays.

Future Activities

The project was completed in 2014.

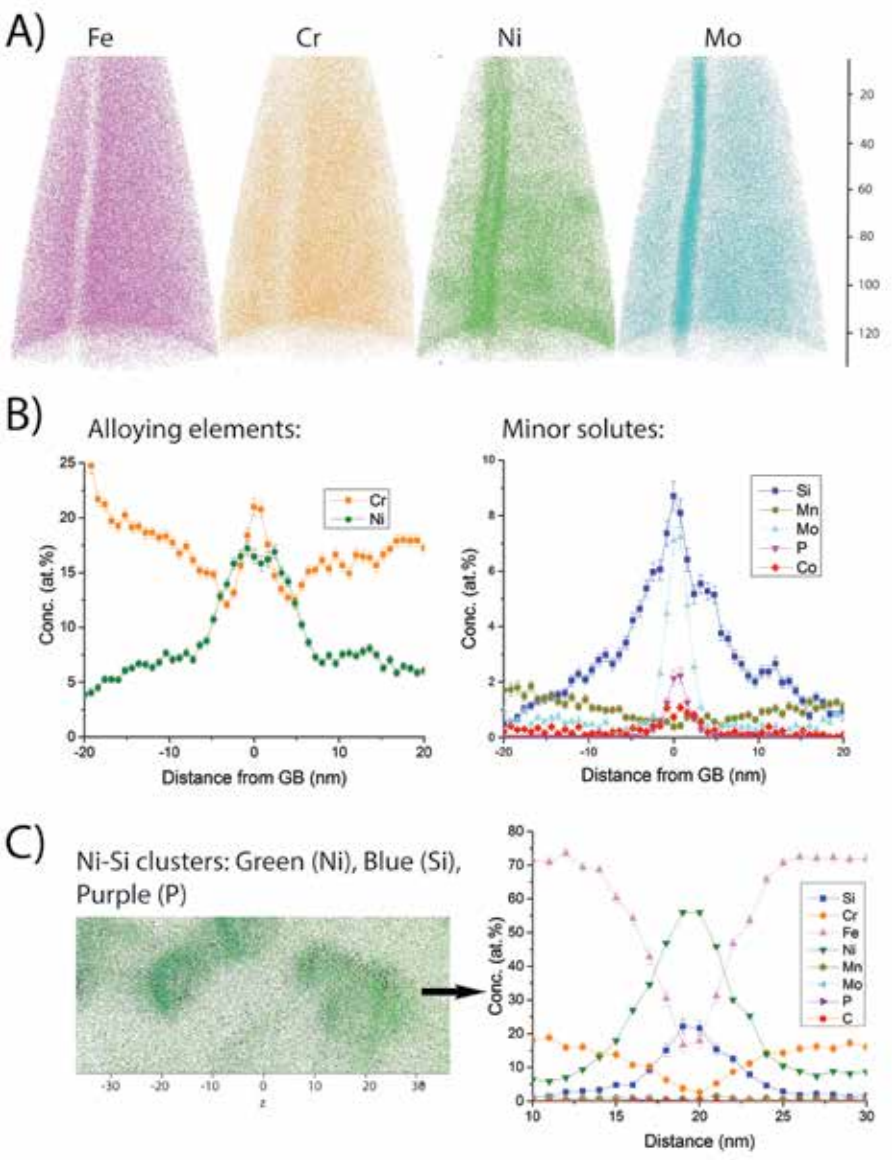


Figure 4. (A) Sequence of atom probe tomography atom map for Fe, Cr, Ni, Mo, (B) highlighted GB microchemistry including Cr depletion with "W" profile, and (C) concentration profile for Ni-Si enriched clusters.

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	PIE facilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
Drexel University	Mitra Taheri (principal investigator), Christopher Barr (co-principal inverstigaator)
Idaho National Laboratory	Jim Cole (principal investigator)

Transducers for In-Pile Ultrasonic Measurement of the Evolution of Fuels and Materials

Bernhard Tittmann – Pennsylvania State University – brt4@psu.edu

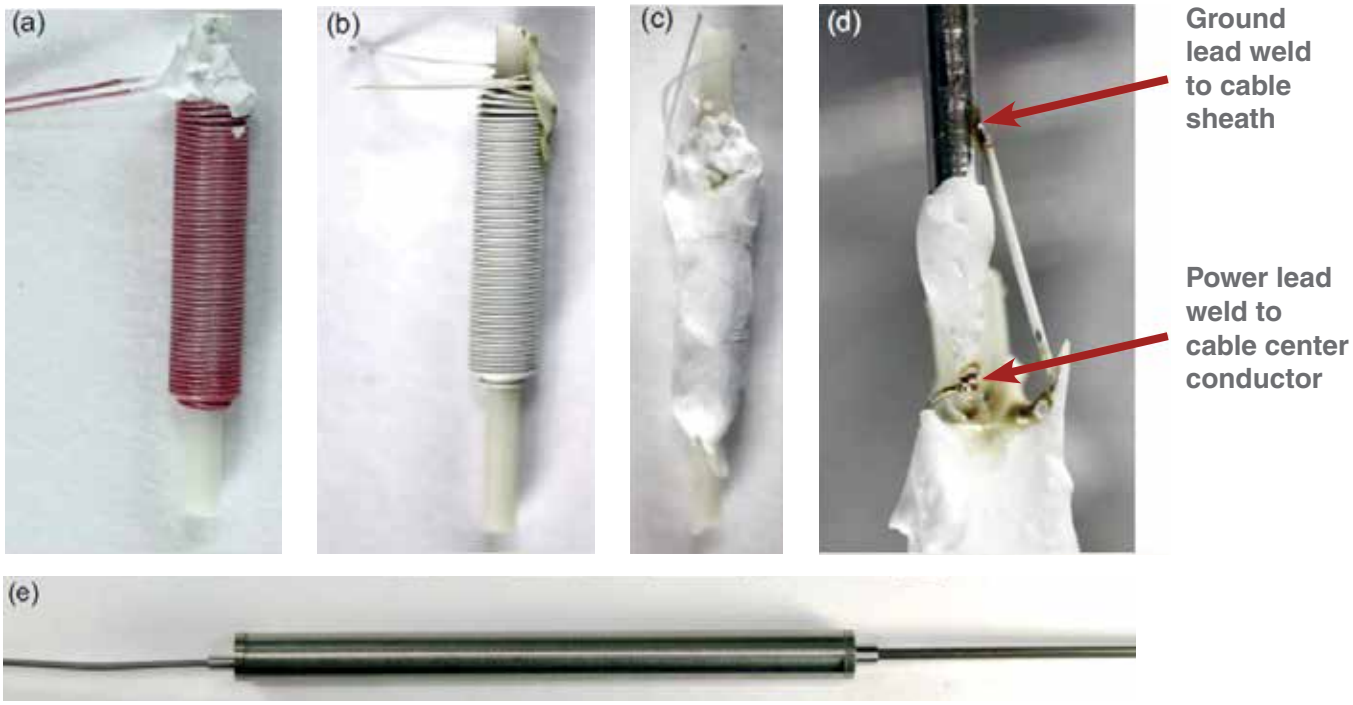


Figure 1. Magnetostrictive transducer fabrication. (a) Silver-palladium is wrapped around an alumina bobbin; (b) the wire is coated with a standoff insulation and heat treated; (c) the wire is coated with an alumina cement and heat-treated a second time; (d) leads are laser-welded to the coax cable and (e) welded into a pre-fabricated housing.

Project Description

Pennsylvania State University (PSU) was awarded an ATR NSUF program to insert both magnetostrictive and piezoelectric transducers into the Massachusetts Institute of Technology Research Reactor (MITR) for radiation at fluences up to 1021 n/cm^2.

Ultrasonic measurements of the transducers are being taken in-situ as the transducers are subjected to the neutron flux. The goals of the experiment are to develop a test design, including selection criteria for candidate materials and optimizing test assembly parameters; obtain data from both out-of-pile and in-pile tests conducted at elevated temperatures; and compare the accrued data with the expected performance of ultrasonic devices under irradiation conditions.

These experiments will enable researchers to design ultrasonic sensors for characterizing the evolution of fuel materials and other nuclear reactor components. The sensors will allow in-situ measurements of material properties and will lead to the development of tools that will monitor the structural health of reactors.

Accomplishments

In 2014, the following progress was made on the project.

The following piezoelectric transducer materials were selected: aluminum nitride (AlN), zinc oxide (ZnO) and bismuth titanate (BiT). Remendur and Galfenol were selected as magnetostrictive transducers.

Assemblies for the magnetostrictive transducers were designed, built and tested (Figures 1 and 2). Transducer performance was characterized at design temperatures (Figures 3–5).

The ZnO transducer did not perform as expected and will only operate successfully during reactor shutdowns (Figure 5). One of ZnO transducers was replaced with an AlN transducer, which operated successfully at the design temperature.

Graduate students Brian Reinhardt (Ph.D.) and Andy Suprock (M.S.) were trained on high-temperature, ultrasonic, nondestructive testing; transducer fabrication principles; and the design of high-temperature, radiation-tolerant transducers.

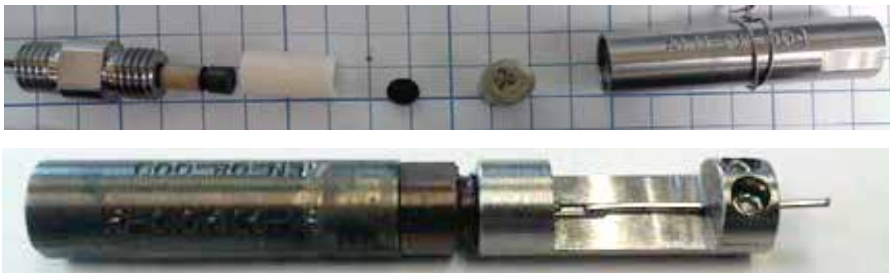


Figure 2. Piezoelectric transducer fabrication. (Top) Transducer components from left to right: stainless steel cap, alumina insulation, nickel plunger, alumina insulation, carbon-carbon backing, Kovar waveguide with ZnO sensor on top, stainless steel outer casing. (Bottom) A fully assembled transducer housing with cable and strain-relief sleeve to support the cable connection.

Results of first power cycle

The first power cycle commenced when the reactor had been operating for 180 hours and lasted until 980 hours of operation. During this time, the reactor was operating at 5 MW. To allow the sensor temperature to rise slowly, the reactor power had been brought to 5 MW incrementally over a period of one week (Figure 6).

The ZnO transducer experienced an electrical malfunction during the insertion process. One of the AlN transducers developed an electrical short during reactor startup, when the temperature reached 400°C.

During the first power cycle, A-scans—or amplitude time series—were periodically collected from the remaining AlN and BiT transducers as well as from the two magnetostrictive transducers. The pulse echo-amplitude was determined by windowing the first returned echo and measuring the amplitude of the fundamental frequency component

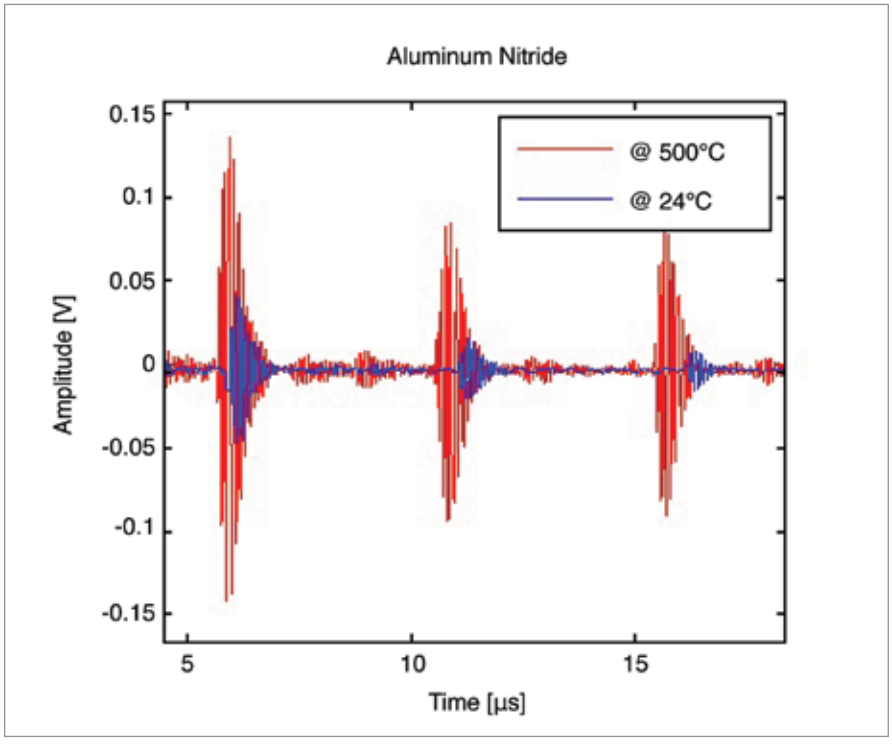


Figure 3. AlN ultrasonic pulse-echo amplitudes recorded at room temperature and at the irradiation temperature.

using fast Fourier transform (FFT). The results of this analysis are shown in Figures 7–9. Figure 7 shows the pulse-echo amplitude measured from the surviving AlN sensor. The plot was normalized to the pre-irradiation amplitude. The pulse-echo amplitude varies by +/- 20% during irradiation.

Figure 8 shows the pulse-echo amplitude measured from the BiT sensor. During the first cycle, the pulse-echo amplitude decreased by approximately 65%. Figure 9 shows the pulse-echo amplitude of the magnetostrictive transducers. These plots were also normalized to the pre-irradiated pulse-echo amplitude. The transient behavior in these sensors seems to be less pronounced as the pulse-echo amplitude only varies by about +/- 10%.

To gauge the material's practical use in harsh radiation environments, the selection criteria of piezoelectric materials for nondestructive evaluation (NDE) and material characterization were analyzed. Piezoelectric AlN was observed to be a viable candidate material. Test results on transducers based on AlN, BiT, Remendur and Galfenol operating in a nuclear reactor within a 40-day window at a fast-neutron flux of 4.05 × 1013 n/cm^2 and a gamma dose rate of 1 × 109 r/hr were also evaluated.

In each case, clear A-Scan measurements were taken at the end of the power cycle. Remendur, Galfenol, and AlN seemed to maintain their initial transduction efficiencies. The pulse-echo amplitude of the AlN sensors varied by +/- 20%, while that of the Remendur and Galfenol varied by +/10%. Conversely, by the end of the first power cycle, the BiT pulse-echo amplitude had decreased by 65%.

The data shows both piezoelectric and magnetostrictive transducers hold promise for use in high-neutron-flux environments. Each shows potential for improving reactor safety and furthering the understanding of the effects of radiation on materials by enabling researchers to monitor a material's structural health and NDE even in the

The in-pile use of ultrasonic transducers during irradiations at MITR is extremely important, because they could provide more accurate, higher-resolution data on the performance of candidate fuels and materials exposed to the harsh conditions of irradiation testing.

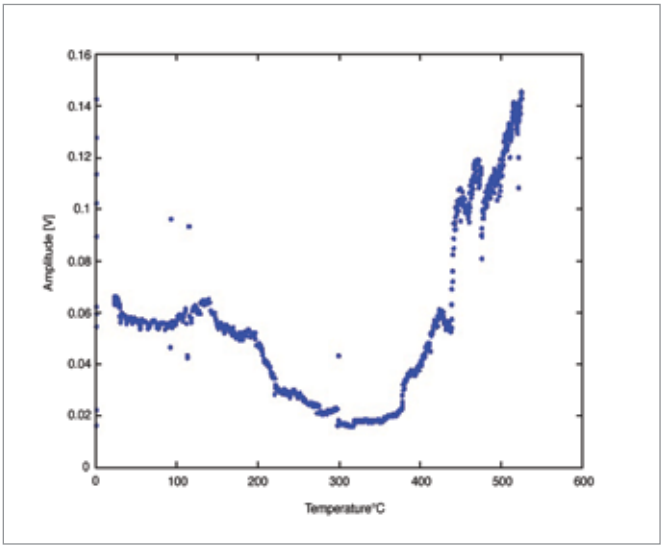


Figure 4. Temperature-dependent performance of an AlN piezoelectric transducer. The transducers operated at their highest efficiencies at the design temperature.

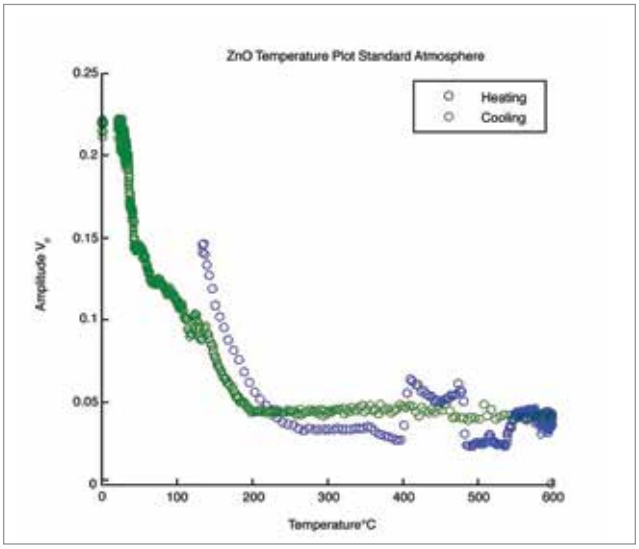


Figure 5. Temperature-dependent performance of the ZnO transducer. The transducer did not perform well at the design temperature, however it recovered its original amplitude upon cooling. This indicates that the sensor will operate satisfactorily during reactor shutdown, enabling us to measure performance degradation at those times.

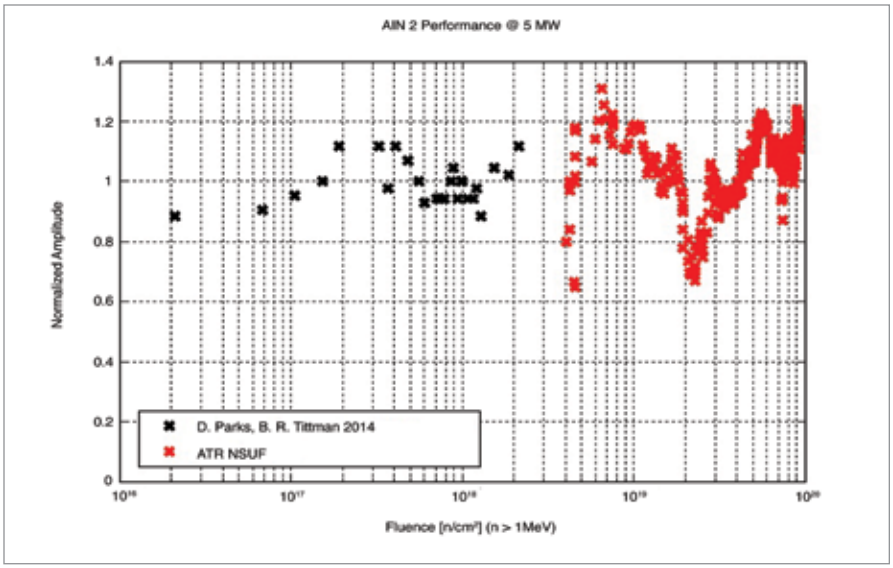


Figure 6. The blue curve indicates the integrated neutron flux (fluence) for neutrons with energy greater than 1 MeV. The green curve represents the reactor power.

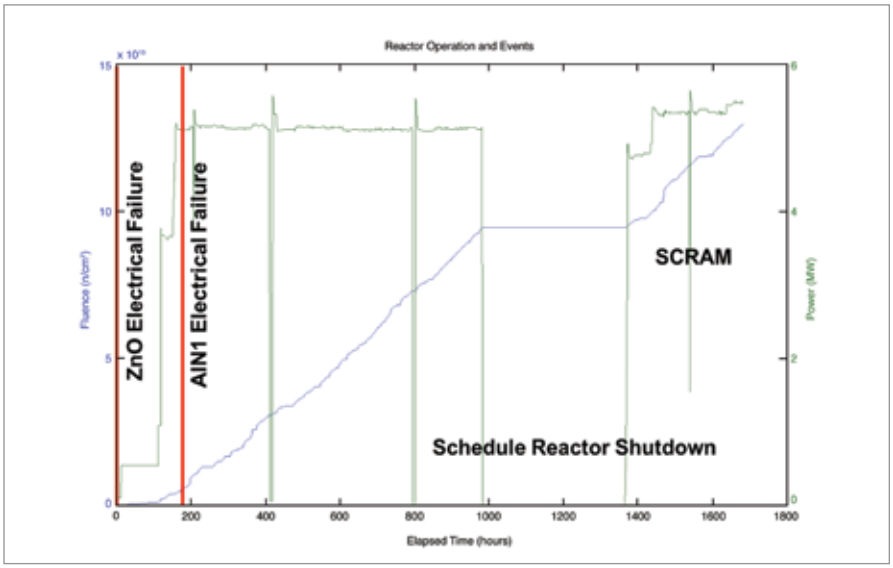


Figure 7. A plot of the pulse-echo amplitude measured by the remaining AlN transducer. The amplitude was normalized to the first measurement made at "0" fluence. The plotted data (red) applies only to the first power cycle and is comparable to the data collected by Parks and Tittmann [2014] (black).

"These experiments will enable researchers to design ultrasonic sensors for characterizing the evolution of fuel materials and other nuclear reactor components. The sensors will allow in-situ measurements of material properties and will lead to tools for structural health monitoring."

— Bernhard Tittmann, Schell Professor and Professor of Engineering Science and Mechanics, Pennsylvania State University

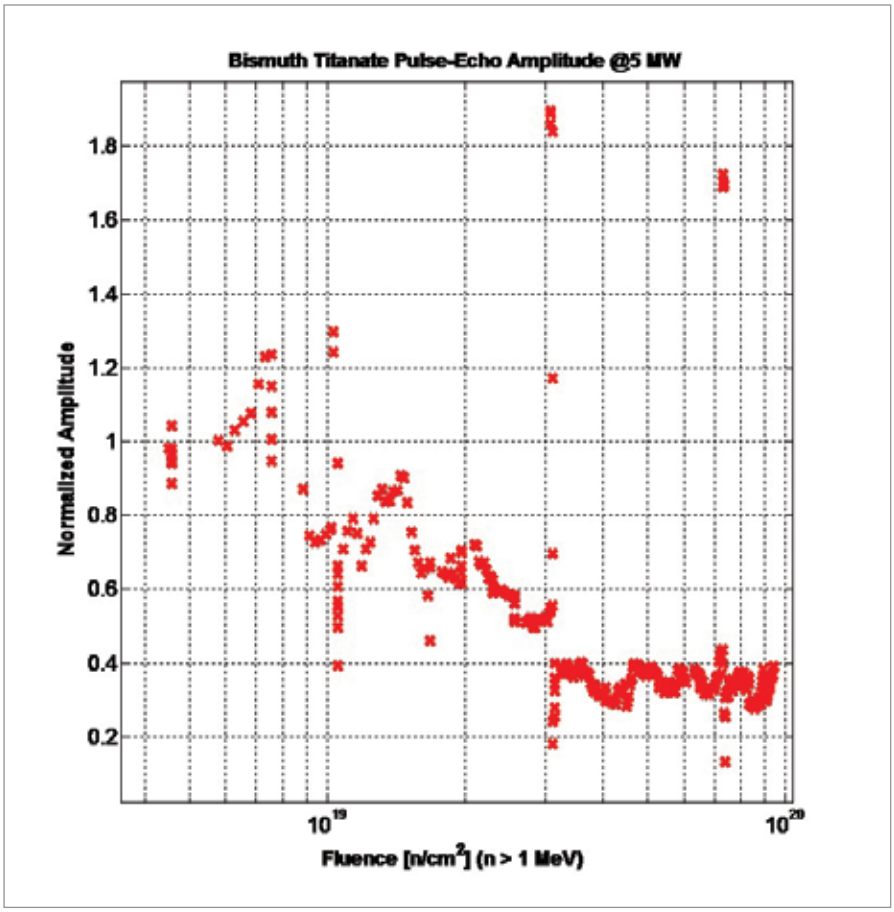


Figure 8. Pulse-echo amplitude measured for the BiT transducer during the first power cycle.

presence of high levels of radiation and high temperatures that would destroy typical commercial ultrasonic transducers.

Future Activities

During the second year of the experiment, PSU personnel shipped signal-processing equipment to MITR and then travelled to MITR to assist with the insertion and setup of the signal-processing equipment. The irradiation started in February 2014. PSU is monitoring and interpreting the data from the irradiation, including supporting laboratory evaluations.

The project's third year will witness the completion of 18 months of irradiation. PSU and INL researchers will travel to MITR to assist with removal of samples from the reactor, final measurements, and post-irradiation examination (PIE), during which the effects of irradiation on the piezoelectric candidate materials will be quantified. System components will be examined at the MIT Hot Box, and specific parameters, including ultrasonic velocity and attenuation, transduction efficiency of piezoelectric, electrical resistivity, color, crystallinity, and physical and electrical robustness, will be evaluated.

The scope of the PIE will be based on the activation level of the transducers. It is likely that the coaxial cables can be left intact as the experiment is moved to the MIT Hot Box. If this is the case, it will be possible to reconnect these cables to non-irradiated cables and interrogate the transducers after they have been placed in the Hot Box. Detailed analysis of the magnetostrictive transducer material will probably not be possible due to the cobalt-bearing materials with which they are made.

At the conclusion of the PIE activities, PSU scientists will prepare a final report summarizing the results of this project, and MIT will dispose of the irradiated materials.

Publications and Presentations*

1. J. Daw, J. Palmer, P. Ramuhalli, P. Keller, R. Montgomery, H-T. Chien, B. Tittmann, B. Reinhardt, G. Kohse, J. Rempe, 2015, "Ultrasonic Transducer Irradiation Test Results." *NPIC & HMIT at Charlotte, NC, February* 23–26, 2015.

2. B. Reinhardt, B. Tittmann, J. Rempe, J. Daw, G. Kohse, D. Carpenter, M. Ames, Y. Ostrovsky, P. Ramuhalli, R. Montgomery, H. T. Chien, and B. Wernsman, 2014, "Progress towards developing neutron tolerant magnetostrictive and piezoelectric transducers," 41st *Annual Review of Progress in Quantitative Nondestructive Evaluation Conference, Boise, ID, July* 20–25, 2014.

3. D.A. Parks and B.R. Tittmann, 2014, "Radiation tolerance of piezoelectric bulk single crystal aluminum nitride," *IEEE Transactions on Ultrasonics Ferroelectrics and Frequency Control*, Vol. 61, No 7, pp. 1216–1222.

**See additional publications from other years in the Media Library on the NSUF website.*

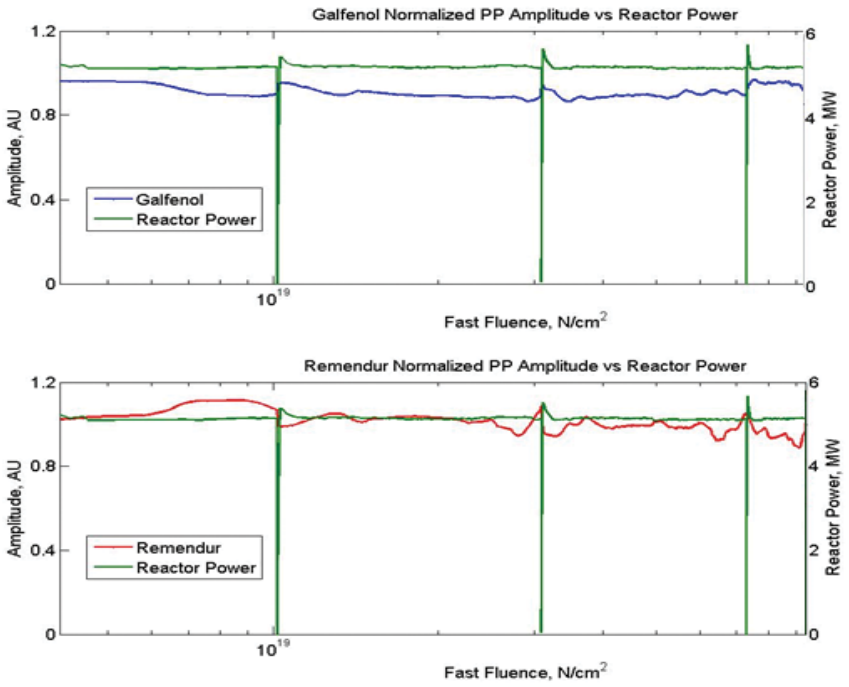


Figure 9. Pulse-echo amplitude of the Galfenol and Remendur magnetostrictive transducers.

Distributed Partnership at a Glance

ATR NSUF and Partners	Facilities and Capabilities
Massachusetts Institute of Technology	Research Reactor, PIE facilities
Idaho National Laboratory	PIE facilities
Collaborators	
Pennsylvania State University	Bernhard Tittmann (principal investigator), Brian Reinhardt (graduate student), Andy Suprock (graduate student)
Idaho National Laboratory	Joy Rempe (co-principal investigator), Joshua Daw (collaborator), Joseph Palmer (collaborator)
Massachusetts Institute of Technology	Gordon Kohse (collaborator)
Pacific Northwest National Laboratory	Pradeep Ramuhalli (collaborator)
Argonne National Laboratory	H.T. Chien (collaborator)
Bettis Atomic Power Laboratory	Ben Wernsman (collaborator)

Electron Backscatter Diffraction and Atom Probe Tomography to Study Krypton Segregation Behavior in Uranium Dioxide

Michele Manuel – University of Florida – mmanuel@mse.ufl.edu

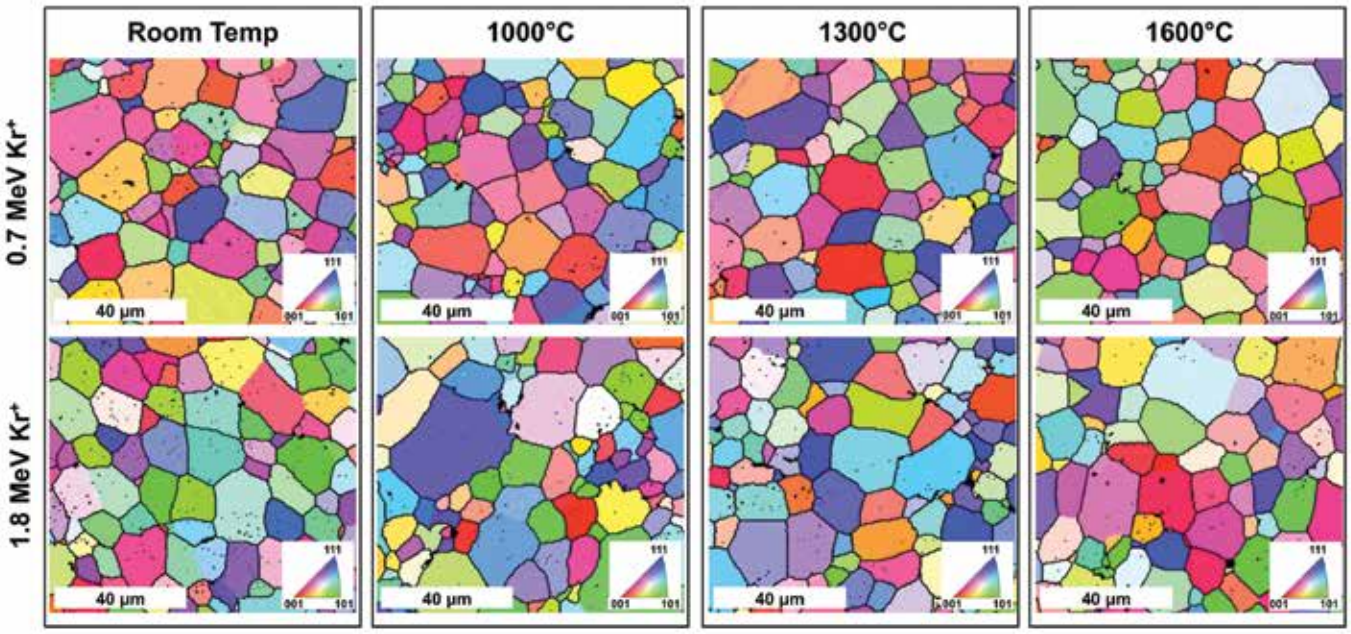


Figure 1. Representative EBSD maps of 0.7 MeV and 1.8 MeV Kr-irradiated UO_2 and the following post-irradiation anneal.

The ability to produce specific fuel microstructures by Pulsed DC Magnetron Sputtering is a powerful sample preparation technique, and the research conducted at CAES pushes this method forward to full validation.

Project Description

The objective of this research was to investigate the structure of a uranium oxide thin film produced by pulsed dc magnetron sputtering. This technique allows films of specific microstructures to be grown for analysis. Uranium dioxide is a pervasive material in nuclear energy, and the ability to produce defined microstructures for testing makes this an exciting technique, especially given the difficulty in sample preparation of nuclear fuel. This specific project was to conduct atom probe tomography on the interface between the yttria-stabilized zirconia substrate and UO_2 thin film to look for possible inter-diffusion that had occurred during processing. This research supports efforts to make the microstructural investigation and testing of nuclear fuels more insightful and cost-effective.

Accomplishments

Samples were fabricated on the focused ion beam at the Center for Advanced Energy Studies (CAES) in Idaho Falls, Idaho and then analyzed with atom probe tomorgraphy. The goal of the project, to visualize the interface between film and substrate, was successful and incorporated into a publication in the journal Applied Surface Science. This research was

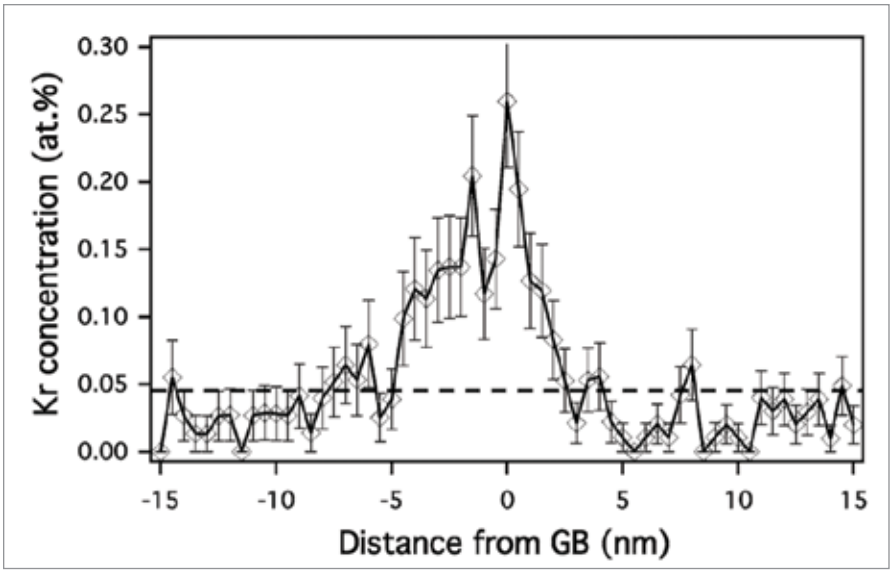


Figure 2. Kr concentration across a high angle grain boundary in UO_2 annealed at 1600°C, showing significant segregation. The horizontal dashed line represents the bulk composition of Kr in the material.

conducted primarily by Billy Valderrama and facilitated in large part by staff at CAES, including Jatuporn Burns, Dr. Yaqaio Wu and Joanna Taylor.

Research to be completed

This research is complete, as it was a single study of one material for a publication.

Publications and Presentations

1. Lin, I Dahan, B Valderrama, M. V. Manuel, 2014, "Structure and properties of uranium oxide thin films deposited by pulsed dc magnetron sputtering," *Applied Surface Science*, Vol. 301, pp. 475–480.

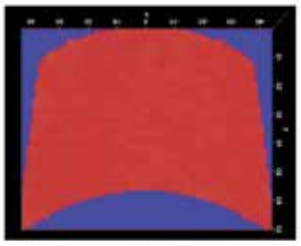
Access to the CAES facility has provided unprecedented insight into the behavior of nuclear fuels.

— Michele Manuel, *Associate Professor, Department of Materials Science and Engineering, University of Florida*

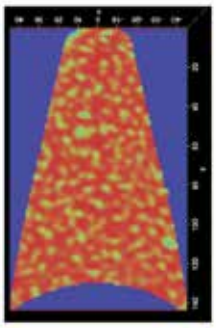
Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
Idaho National Laboratory	Jian Gan (collaborator)
University of Florida	Michele Manuel (principal investigator), Billy Valderrama (collaborator), Hunter Henderson (collaborator)

Irradiation Effects in Aged Cast Duplex Stainless Steels

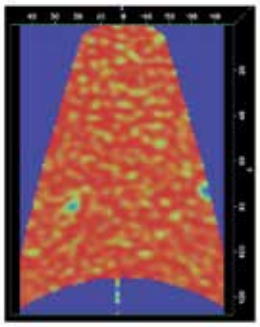
Yong Yang – University of Florida – yongyang@ufl.edu



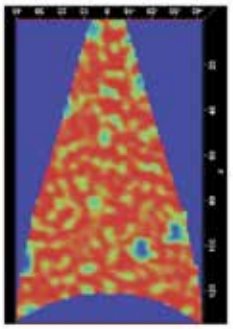
As-cast



Aged



As-cast & Irradiated



Aged & Irradiated

Figure 1. Spinodal decomposition of ferrite phase in cast stainless steels, where the red color represents Fe atoms and the blue color is the background.

By correlating microstructural studies with associated mechanical tests, this research provides a fundamental understanding of the aging behavior of cast stainless steels during the 40 years of as-designed reactor life, and, more importantly, during the 60 years and beyond of a reactor's anticipated extended life.

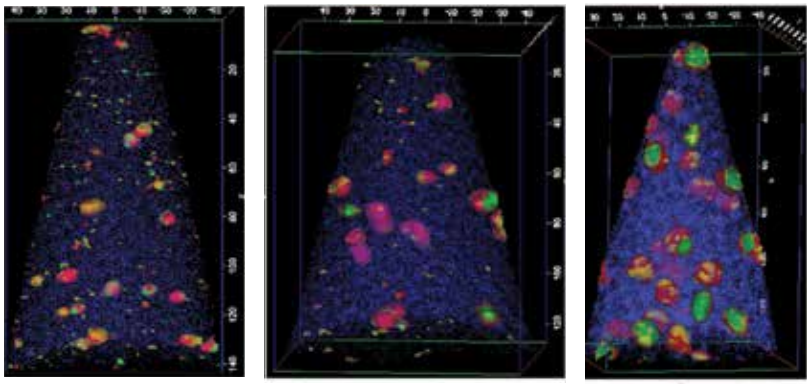
Project Description

Researchers used transmission electron microscopy (TEM) and atom probe tomography (APT) to systemically characterize the neutron-irradiated cast stainless steels under non-aged and aged conditions, focusing on precipitations and elemental segregation. The results of the study will provide some of the first knowledge of the synergistic effects of thermal aging and neutron irradiation on microstructural evolution in reactor components made of cast stainless steel.

Accomplishments

Microstructural changes in the ferrite atoms of thermally aged only, neutron-irradiated only and neutron-irradiated after thermal aging cast austenitic stainless steels (CASS) were investigated using APT. It was found that low-dose neutron irradiation could effectively induce spinodal decomposition in the ferrite atoms of non-aged CASS, while the neutron irradiation of aged CASS would further enhance the spinodal decomposition (Figure 1).

Determining the combined effects of thermal aging and low-dose neutron irradiation on duplex stainless steels (cast stainless and austenitic stainless steel welds) is going to have major implications for the sustainability of light water reactors (LWR) when their service lives are extended to 80 years.



Aged | As-cast & Irradiated | Aged & Irradiated

Figure 2. G-phase precipitates in aged, cast-irradiated, and aged-irradiated CF-3. Image sizes reflect identical scales, and the blue dots represent Cr atoms. The concentrations of the iso-surface plots of Mn, nickel (Ni), and silicon (Si) are 5, 20, and 15 at.%, respectively.

Neutron irradiation not only dramatically increases the size of G-phase precipitates in the stainless steel samples, but increases the amount of chromium (Cr), phosphorous (P), and molybdenum (Mo) and decreases the iron (Fe) and manganese (Mn) (Figure 2). This study proves that the effects of neutron irradiation on ferrite degradation are highly dependent on the irradiation dose rate at an LWR's operational temperatures.

Researchers postulate that a synergistic effect exists in duplex stainless steel components between thermal aging and neutron irradiation even at very low doses.

Future Activities

The project was completed in 2014.

Publications and Presentations

1. W-Y Lo, Y. Chen, J. Pakarinen, Y. Wu, T. Allen, and Y. Yang, "Irradiation response of delta ferrite in as-cast and thermally aged cast stainless steel," submitted to *Journal of Nuclear Materials*, 2014, under revision.

Understanding the effects of long-term thermal aging and neutron irradiation on duplex stainless steels in LWRs will provide a strong scientific basis for managing LWRs as they age beyond their designed-for lifetimes.

— Yong Yang, Assistant Professor, Nuclear Engineering, University of Florida

Distributed Partnership at a Glance

ATR NSUF and Partners	Facilities and Capabilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
University of Florida	Yong Yang (principal investigator), Wei-yang Lo (graduate student, collaborator), Zhangbo Li (graduate student, collaborator)
Argonne National Laboratory	Yiren Chen (co-principal investigator)
University of Wisconsin - Madison	Janne Pakarinen (co-principal investigator)

Scanning Transmission Electron Microscopy/Local Electrode Atom Probe Study of Fission Product Transportation in Neutron Irradiated Tri-structural Isotropic Fuel Particles

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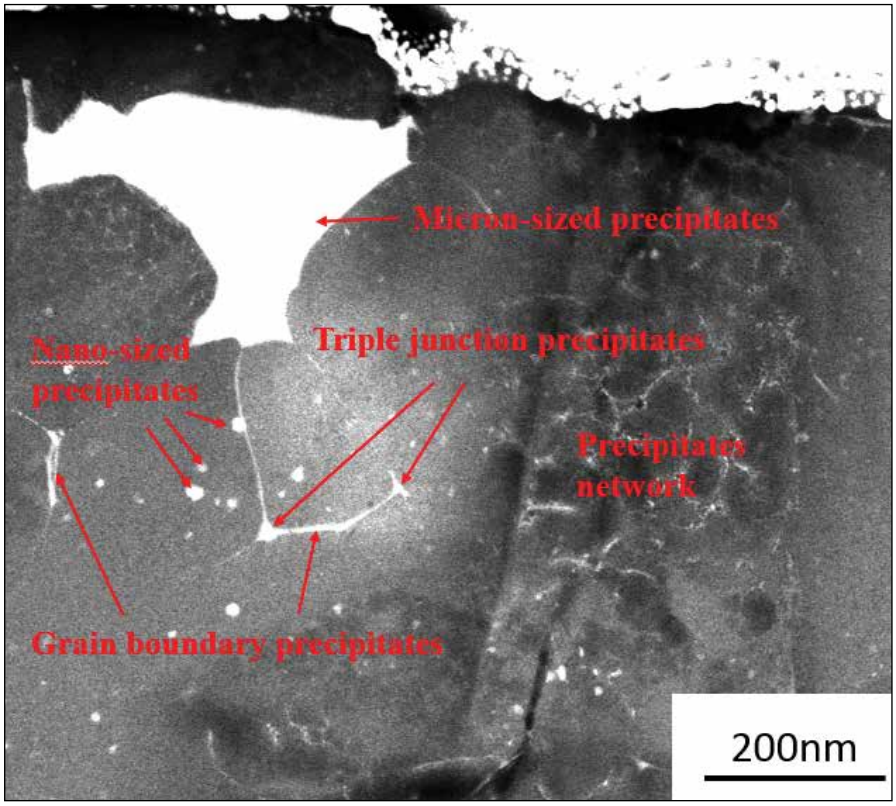

Figure 1. Distribution of FP precipitates in the SiC layer (~5 µm from SiC/IPOyC interface) of a neutron-irradiated TRISO fuel particle from AGR-1 experiments.

Tristructural-isotropic (TRISO)-coated particle fuel, consisting of a spherical fuel kernel encapsulated in successive layers of pyrolytic graphite (PyC) and silicon-carbide (SiC) is being considered as fuel for both the Generation IV Very High Temperature Reactor (VHTR) and the Fluoride Salt-Cooled High-Temperature Reactor (FHR). The SiC layer is intended to be the primary barrier to the release of radioactive metallic fission products (FP). However, the release of FP, particularly silver (Ag) from seemingly intact TRISO particles has been observed. Therefore, understanding the mechanisms of FP Ag transport through SiC is necessary to limit its release and consequently promote the safe operation of the reactor.

Because research has been limited by the inability of nanoscale analysis equipment to handle irradiated materials, little is known about how FPs are transported through the SiC layer. In

fact, Ag was first identified in the SiC layer of a neutron-irradiated TRISO fuel particle only in 2013 by a Boise State University research team at INL using scanning transmission electron microscopy (STEM) [1].

Project Description

As a continuation of the 2013 preliminary STEM study, this project utilized STEM and the local electrode atom probe (LEAP) to further investigate the morphology, composition and distribution of FPs in the SiC layer of the same neutron-irradiated TRISO fuel particles. The goal was to gain further insight into the FP's transport mechanism, particularly that of Ag. The various fission products found have improved our understanding of FP release mechanisms in this type of fuel.

Accomplishments

STEM-EDS Analysis: Two transmission electron microscopy (TEM) lamellae lifted from the SiC layer close to the SiC/innerPyC interface were examined in detail using STEM and energy dispersive spectroscopy (EDS). The excellent Z contrast observed under STEM clearly revealed the existence of various types of FP precipitates (Figure 1), and EDS aided in the determination of the composition of these precipitates. This study confirmed previous reports of Ag-enriched grain boundary and triple-junction precipitates, and identified for the first time the Ag-palladium (Pd) intragranular precipitates as well as a network of small uranium-rich precipitates. These findings advance our understanding of FP transport through the SiC layer in TRISO fuel particles.

LEAP Analysis: A batch of 15 LEAP tips from the regions where the TEM lamellae were fabricated and examined in 2013 was prepared. The first phase of this work resulted in some fracturing of the tips, primarily due to the weakness of the TEM sample grid on which the samples were placed. The largest data set obtained was only from depths of about ~30 μm.

In the second phase of this study, another batch of five tips was fabricated and tested in 2014. These tips were made from material taken from the same location as in the first phase, but this time they were mounted in a much sturdier copper focused ion beam (FIB) grid. With the optimized equipment parameters (laser energy of 80 to 100 petajoules (pJ), laser pulse of 160 to 200 kilohertz (kHz), temperature of 30–60 K), better grid data could be obtained from deeper in the material than was possible in the first phase of the project. The largest data set obtained is shown in Figure 2.

Although the results were superior to those obtained from Phase one of this study, the analysis depth was

"The cutting-edge analytical equipment in the Microscopy and Characterization Suite at CAES can shed new light on thermodynamically and kinetically driven nanoscale physical and compositional changes in materials."

— Dr. Bin Leng, Research Associate, University of Wisconsin - Madison

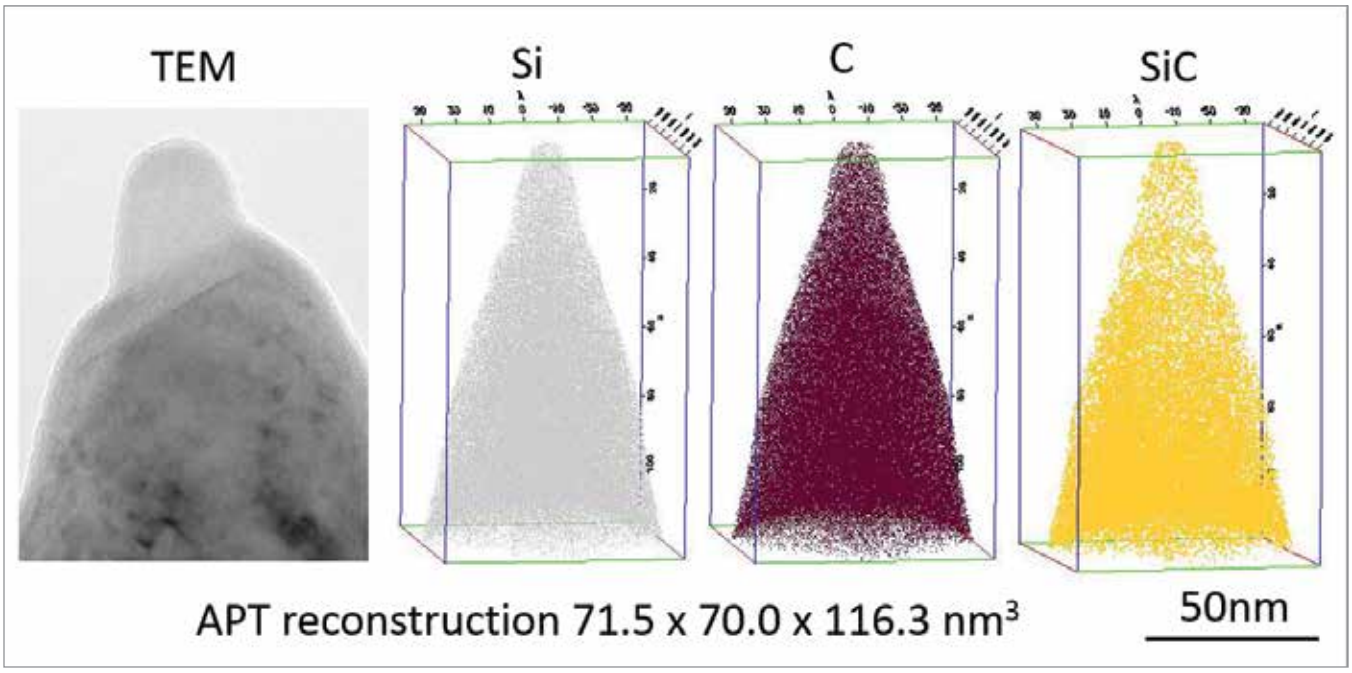

Figure 2. LEAP results showing the distribution of elements in a tip fabricated from a neutron-irradiated TRISO fuel particle.

Various fission products found in the SiC layer of a neutron-irradiated TRISO fuel particle have improved our understanding of the fission product release mechanisms in this type of fuel.

still limited and no FP were detected. Therefore, in the third phase of this study, ten tips were fabricated at the University of Wisconsin – Madison from a surrogate Ag ion-implanted SiC sample and shipped to CAES for LEAP examinations. Compared with the neutron-irradiated samples, the surrogate tips were far less prone to fracture, and data was obtained from depths of up to ~240 nm. However, no Ag was detected (Figure 3).

It was concluded that LEAP examination of SiC is limited by the relatively low electrical and thermal conductivity of this material, as well as its brittleness, which is exacerbated by radiation-induced defects. More extensive research is needed to exploit the full benefits of the LEAP technique for the examination of irradiated SiC samples.

Future Activities

This project has been completed.

References

[1.] I. J. van Rooyen, Y. Q. Wu, T. M. Lillo, 2014, "Identification of silver and palladium in irradiated TRISO coated particles of the AGR-1 experiment," *Journal of Nuclear Materials*, 446, pp. 178–186.

Publications and Presentations

1. I.van Rooyen, B. Leng, Y. Wu, T. Lillo, I. Szlufarska, K. Sridharan, T. Gerczak, J. Madden, 2014, "Identification of Fission Products in irradiated SiC Using Scanning Transmission Electron Microscopy and Atom Probe Tomography," *ATR NSUF User's Week*.

2. B. Leng, I. van Rooyen, Y. Wu, I. Szlufarska, K. Sridharan, 2015, "STEM-EDS analysis of fission products in neutron-irradiated TRISO fuel particles from AGR-1 experiment," submitted to the *Journal of Nuclear Materials*.

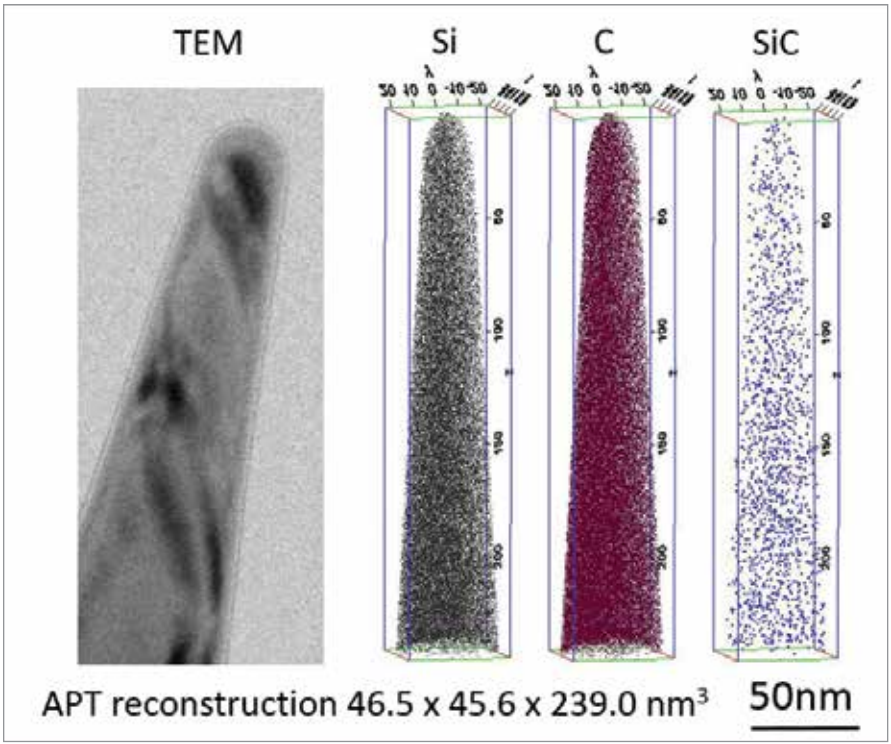

Figure 3. LEAP results showing the distribution of elements in a tip fabricated from surrogate Ag ion-implanted SiC.

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	Advanced Test Reactor
Center for Advanced Energy Studies	Microscopy and Characterization Suite
University of Wisconsin – Madison	PIE facilities
Collaborators	
University of Wisconsin – Madison	Izabela Szlufarska (principal investigator), Bin Leng (collaborator), Tyler Gerezak (collaborator), Kumar Sridharan (collaborator)
Idaho National Laboratory	Isabella van Rooyen (principal investigator), James Madden (collaborator), Tim Lillo (collaborator)
Center for Advanced Energy Studies	Yaqiao Wu (collaborator)

Irradiation-Assisted Diffusion of Uranium-Iron Diffusion Couples

Lin Shao – Texas A&M University – lshao@tamu.edu

Since the chemical compatibility between the fuel and the cladding is imperative for safe operation of a reactor, it is essential to limit the interdiffusion between the two.

Irradiation of nuclear fuel results in fuel swelling and the production and transport of fission products to the cladding. This contact may allow chemical and mechanical interactions with the cladding. Since the chemical compatibility between the fuel and the cladding is imperative for safe operation of a reactor, it is essential to limit the interdiffusion between the two.

Project Description

This project is investigating fuel-cladding interactions with prototypic material systems that have traditionally been difficult to study these interactions under extreme conditions.

Accomplishments

The objective of this research is to provide an understanding of thermally activated and irradiation-enhanced multicomponent-multiphase diffusion, and microstructural evolution in transition and rare earth metals, relevant to fuel-cladding interactions. Studies to date have validated equilibrium phases predicted by phase, diagrams of uranium-iron (U-Fe), uranium-nickel (U-Ni), and uranium-zirconium (U-Zr) binary systems. In addition, researchers observed unusual microstructures in several phases which were not reported before. Discrepancies with previous publications were documented for crystalline structures and lattice parameters of certain phases. Integrated diffusion coefficients at different temperatures and their activation energies were extracted for each diffusion couple.

In the U-Fe system, researchers found that integrated diffusion coefficients for diffusion couples prepared using polycrystalline Fe and single-crystalline Fe are largely different. Figure 1 shows that diffusion coefficients for the polycrystalline Fe sample are systematically larger and the diffusion activation energy is lower than that of the single crystalline Fe sample. This is because atoms can use grain boundaries as quicker diffusion paths to broaden the widths of interfacial phases. This finding also points to the necessity of using single crystal diffusion couples for better comparisons between modeling predictions and actual results. Figure 2a-d plot the U and Fe elemental distributions in U-Fe (single crystal) diffusion couples obtained at different annealing temperatures. Figure 2e-f shows typical back-scattered-electron (BSE) images of diffusion annealed at 625°C and 650°C, respectively.

Future Activities

All the focused ion beam (FIB) time allocated for this project has been used. Researchers plan to write another proposal for continuing support.

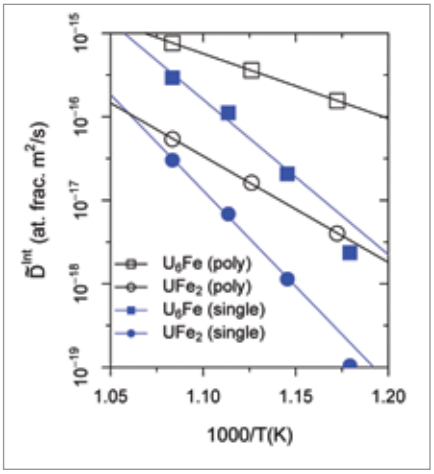

Figure 1. Comparisons of integrated diffusivities forming UFe_2 and U_6Fe phases in U-Fe diffusion couples using polycrystalline Fe and single crystal Fe, respectively.

Publications and Presentations*

1. T. Chen, T. Smith, J. Gigax, D. Chen, R. Balerio, B. H. Sencer, J. R. Kennedy, L. Shao, "Diffusion kinetics and grain boundary effects in interface reactions of U–Fe diffusion couples," *Journal of Nuclear Materials, in press.*

2. L. Shao, D. Chen, C-C. Wei, M. Martin, X. Wang, Y. Park, E. Dein, K. R. Coffey, Y. Sohn, B. H. Sencer, and J. R. Kennedy, 2015, "Radiation effects on interface reactions of U/Fe, U/(Fe+Cr), and U/(Fe+Cr+Ni)," *Journal of Nuclear Materials*, vol. 456, pp. 302.

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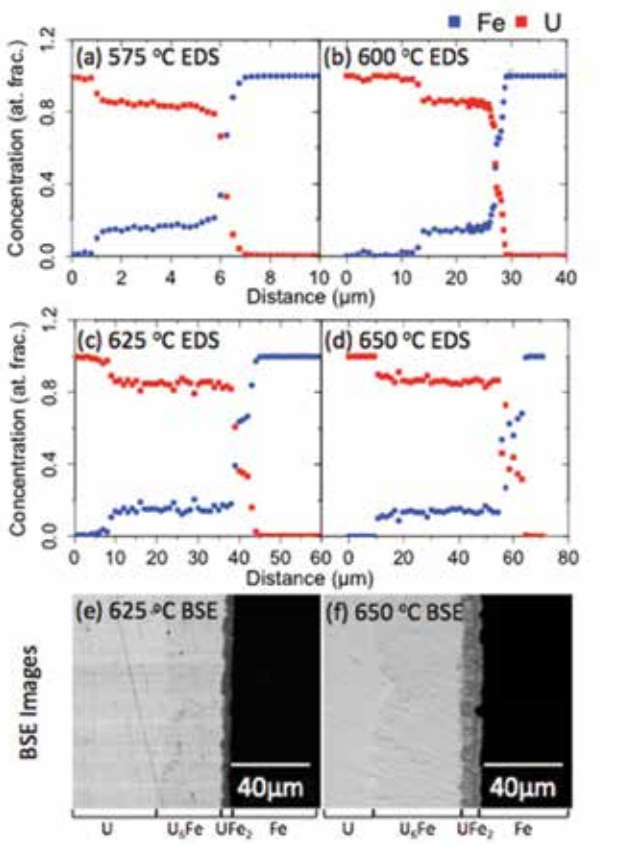

Figure 2. Concentration profiles obtained for diffusion couples U-Fe (single crystal) annealed at different temperatures (a-d) and typical BSE image at 625°C and 650°C diffusion couples showing formation of intermetallic crystals (e-f).

> *"The Center for Advanced Energy Studies is second to none, and we are lucky to have access to it."*
>
> *— Lin Shao, Associate Professor, Texas A&M University*

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	Advanced Test Reactor
Center for Advanced Energy Studies	Microscopy and Characterization Suite
University of Wisconsin – Madison	PIE facilities
Collaborators	
University of Wisconsin – Madison	Mahima Gupta (principal investigator), Janne Parkarinen (collaborator)
Idaho National Laboratory	Todd Allen (co-principal investigator), Jian Gan (collaborator)
Los Alamos National Laboratory	Steve Conradson (collaborator)

Toward an Understanding of the Effect of Dose Rate on the Irradiation Response of F-M Alloys

Janelle P. Wharry – Boise State University – janellewharry@boisestate.edu

With their high strength, resistance to thermal stresses, dimensional stability, and low activation, ferritic-martensitic (F-M) alloys are leading candidates for cladding and structural components in fusion and advanced fission reactors.

Throughout their service lifetimes, F-M alloys are exposed to neutron irradiation at doses as high as several hundred displacements per atom (dpa). To simulate the neutron damage produced at these high doses, ions are often used in test reactors to irradiate F-M samples. The ions' higher damage rates and minimal residual radioactivity allow for quicker, more cost-effective experiments; however increasing the irradiation dose rate also increases the rate of point defect recombination. This means researchers may not be able to directly compare the microstructure development between the proton- and neutron-irradiated specimens.

Even so, understanding the differences produced by the irradiating particle types and dose rates, as well as their implications, is key to using ion-irradiation techniques to assess the long-term viability of F-M alloys as materials in advanced reactor systems.

Limited studies have been carried out on neutron-irradiated F-M materials, so the effects of particle types and dose rates in F-M alloys are yet to be resolved.

	HCM12A			HT9		
	As Received	Proton, 3 dpa, 500°C	Neutron, 3 dpa, 500°C	As Received	Proton, 3 dpa, 500°C	Neutron, 3 dpa, 500°C
Grain diameter (10^{-6} m)	0.66	0.66	0.66 ± 0.36	0.30	0.30	0.30 ± 0.10
Dislocation line density (10^{14} m^{-2})	12.84	12.1 ± 4.21	13.6 ± 3.55	13.9	14.1 ± 4.0	13.8 ± 4.3
Carbide precipitate diameter (10^{-6} m)	0.05	0.05	0.05 ± 0.03	0.06	0.06	0.06 ± 0.03
Carbide number density (10^{20} m^{-3})	0.92	0.92	0.92 ± 0.12	0.71	0.71	0.71 ± 0.41
Void diameter (10^{-9} m)	–	–	–	–	–	4.20 ± 1.03
Void number density (10^{21} m^{-3})	–	–	–	–	–	0.19
Dislocation loop diameter (10^{-9} m)	–	10.8 ± 4.20	12.5 ± 4.11	–	11.9 ± 6.12	10.0 ± 3.62
Dislocation loop density (10^{21} m^{-3})	–	0.9	0.9	–	2.2	0.9
Si-Mn-Ni-P cluster diameter (10^{-9} m)	–	5.51 ± 1.91	1.56 ± 0.65	–	4.31 ± 2.09	1.71 ± 0.96
Si-Mn-Ni-P cluster no. density (10^{21} m^{-3})	–	23	788	–	19	582
Cu-rich cluster diameter (10^{-9} m)	–	3.96 ± 1.42	1.97 ± 0.47	–	–	–
Cu-rich cluster number density (10^{21} m^{-3})	–	23	447	–	–	–
Cr-rich cluster diameter (10^{-9} m)	–	–	0.54 ± 0.13	–	–	0.47 ± 0.10
Cr-rich cluster number density (10^{21} m^{-3})	–	–	1312	–	–	1775

Table 1. Microstructure measurements of HCM12A and HT9 samples, as-received and proton- or neutron-irradiated to 500°C and 3 dpa.

Project Description

The goal of this project is to better understand the effects of varying dose rates on F-M alloys by comparing the damage inflicted on two identically heated commercial alloys, HCM12A and HT9, by neutron and proton irradiation. Both alloys were irradiated in the ATR with neutrons at a dose rate of ~10^{-7} dpa/sec and with 2.0 MeV protons at ~10^{-5} dpa/sec. Both irradiations were carried out to 3 dpa at a temperature of 500°C.

Because it aims to (1) understand the response of advanced reactor candidate structural materials to irradiation, and (2) assess the ability of proton irradiations to emulate in-reactor neutron irradiation damage, this project has direct relevance to the Advanced Reactor Technologies program being conducted by the Department of Energy's Office of Nuclear Energy (DOE-NE).

The primary DOE-NE mission is to advance nuclear power as a viable resource for meeting the nation's energy, environmental, and national security needs. Generation IV advanced reactor designs, such as high-temperature reactors and fast-neutron spectrum reactors, fulfill this mission by combining high-efficiency power generation with the environmental and national security benefits of consuming the extended-life radioactive isotopes found in spent nuclear fuel.

However, along with the promise of Generation IV designs comes the challenge of finding suitable structural

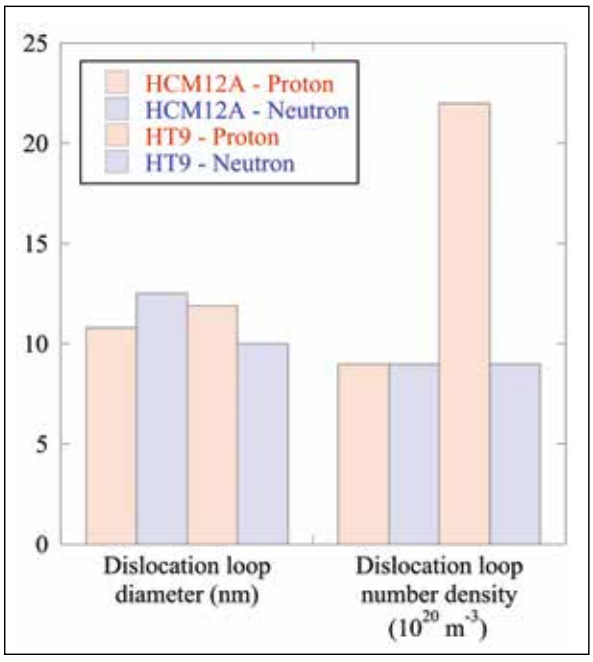

Figure 1. Irradiation-induced dislocation loop diameter and number density produced by proton and neutron irradiations to 3 dpa at 500°C in HCM12A and HT9.

"This project enabled two graduate students to develop proficiency in microscopy techniques, and has laid the groundwork for their thesis projects."

— Janelle Wharry, Assistant Professor, Materials Science & Engineering, Boise State University

materials that will withstand the harsh operating conditions in these new reactors. Ensuring the integrity of these materials under high temperatures, corrosive environments, cyclic loading and high irradiation damage is paramount to the safety, performance and long-term success of the Generation IV nuclear fleet.

Accomplishments

This project compared the microstructure evolution of the commercial F-M alloys HCM12A and HT9 exposed to proton and neutron irradiations at 500°C to 3 dpa. Results of this project suggest that proton irradiation produces a dislocation loop morphology comparable to that of neutron irradiation, but that irradiation-induced nanoscale clustering behavior varies considerably between the two particle types.

Prior to this project, neutron irradiations of HCM12A and HT9 had been completed in ATR at a dose rate of approximately 10^{-7} dpa/sec. Specimens from the same heats were also irradiated with 2.0 MeV protons at the Michigan Ion Beam Laboratory at a dose rate of approximately 10^{-5} dpa/sec. Researchers performed a full microstructure characterization on both the neutron- and proton-irradiated materials. Since the response of F-M alloys to irradiation has been found to be sensitive to variations in alloy heats, examining these materials offered a tremendous opportunity to compare the effects of both proton and neutron irradiations on identical alloy heats.

A combination of transmission electron microscopy (TEM) and local electrode atom probe (LEAP) analysis enabled thorough characterizations of the irradiated microstructures and phase evolutions. All material characterization work was conducted in the Microscopy and Characterization Suite (MaCS) at CAES, utilizing the FEI Quanta focused ion beam (FIB), CAMECA 4000X HR LEAP, and FEI Tecnai STwin TEM.

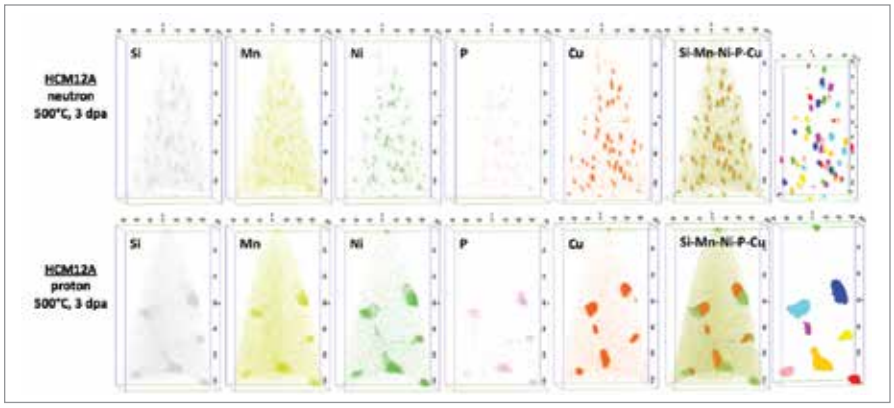

Figure 2. LEAP tip reconstructions of neutron- (top) and proton-irradiated (bottom) HCM12A to 3 dpa at 500°C, showing (left to right) Si, Mn, Ni, P, Cu; Si-Mn-Ni-P-Cu; and cluster identification.

Generation IV reactors combine high-efficiency power generation with the environmental and national security benefits of consuming the long-lived radioactive isotopes found in spent nuclear fuel.

TEM results showed that grain size, dislocation line density and carbide precipitates had not been changed by either proton or neutron irradiation (Table 1). Irradiation-induced dislocation loops were also observed by TEM in both HCM12A and HT9. In both alloys, dislocation loop morphologies and number densities were consistent across both irradiation types, although proton irradiation generated a slightly higher number density of loops in HT9 than did neutron irradiation (Figure 1).

Voids were found only in the neutron-irradiated HT9. These voids were small, with diameters <5 nm, and very sparsely populated.

At TEM resolution, the neutron-irradiated microstructures in HCM12A and HT9 can be replicated using proton irradiation at the same dose and temperature. However, below TEM resolution, proton irradiation produces significantly different nanoscale features than neutron irradiation.

LEAP analysis observed nucleation of three types of nanoscale clusters: (1) those rich in silicon (Si), manganese (Mn), nickel (Ni), and phosphorous (P); (2) those rich in copper (Cu); and (3) those rich in chromium (Cr).

Si-Mn-Ni-P clusters are found in both HCM12A and HT9 samples following proton and neutron irradiations. However, these clusters are three to four times finer and occur at more than an order of magnitude greater number density in the neutron-irradiated specimens than in the proton-irradiated specimens

(Figures 2 and 3). These clusters are comprised of approximately 3.5–7 at.% Si, 3–5 at.% Mn, 2.5–7 at.% Ni, and 0.25–1.0 at.% P. Clusters in the proton-irradiated specimens contain a higher at.% Si, Mn, Ni, and P than do those in the neutron-irradiated specimens.

Cu-rich clusters are found in both the proton- and neutron-irradiated HCM12A. Much like the Si-Mn-Ni-P clusters, they are finer and more populous in the neutron-irradiated specimen than in the proton-irradiated specimen. They are often, but not always, found coincident with the Si-Mn-Ni-P clusters (Figure 2). Cu-rich clusters are not found in HT9 (Figure 3) due to the low bulk concentration of Cu in HT9.

Cr-rich clusters are found in both HCM12A and HT9 specimens, but only in the alloys' neutron-irradiated samples. These clusters are found at a very high number density, on the order of 10^{24} m^{-3}. Their sizes are on the order of 0.5 nm. As such, these clusters can only contain a few atoms each. LEAP tip reconstructions from irradiated HT9 (Figure 3) show the differences in Cr clustering between neutron and proton irradiations. The mechanism for this Cr clustering is not yet understood.

Work on this project was completed by Corey Dolph and Matthew Swenson, graduate students at Boise State University. The project team wishes to acknowledge the assistance of Yaqiao Wu, Jatuporn Burns and Joanna Taylor, all of whom were instrumental in assisting with scheduling, instrument training and LEAP analysis.

Future Activities

This project was completed in 2014. Future work will include additional irradiation experiments and computational studies to further understand the mechanisms of nanoscale clustering in these alloys and their implications on mechanical behavior, irradiation dose rate, and particle type effects, and to compare Cr clustering with radiation-induced segregation of Cr.

Publications and Presentations

1. M.J. Swenson, J.P. Wharry, 2015, "The strengthening mechanism transition in nanofeatured ferritic-martensitic alloys." *The Minerals, Metals & Materials Society Annual Meeting, Orlando, FL. March 2015.*
2. M.J. Swenson, J.P. Wharry. "The comparison of microstructure and nanocluster evolution in proton and neutron irradiated Fe-9%Cr oxide dispersion strengthened (ODS) steel to 3 dpa at 500°C," Submitted to *Journal of Nuclear Materials.*

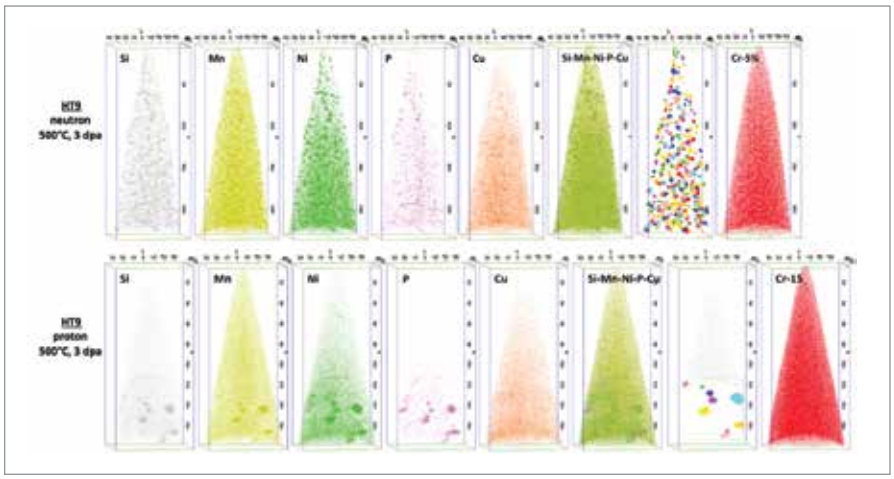

Figure 3. LEAP tip reconstructions of (top) neutron- and (bottom) proton-irradiated HT9 to 3 dpa at 500°C, showing (left to right) Si, Mn, Ni, P, Cu; Si-Mn-Ni-P-Cu; cluster identification; and Cr.

Distributed Partnership at a Glance

ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	Advanced Test Reactor, PIE facilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
Boise State University	Janelle Wharry (principal investigator), Jatuporn Burns (collaborator), Yaqiao Wu (collaborator), Corey Dolph (M.S. graduate student), Matthew Swenson (Ph.D. graduate student)
University of Idaho	Joanna Taylor (collaborator)

Electron Backscatter Diffraction and Atom Probe Tomography to Study Grain Boundary Chemistry Variation in Off Stoichiometric Uranium Dioxide Thin Films

Michele Manuel – University of Florida – mmanuel@mse.ufl.edu

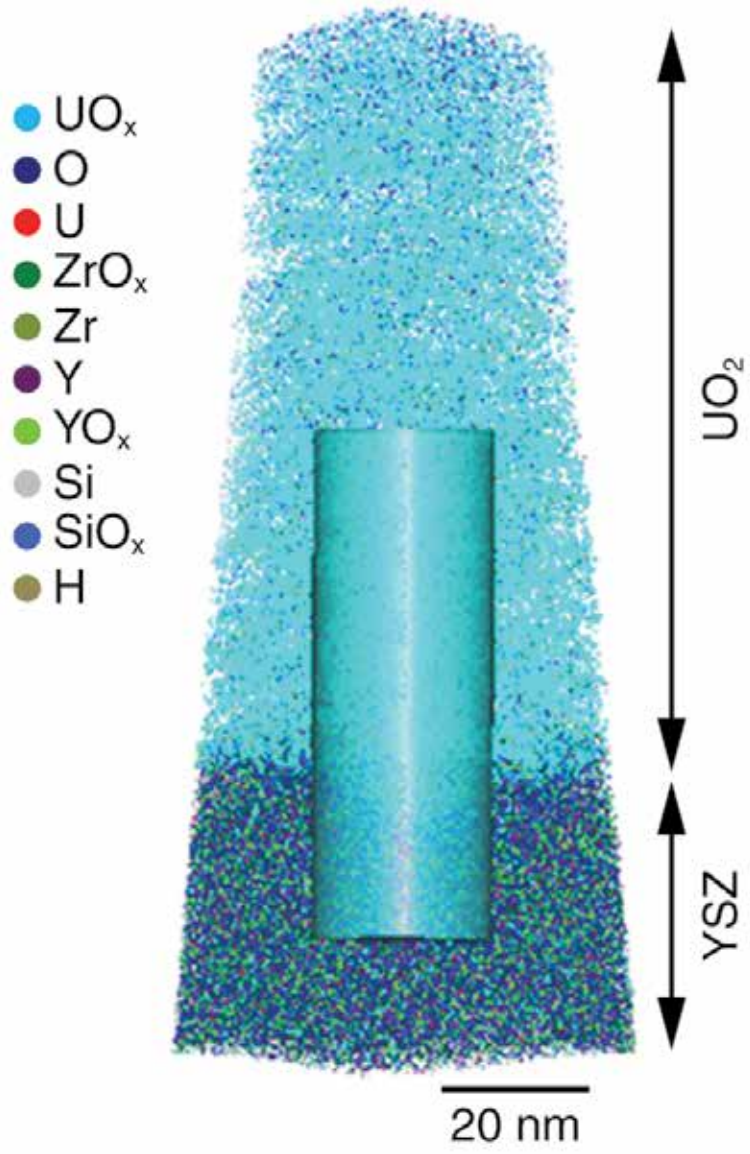

Figure 1. 3D APT reconstruction of the elements in the UO2/YSZ thin film. Complex ions represented as UO_x, ZrO_x, YO_x, where x = 1, 2, or 3.

Project Description

The objective of this research was to investigate the structure of a uranium oxide thin film produced by pulsed dc magnetron sputtering. This technique allows films of specific microstructures to be grown for analysis. Uranium dioxide is a pervasive material in nuclear energy, and the ability to produce defined microstructures for testing makes this an exciting technique, especially given the difficulty in sample preparation of nuclear fuel. This specific project was to conduct atom probe tomography on the interface between the yttria-stabilized zirconia substrate and UO_2 thin film to look for possible inter-diffusion that had occurred during processing. This research supports efforts to make the microstructural investigation and testing of nuclear fuels more insightful and cost-effective.

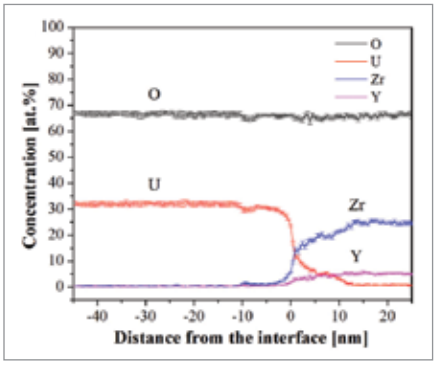

Figure 2. Atom probe 1-D concentration profile taken from the cylinder.

The ability to produce specific fuel microstructures by Pulsed DC Magnetron Sputtering is a powerful sample preparation technique, and the research conducted at CAES pushes this method forward to full validation.

Accomplishments

Samples were fabricated on the focused ion beam at the Center for Advanced Energy Studies (CAES) in Idaho Falls, Idaho and then analyzed with atom probe tomorgraphy. The goal of the project, to visualize the interface between film and substrate, was successful and incorporated into a publication in the journal Applied Surface Science. This research was conducted primarily by Billy Valderrama and facilitated in large part by staff at CAES, including Jatuporn Burns, Dr. Yaqaio Wu and Joanna Taylor.

Research to be completed

This research is complete, as it was a single study of one material for a publication.

Publications

1. J. Lin, I. Dahan, B. Valderrama, M. V. Manuel, 2014, "Structure and Properties of Uranium Oxide Thin Films Deposited by Pulsed DC Magnetron Sputtering," *Applied Surface Science*, Vol. 301, pp. 475–480.

"Access to the CAES facility has provided unprecedented insight into the behavior of nuclear fuels."

— Michele Manuel, Associate Professor, Department of Materials Science and Engineering, University of Florida

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
Idaho National Laboratory	Jian Gan (collaborator)
University of Florida	Michele Manuel (principal investigator), Billy Valderrama (collaborator), Hunter Henderson (collaborator)

Study of the Microstructures of Krypton and Xenon In Irradiated Uranium Dioxide by Advanced Microscopy Techniques

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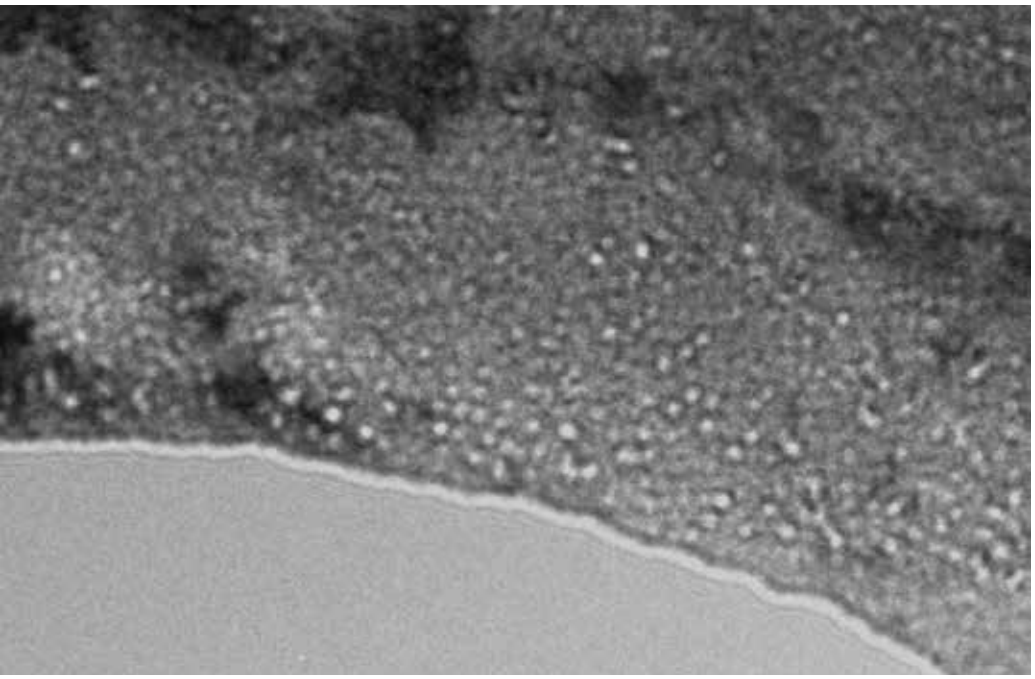


Figure 1. TEM images of bubbles in a UO_2 crystal implanted with Xe at a dose of 5 ×10^{14} ions/cm^2.

Initiated in 2012, this project is a collaborative effort of INL, Argonne National Laboratory, and the University of Wisconsin. The purpose of this rapid turnaround experiment is to clarify the microstructure and stoichiometry of uranium dioxide (UO_2) under irradiations of krypton (Kr) and xenon (Xe) ions.

Project Description

Kr and Xe are the main fission gases in UO_2 nuclear fuel. The radiation defects (i.e., dislocation loops) and precipitates (i.e., gas bubbles) induced during fission affect the fuel's structure and thermal transport properties. It is therefore important to understand and model the behaviors of Kr and Xe in order to develop optimized fuel microstructures that allow improved performance of the UO_2 fuel.

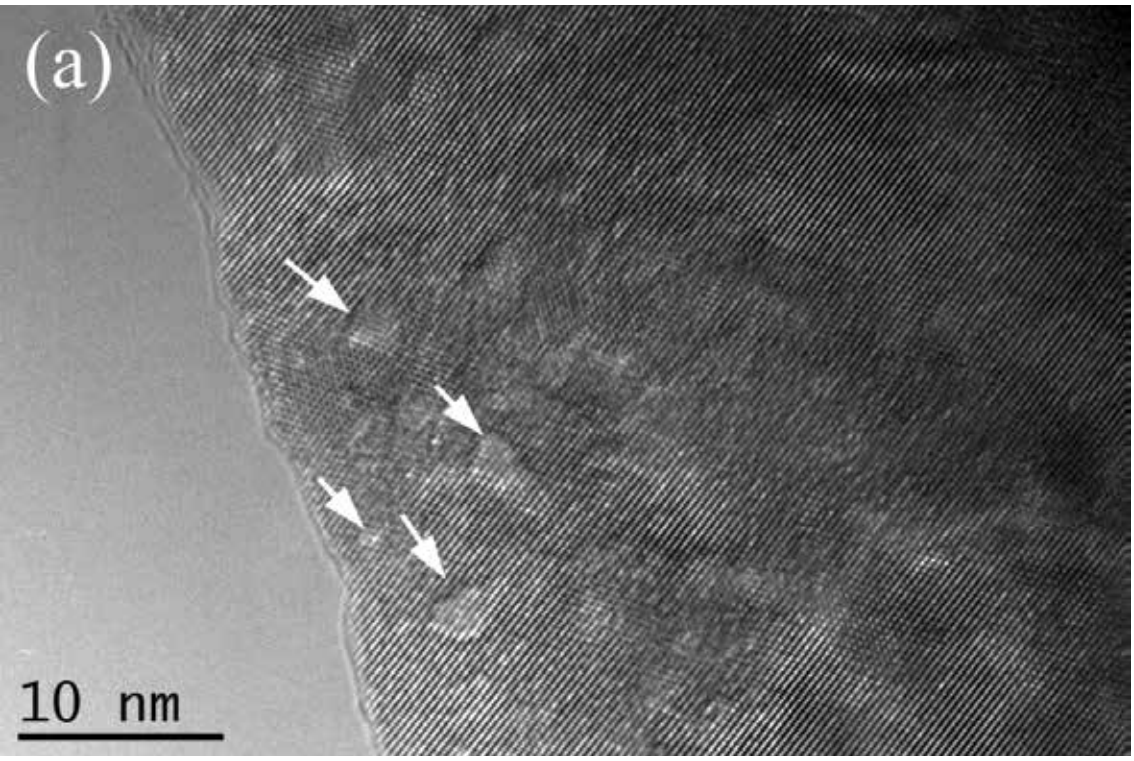
Accomplishments

To simulate the fragment damage to UO_2 caused by fission, depleted UO_2 samples were irradiated with Kr and Xe ions. To date, the research team has used a focused ion beam (FIB) to prepare cross-section lamina of the irradiated samples and transmission electron microscopy (TEM) to study the extended defects, including bubbles, dislocation loops, and dislocation lines as well as stoichiometry change.

The results indicate bubble formation under irradiation at room-temperature (Figure 1) with no solid precipitates forming in the bubbles (Figure 2). This indicates that uranium (U) vacancies may be mobile at temperatures below room temperature. It could also imply that Kr/Xe bubbles may directly nucleate at the vacancy clusters that are produced in cascades at room temperature. Such a process might not require that Kr/Xe and U vacancies be diffusive.

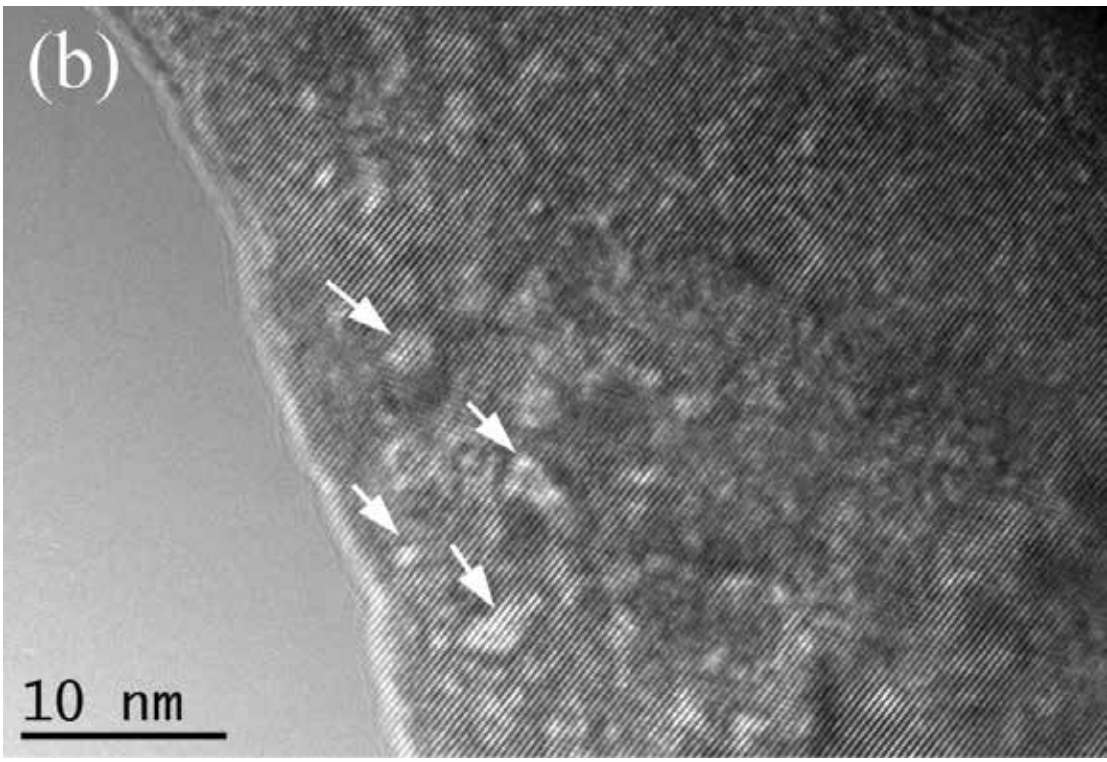
In addition, electron energy loss spectroscopy (EELS) shows that, under a vacuum, the stoichiometry of UO_2 is relatively stable during irradiation at room temperatures. At the same time, the branching ratio of the M-edge in EELS is 0.695, which is within the range of 0.695–0.720 for U^{4+}. This implies that the stoichiometry in UO_2 is unaffected by Xe irradiation (Figure 3).

We also found that the formation of dislocation-denuded zones is temperature sensitive. At 800°C, enough U interstitials migrate toward the grain boundaries so that the concentra



This experiment will improve our understanding of microstructure evolution in uranium dioxide under irradiation.

Figure 2. a) High-resolution TEM images of bubbles in a UO_2 single crystal irradiated with 300-keV Xe at room temperature at a dose of 5 ×10^{14} ions/cm^2. (b) An under-focus image of (a). Some bubbles are marked with arrows.



"Our understanding of nuclear fuels has improved greatly thanks to the state-of-the-art techniques used in this project."

— Lingfeng He, Assistant Scientist, University of Wisconsin-Madison (Currently Nuclear Fuels Engineer at Idaho National Laboratory)

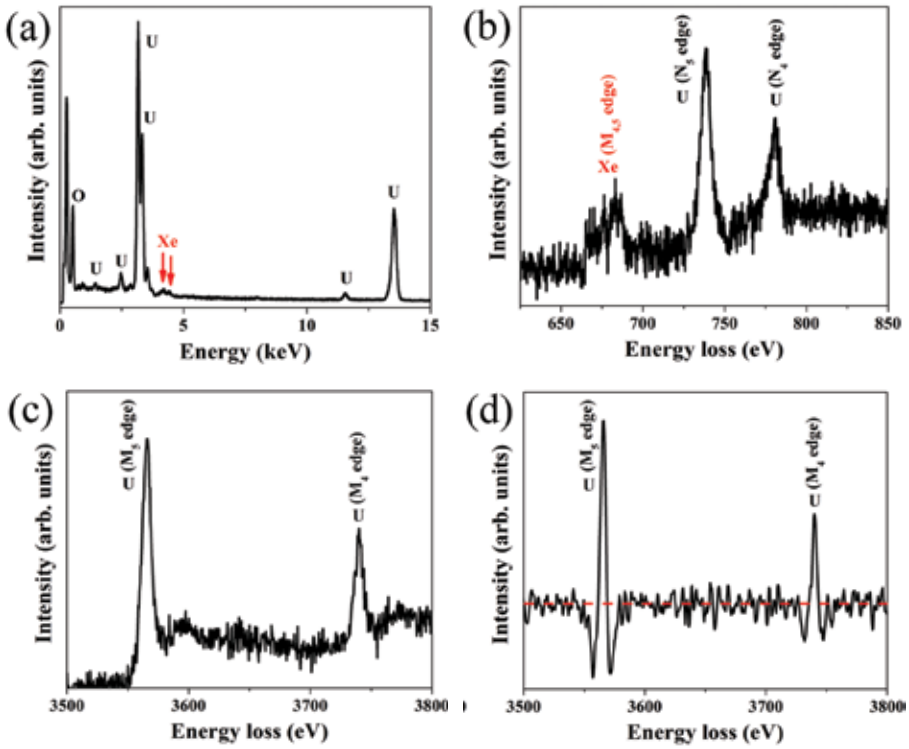


Figure 3. (a) Energy dispersive x-ray (EDX) and (b) and (c) EELS spectra of a UO_2 single crystal irradiated with 300 keV Xe at room temperature up to a dose of 1 $\times 10^{16}$ ions/cm^2. (d) The second derivative EELS spectrum of part (c).

tion of interstitials near the grain boundaries becomes too low to form loops, resulting in the formation of dislocation-denuded zones near the grain boundaries. However, at 600°C, the dislocation-denuded zones were not found, even at high-angle grain boundaries (Figure 4).

Future Activities

Circular features having diameters of 100–200 nm were found in UO_2 irradiated with Xe at high doses. However, the nature of these features is still not clear, and further TEM study is planned in 2015.

Publications and Presentations

1. L.F. He, M. Gupta, M.A. Kirk, J. Pakarinen, J. Gan, T.R. Allen, 2014, "In Situ TEM Observation of Dislocation Evolution in Polycrystalline UO2," *JOM*, Vol. 66, pp. 2553–2561.
2. L.F. He, J. Pakarinen, M.A. Kirk, J. Gan, A.T. Nelson, X.-M. Bai, A. El-Azab, T.R. Allen, 2014, "Microstructure evolution in Xe-irradiated UO_2 at room temperature," *Nuclear Instrument and Methods in Physics Research B*, Vol. 330, pp. 55–60.
3. L.F. He, B. Valderrama, A.-R. Hassan, J. Yu, M. Gupta, J. Pakarinen, H.B. Henderson, J. Gan, M.A. Kirk, A.T. Nelson, M.V. Manuel, A. El-Azab, and T.R. Allen, 2015, "Bubble formation and Kr distribution in Kr-irradiated UO_2", *Journal of Nuclear Materials*, Vol. 456, pp. 125–132.

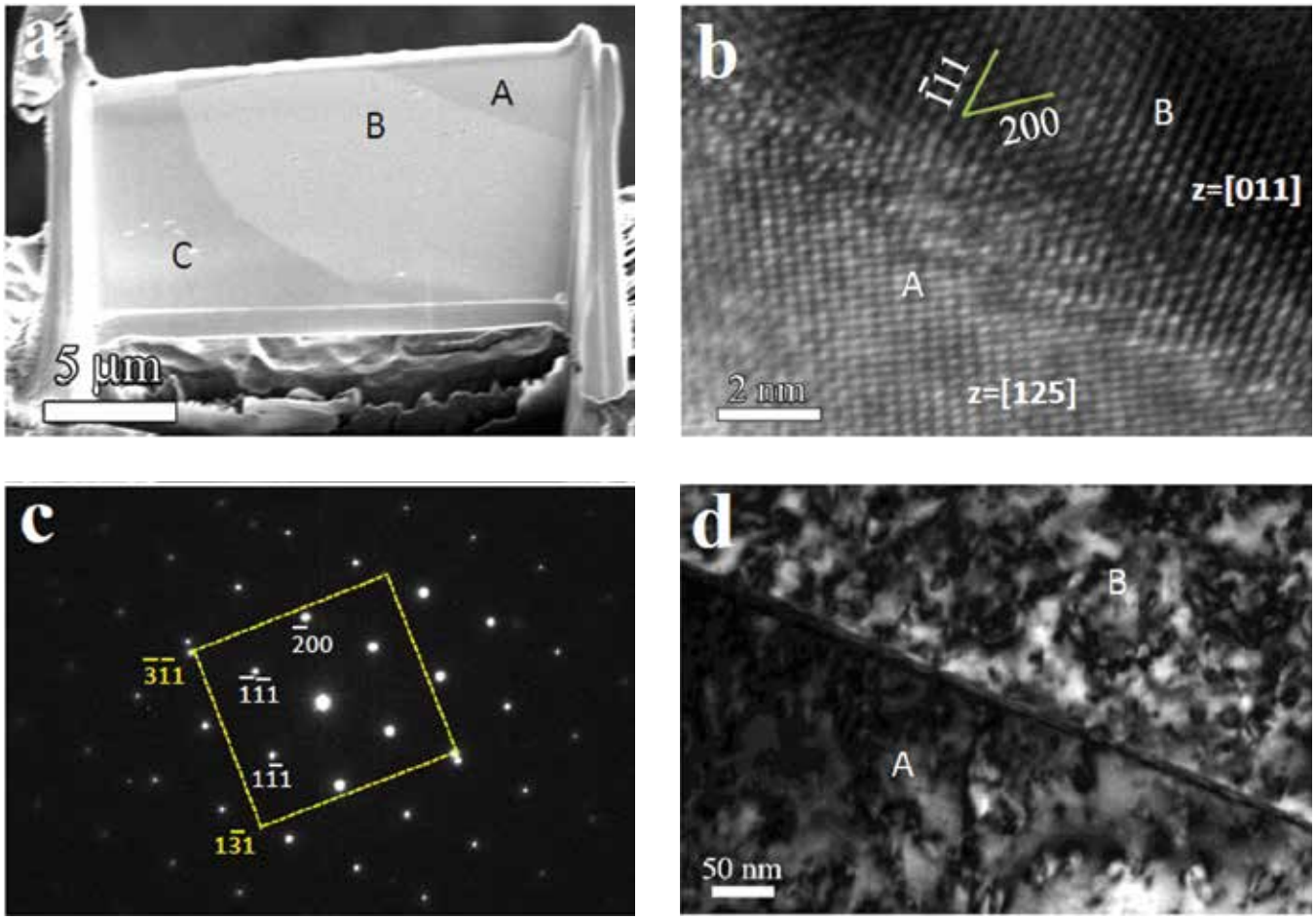


Figure 4. (a) A scanning electron microscope (SEM) image of a TEM lamina prepared by FIB showing three grains, A, B, and C. (b) High-resolution TEM image showing the grain boundary between grains A and B, the orientations of grains A and B are close to [125] and [011], respectively. (c) Selected-area diffraction pattern of grains A and B. (d) Bright-field TEM images showing the dislocation loops in grains A and B that have been irradiated with 1 MeV Kr at 600°C at a dose of 5 × 10^{14} ions/cm^2.

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	Advanced Test Reactor, PIE facilities
Argonne National Laboratory	Intermediate Voltage Electron Microscopy (IVEM)-Tandem facility
Collaborators	
University of Wisconsin - Madison	Lingfeng He (principal investigator), Mahima Gupta (collaborator)
Idaho National Laboratory	Jian Gan (collaborator)
Argonne National Laboratory	Marquis Kirk (collaborator)

X-ray Absorption Near Edge Spectroscopy and Extended X-ray Absorption Fine Structure Study of Technetium-99 Relevant to the Nuclear Fuel Cycle

Silvia Jurisson – University of Missouri – jurissons@missouri.edu

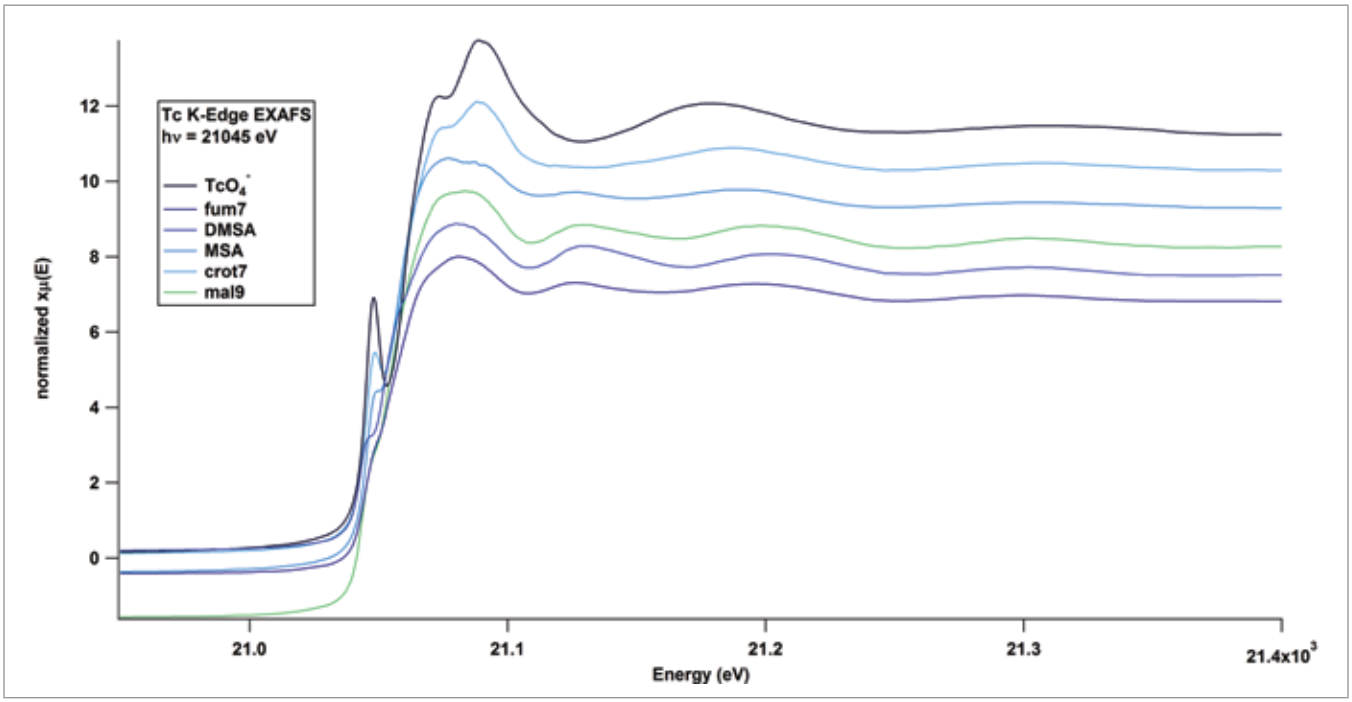


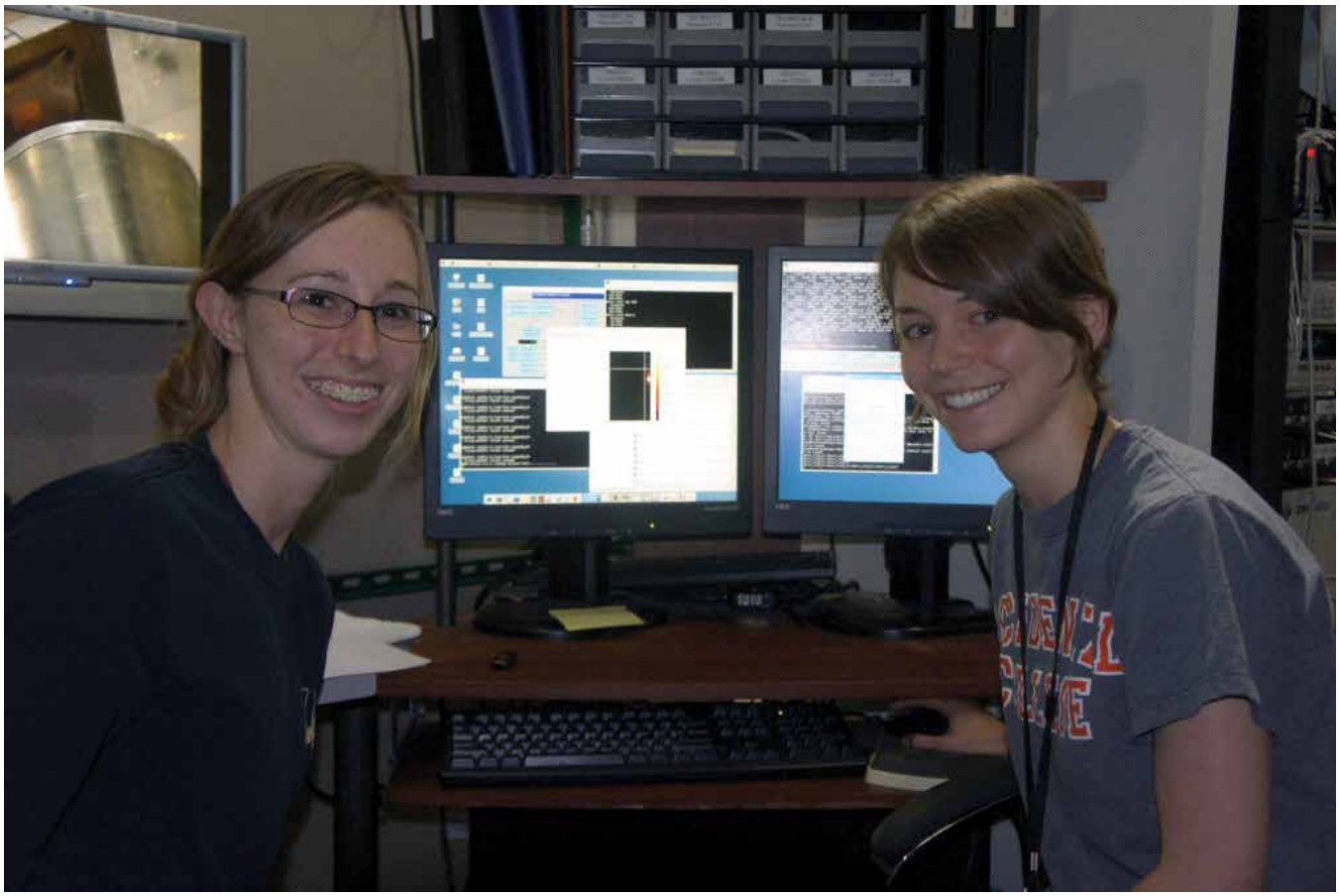
Figure 1. Tc K-edge EXAFS spectra of the standard complexes and experiment samples.

The synchrotron radiation techniques found in x-ray absorption near-edge spectroscopy (XANES) and extended x-ray absorption fine-structure spectroscopy (EXAFS) are among the very few that provide oxidation-state and nearest-neighbor information about compounds and materials that do not crystallize suitably or are not available in sufficient quantities for x-ray diffraction analysis (XRD).

Technetium-99 (Tc-99) is an important radionuclide in the nuclear fuel cycle. It is a major fission by-product of both uranium (U) and plutonium (Pu) and serves a vital role in environmental remediation at nuclear sites such as the Department of Energy's reactor near Hanford, Washington.

Project Description

We proposed to perform XANES and EXAFS experiments at the Advanced Photon Source (APS) at Argonne National Laboratory (ANL) to determine the end products of immobilization reactions between pertechnetate (TcO_4) and sulfide in the presence of various olefinic acids. The results will allow us to determine the oxidation states of technetium (Tc) as well as those of its nearest neighbors in the Tc metal center (i.e., its coordination environment). The resulting data will provide mechanistic



Graduate students Kim Reinig (left) and Rachel Seibert (right) in the Advanced Photon Source at Argonne National Laboratory.

information on the factors influencing the immobilization reactions that are important to both environmental remediation and nuclear fuel reprocessing.

We believe that understanding the products of these Tc reactions will enable the development of better immobilization methods and, potentially, superior separation methods. Since examining the Tc compounds should be a fairly straightforward process, our goal is to analyze this data quickly. In order to expedite the development of a base of information for further exploration, we have proposed that the work and analysis be performed in 2013 and 2014.

Accomplishments

Under the guidance of Jeff Terry at the Illinois Institute of Technology, graduate students Kim Reinig and Rachel Seibert ran the EXAFS and XANES spectra at the APS.

They used X-ray absorption spectroscopy (XAS) to investigate the reaction of TcO_4 with the unsaturated acids maleic acid and fumaric acid in the presence of sulfide. Nuclear magnetic resonance (NMR) of these reactions has shown that sulfide reacts across the double bond, forming mercaptosuccinic acid (MSA) or dimercaptosuccinic acid (DMSA). The resulting mercaptoacids then chelate the Tc and hinder or even prevent its immobilization to technetium heptasulfide (Tc_2S_7). They also prevent or hinder Tc's adsorption into another mineral's surface.

Tc_2S_7 is the product formed through the reaction of TcO_4 with sulfide in the absence of unsaturated acids. The Tc standard complexes, $[TcO(MSA)_2]^{1-}$ and $[TcO(DMSA)_2]^{1-}$, were used as comparators for the reactions of maleic acid and fumaric acid with TcO_4 in the presence of sodium sulfide (Na_2S).

The following seven solid samples were analyzed by Reinig and Seibert at the APS.

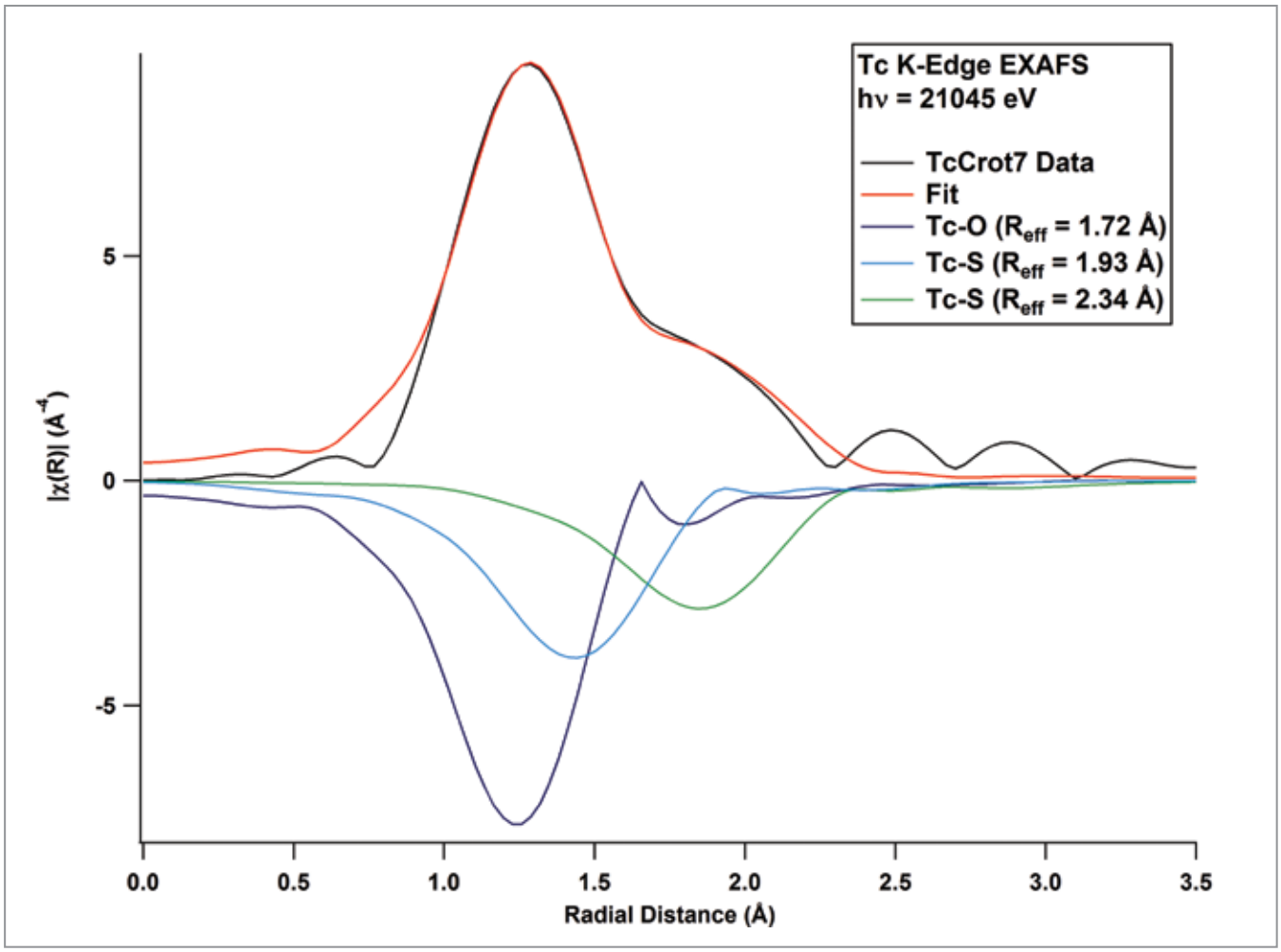


Figure 2. Fourier Transformation of the EXAFS spectra for the crotonic acid reaction.

1. (NH_4TcO_4)
2. $Na[Tc^{v}O(MSA)_2]$
3. $Na[Tc^{v}O(DMSA)_2]$
4. Tc_2S_7
5. TcO_{4^-} + SH^- + maleic acid at pH 7
6. TcO_{4^-} + SH^- + fumaric acid at pH 9
7. TcO_4^- + SH^- + crotonic acid at pH 7

Reinig and Seibert also investigated the effects unsaturated carboxylic acids had on the reaction of TcO_4^- with sulfide, which followed on the project's previous work [1]. The reaction that formed Tc_2S_7 is shown below. A phosphate buffer was used to control the pH.

$$2\ ^{99}TcO_{4^-} + 7\ SH^- + H_2O \longleftrightarrow Tc_2S_7 + 9\ OH^-$$

In the presence of unsaturated carboxylic acids, the sulfide appears to be building across the double bond to form the mono- or dithiol-carboxylic acids, but only when TcO_4^- is present. The product formed during the above reaction is a pentavalent technetium complex [Tc(V)oxo] species containing the newly formed mono- or dithiol-carboxylic acid. NMR spectra of the end products of the purified reaction show the presence of these species (compared to the standard complexes). EXAFS and XANES helped confirm the identities of the end products.

EXAFS and XANES were also used to determine whether a Tc-S bond similar to that observed in $Tc_vO(MSA)_2$ or $Tc_vO(DMSA)_2$ occurred. Comparisons of each sample's EXAFS spectra to the control standards showed profiles similar to the $[TcO(MSA)_2]^{1-}$ and $[TcO(DMSA)_2]^{1-}$ samples (Figure 2).

This work will provide important data on the immobilization reactions of the Tc-based compounds that are important to both environmental remediation and nuclear fuel reprocessing.

The crotonic acid sample contains a TcO_4 contaminant, but also shows evidence of a Tc-S bond (Figure 3).

Future Activities

The collected data continues to be analyzed, and a manuscript describing the chemistry of TcO_4 with sulfide in the presence of unsaturated acids is being drafted. The EXAFS and XANES data along with the 500-MHz NMR spectra will show that the presence of unsaturated acids can hinder the immobilization of Tc-99.

References

[1.] Y. Liu, J. Terry, S. Jurisson, 2009, "Potential Interferences on the Pertechnetate-Sulfide Immobilization Reaction,". *Radiochimica Acta*, Vol. 97, pp. 33–41.

Publications and Presentations

1. We are working on the manuscript and plan to submit it in 2015.

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Argonne National Laboratory	Advanced Photon Source
Collaborators	
University of Missouri	Silvia Jurisson (principal investigator), Kimberly Reinig (graduate student, collaborator)
Illinois Institute of Technology	Jeff Terry (co-principal investigator), Rachel Seibert (graduate student, collaborator)

"The EXAFS/XANES experiments at the APS allowed us to confirm the coordination environments about the Tc(V) centers on reaction of pertechnetate with sulfide in the presence of the unsaturated acids. Without these results, the product identities would have been more speculative."

— Dr. Silvia Jurisson, Professor of Chemistry and Radiology, University of Missouri

Post-Irradiation Examination of ATR-Irradiated Ultra-Fine Grained Steel

K. Linga Murty – North Carolina State University – murty@ncsu.edu

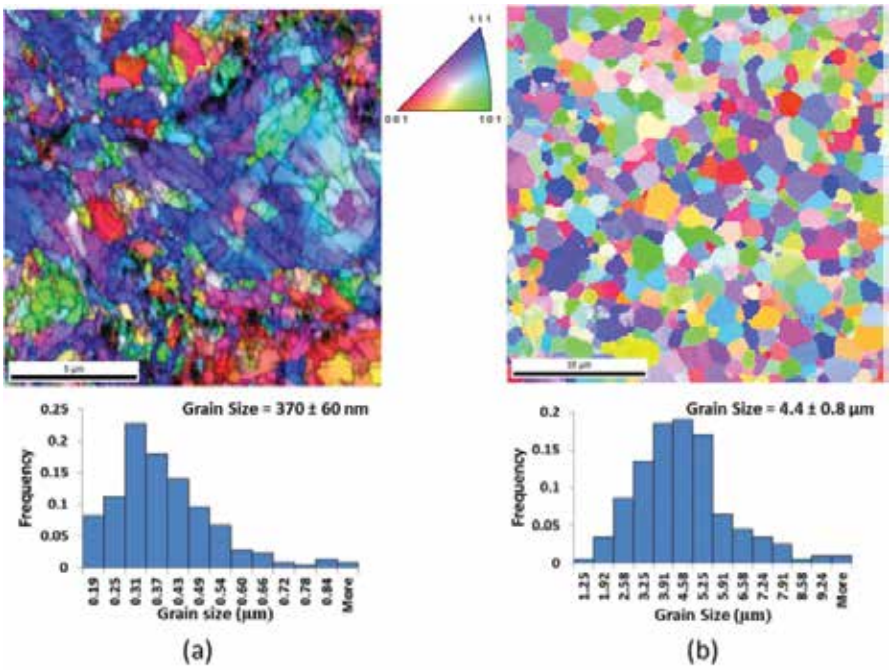


Figure 1. Pre-irradiation EBSD patterns and grain size distributions for (a) UFG and (b) CG steels.

Currently, 436 nuclear reactors currently provide an average of about 20% of the energy in the 30 countries that have nuclear power programs [1]. However, the structural materials that will be used for the new, advanced reactor systems will be subjected to far higher neutron fluences (150 dpa) than those in today's operating reactors (50 dpa) [2]).

Therefore, it is imperative that we develop new, more-radiation-tolerant materials and investigate the effects irradiation has on them. Ultra-fine grained (UFG) materials have shown promising mechanical properties, and extensive research has been conducted on the different processing techniques that could improve their properties across various applications [3].

Previous studies have shown that the high densities of grain boundaries in UFG materials act as sinks for irradiation-induced defects, reducing the influence of the irradiation defects [4].

Project Description

In this study, researchers investigated the effects of neutron irradiation on UFG ferritic steel that had been prepared using the equal-channel-angular-pressing (ECAP) technique [3, 4]. The experimental material is an ECAPed, low-carbon, mild steel consisting, by weight, of 0.1% carbon (C), 0.5% manganese (Mn), 0.27% silicon (Si), and the balance (99.13%) iron (Fe). Both UFG samples along with their conventional grain (CG) steel counterparts—prepared by annealing UFG samples at 800ºC for 1 hour—were irradiated to 1.37 displacements per atom (dpa) in ATR at INL. The irradiation capsules were designed to keep the irradiation temperature of the samples below 100ºC. The mean grain sizes for the UFG and CG steels are 0.35 μm ±0.18 μm and 4.4 μm ±1.8 μm, respectively (Figure 1).

Accomplishments

After irradiation, microstructural and mechanical properties of the CG and UFG steels were investigated using transmission electron microscopy (TEM), electron back scattered diffraction (EBSD), atom probe tomography (APT), x-ray diffraction (XRD), and micro hardness and tensile testing. TEM micrographs showed no grain growth post irradiation in the UFG steel. ESBD showed similar results for CG steel. XRD was used to determine the dislocation density for both steels before and after irradiation by fitting the XRD patterns to a pseudo-Voigt (pV) function using the Modified Rietveld technique [5].

CG steel exhibited an increase in the dislocation density post irradiation. On the other hand, with values well within error bars, UFG steel showed no significant change. APT analysis revealed a high number of nano Mn-Si-enriched precipitates in both CG and UFG steels (Figure 2). However,

	ρ Dose(dpa)	ρ(m^{-2})	Yield Strength (MPa)	Tensile Strength (MPa)	Vickers Hardness (MPa)	Ductility (%)
CG	0	1.06 (± 0.13)×10^{14}	296±17	390±18	1284±13	63±3
	1.37	4.26 (± 0.56)×10^{14}	687±20	754±20	2081±51	11±1
UFG	0	9.50 (± 1.24)×10^{14}	775±9	980±11	3088±54	18±2
	1.37	8.98 (± 1.39)×10^{14}	1009±49	1060±43	3353±68	8±1

Table 1. Mechanical properties for both CG and UFG steels before and after irradiation.

while the radii of the clusters in both steels are similar (0.97 μm ±0.23 μm and 0.9 μm ±0.16 μm for UFG and CG steels, respectively) the density of clusters in the UFG steels (1.2 ×10^{24} m^{-3}) is about twice that of CG steel (6.7×10^{23} m^{-3}) (Figure 3).

Researchers believe this is due to the shorter distance defects have to diffuse in UFG before they reach the grain boundary and the resulting lower probability of defect recombination in the matrix. No clusters were observed before irradiation, indicating that their formation was radiation-induced. Vickers micro hardness, tensile test and dislocation density results are shown in Table 1.

The average micro hardness values for CG steel increased by 62% after irradiation compared to only 8.6% for the UFG steel (Figure 4). Tensile test results revealed that CG yield strength increased by 132% after irradiation and its ductility decreased by 82%, while the yield strength of UFG steel increased by 30% and the ductility reduced by 56% (Figure 5). Although irradiation hardening was minute in the UFG steel compared to its CG counterpart, the irradiation-induced embrittlement is clearly apparent in the UFG steel after irradiation, albeit the percentage decrease in the ductility of UFG steel is quite less than that of its CG counterparts.

	$\sigma_{Oro\text{-}Ash}$ (MPa)	σ_{Dis} (MPa)	$\sigma_{Calculated}$ (MPa)	$\sigma_{Measured}$ (Mpa)
CG Steel	137± 44	245 ± 35	382 ± 57	391 ± 27
UFG Steel	230 ± 90	-20 ± 73	210± 115	234 ± 50

Table 2. Estimated strength increments for different strengthening mechanisms for both CG and UFG steels.

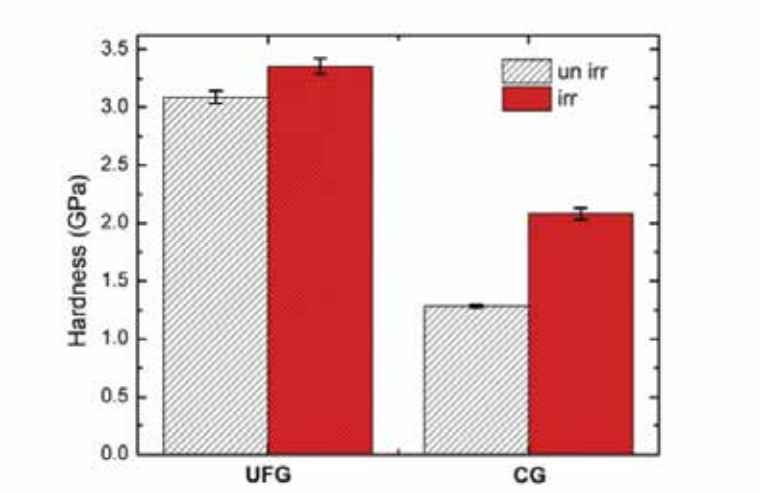


Figure 4. Micro hardness before and after irradiation for both UFG and CG low carbon steel

According to Odette and Lucas [6], the primary mechanism for embrittlement in ferritic steels is the hardening produced by nanometer-sized features that develop as a consequence of radiation exposure. Since our results showed that there is no significant change in dislocation density in UFG steel after irradiation, the observed hardening in UFG steel is likely a product of the high density of irradiation-induced, Mn-Si-enriched clusters found in UFG steel (Table 2).

To gain a better understanding of the effect of irradiation on microstructural changes in the steel's mechanical properties, refer to Alsabbagh et. al's 2014 journal article [7] for a discussion of how the increase in yield stress

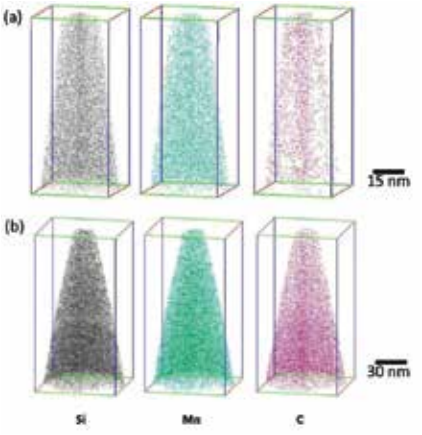


Figure 2. Representation of a three-dimensional (3D) reconstruction of CG low-carbon steel, (a) before and (b) after neutron irradiation by 3D atom-probe microscopy.

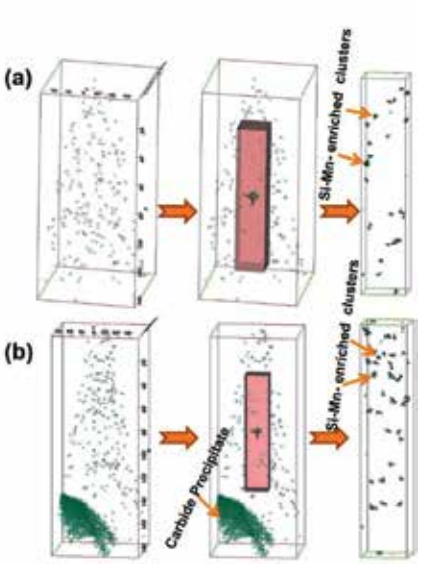


Figure 3. Si-Mn-enriched cluster distribution post-neutron irradiation for (a) CG and (b) UFG steel. The dimension of the interior (colored) boxes is 20 x 20 x 100 nm.

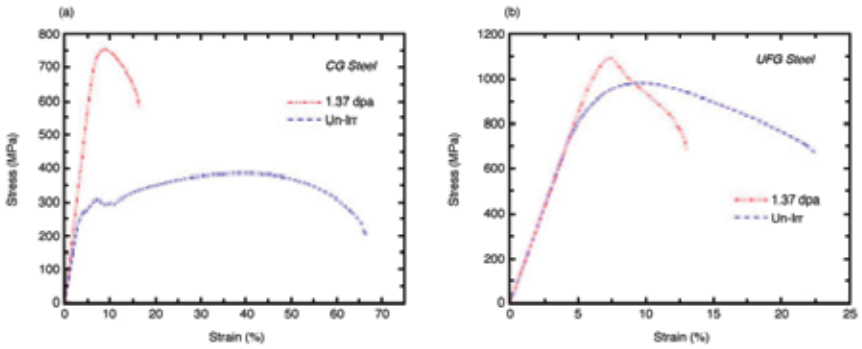


Figure 5. Engineering stress strain curves for (a) UFG and (b) CG before and after irradiation to 1.37 dpa.

Nanocrystalline and UFG metals with relatively large volumes of interfaces are expected to be more radiation resistant than conventional metals.

is related to different strengthening mechanisms, for example strengthening due to grain size, solid solution, clusters and dislocations.

The demonstrated agreement between the experimental results and the actual physical strength mechanisms indicates that the Orowan-Ashby and Taylor strengthening models can be useful for explaining how nanocluster strengthening and dislocation forest hardening mechanisms contribute to the overall strength of the ECAP UFG and the CG steels. The results show that while irradiation-induced dislocation density is an important factor in the increase in total yield stress in neutron-irradiated CG steel, irradiation hardening in the UFG steel is mainly due to the irradiation-induced clusters.

In summary, as the area of grain boundaries (which act as sinks for radiation-induced defects) is significantly increased by grain refinement, UFG steel showed better irradiation tolerance compared to its CG counterparts. However, irradiation-induced solute clustering in UFG alloys needs to be carefully considered.

Future Activities

Investigations are underway at the Center for Advanced Energy Studies (CAES) and INL to determine the effects of irradiating UFG and CG steels to higher doses, which will shed light on the effects of dose on strengthening and embrittlement.

Publications and Presentations*

1. A. Alsabbagh, A. Sarkar, B. Miller, J. Burns, L. Squires, D. Porter, J. Cole, and K. Murty. 2014, "Microstructure and mechanical behavior of neutron irradiated ultrafine grained ferritic steel," *Materials Science & Engineering A*, Vol. 615, pp. 128–138.

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[2.] S.J. Zinkle, J.T. Busby, 2009, "Structural materials for fission & fusion energy," *Materials Today*, Vol. 12, No. 11, pp. 12–19.

[3.] T.C. Lowe, R.Z. Valiev, 2004, "The use of severe plastic deformation techniques in grain refinement," *JOM*, Vol. 56, No. 10, pp 64–68.

[4.] A. Alsabbagh, R.Z. Valiev, and K.L. Murty, 2013, "Influence of grain size on radiation effects in a low carbon steel," *Journal of Nuclear Materials*, Vol. 443, pp. 302.

[5.] L. Lutterotti and P. Scardi, 1990, "Simultaneous structure and size–strain refinement by the Rietveld method," *Journal of Applied Crystallography*, Vol. 23, pp. 246.

[6.] G.R. Odette and G.E. Lucas, 2001, "Embrittlement of nuclear reactor pressure vessels, *JOM*, Vol. 53, No. 7, pp. 18–22.

[7.] A. Alsabbagh, A. Sarkar, B. Miller, J. Burns, L. Squires, D. Porter, J.I. Cole, K.L. Murty, 2014, "Microstructure and mechanical behavior of neutron irradiated ultrafine grained ferritic steel," *Materials Science and Engineering A*, Vol. 615, pp. 128.

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	Advanced Test Reactor, PIE facilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
North Carolina State University	K.L. Murty (principal investigator), Ahmad Alsabbagh (graduate student)
Idaho National Laboratory	Doug Porter (principal investigator), Brandon Miller (collaborator)

"This collaborative research enabled me to work with advanced microstructural characterization techniques through the ATR-NSUF program. I was also able to learn from many working professionals at CAES and INL that resulted in my receiving the 2014 ANS Mark Mills Award for a technical article I wrote based on the research I did to earn my doctoral degree in nuclear science and technology."

— Ahmad Alsabbagh, Ph.D., Graduate Student until June 2014, Nuclear Engineering, North Carolina State University

Microstructural and Mechanical Characterization of Self-Ion Irradiated 14LMT Nanostructured Ferritic Steel

Indrajit Charit – University of Idaho – icharit@uidaho.edu

This work could lead to the development of high-performance fuel cladding materials for advanced fast reactors.

Advanced reactors need high-performance materials to serve under harsh conditions such as higher temperature, higher radiation doses, and extremely corrosive environments. Nanostructured ferritic steels (NFS) are such a class of materials. They are produced by mechanical alloying (MA) of the elemental (or pre-alloyed) metallic powder, typically incorporating nanosized yttria (Y_2O_3) powder followed by a traditional consolidation process such as hot extrusion or hot isostatic pressing (HIP). NFS have excellent potential for advanced fuel cladding and structural materials applications in fast reactors.

Project Description

Since neutron irradiation is out of the scope of this Rapid Turnaround Experiment (RTE), the aim of the project is to investigate a new NFS known as 14LMT (Fe-14Cr-1Ti-0.3Mo-0.5La_2O_3, wt.%) that was recently developed by this research group and other collaborators. Samples were irradiated at doses up to 100 displacements per atom (dpa) and relevant microstructural characterization and mechanical properties evaluations were performed.

Titanium is generally added to Y_2O_3 to form complex yttria-titanium-oxygen (Y-Ti-O)-based particles in order to make the dispersed oxides much finer, and stable at elevated temperatures. The 14LMT alloy uses lanthana (La_2O_3) instead of the traditional Y_2O_3. Spark plasma sintering (SPS) was used to consolidate the mechanically alloyed powder.

NFS performance is largely determined by the ultra-high number density of nanosized oxide particles dispersed throughout the microstructure. These nanofeatures are stable at high temperatures and are expected to impart excellent high-temperature strength and irradiation stability to NFS. This work could lead to the development of high-performance fuel cladding materials for advanced fast reactors.

Accomplishments

The stability of nanoclusters in NFS under irradiation is critical. Collision cascades can eject solute atoms from them and change their physical characteristics. In order to understand their stability, the lanthana-bearing 14LMT alloy was exposed to self-ion (Fe^{2+}) irradiation at both room temperature (30°C) and elevated temperature (500°C) as a function of ion dose at 10, 50, and 100 dpa. Subsequently, the irradiated material was characterized by transmission electron microscopy (TEM) for microstructural characteristics, atom probe tomography (APT) for nanocluster size/compositional analyses, and nanoindentation to measure hardness.

Overall morphology and number density of the nanofeatures remained largely unchanged after irradiation. The average radius of the nanofeatures in the sample irradiated at 500°C/100 dpa was slightly reduced. A notable level of irradiation hardening and enhanced dislocation activity occurred after ion irradiation, except at 30°C and ≥50 dpa. Other microstructural features such as grain boundaries and a high density of dislocations also provided defect sinks to assist in defect removal. A comprehensive paper based on this work is under review with the Journal of Nuclear Materials.

Future Activities

While this project has been completed, a new RTE project (Microstructural and Nanomechanical Characterization of a Lanthana-Bearing Nanostructured Ferritic Steel Irradiated with High Dose Iron Ions) has recently been approved. This will allow the project team to continue its work on understanding ion-irradiation response of 14LMT to higher dose levels (up to 400 dpa).

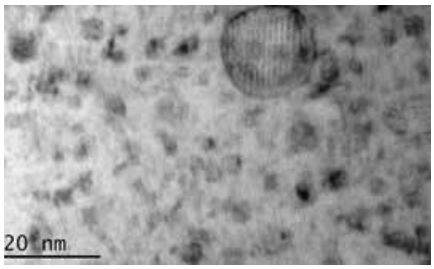

Figure 1. A TEM image of the oxide particles observed in the 14LMT alloy specimen unirradiated at 500°C.

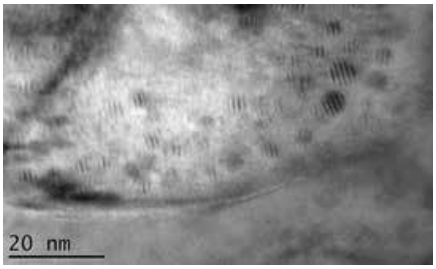

Figure 2. A TEM image of the 14LMT alloy specimen irradiated at 500°C/100 dpa.

Publications and Presentations*

1. S. Pasebani, I. Charit, J. Burns, S. Alsagabi, D.P. Butt, J.I. Cole, L.M. Price, and L. Shao, "Microstructural stability of a self-ion irradiated lanthana-bearing nanostructured ferritic steels," *Journal of Nuclear Materials*, under review.
2. S. Pasebani, I. Charit, 2014, "Effect of alloying elements on the microstructure and mechanical properties of nanostructured ferritic steels produced by spark plasma sintering," *Journal of Alloys & Compounds*, 599, pp. 206–211.
3. S. Pasebani, 2014, *Processing of Oxide Dispersion Strengthened Alloys via Mechanical Alloying and Spark Plasma Sintering*, Doctoral Dissertation: University of Idaho, Moscow, Idaho.

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Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
University of Idaho	Indrajit Charit (principal investigator), Somayeh Pasebani (collaborator)
Boise State University	Darryl Butt (collaborator)
Boise State University/ Center for Advanced Energy Studies	Yaqiao Wu (collaborator), Jatu Burns (collaborator)
Texas A&M University	Lloyd Price (collaborator)
Idaho National Laboratory	James Cole (principal investigator)

The ATR NSUF MaCS microstructural characterization facility provides easy access to state-of-the-art instruments that are not easily available at our campus. This has been extremely beneficial to our research productivity.

— Indrajit Charit, Associate Professor, University of Idaho

Microstructural and Mechanical Characterization of Self-Ion Irradiated Grade 92 Steel

Indrajit Charit – University of Idaho – icharit@uidaho.edu

This experiment could lead to a better understanding of Grade 92's irradiation performance in fuel cladding and structural materials for advanced reactors.

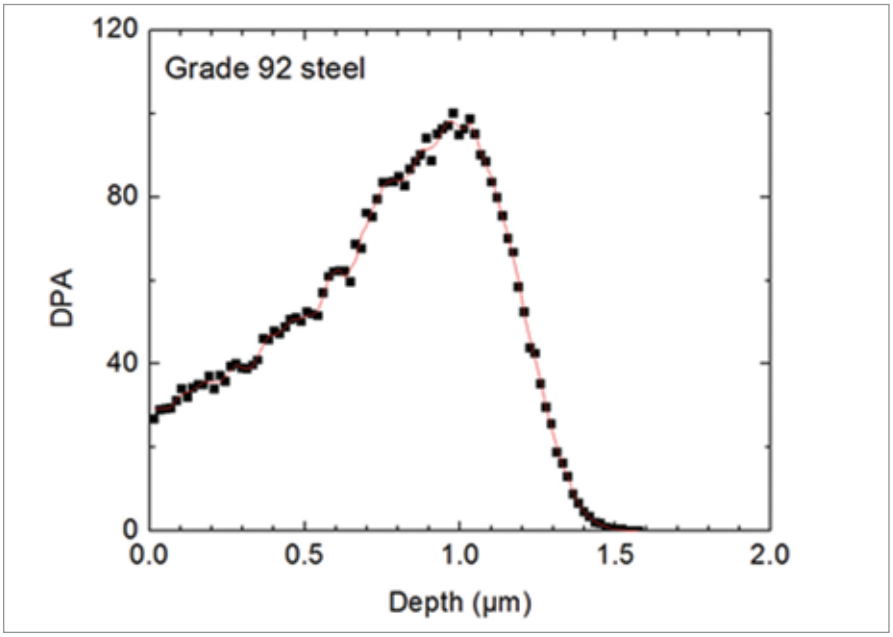


Figure 1. The calculated irradiation damage profile in terms of dpa versus length.

Ferritic-martensitic (F-M) steels have the potential to be employed in advanced reactors. One of these, Grade 92 (9Cr-2W) steel, is being considered for structural applications in advanced nuclear energy systems. It has good mechanical and thermophysical properties, and is considered to have potential for both in-core and out-of-core applications. Still, the irradiation performance of this alloy is not fully understood because of limited available data.

This experiment could lead to a better understanding of Grade 92's irradiation performance in fuel cladding and structural materials for advanced reactors. If proven successful, high-performance materials such as Grade 92 steel would help improve the safety and reliability of future reactors, and give them a longer service life.

Project Description

In this project, Grade 92 samples were irradiated with high energy Fe^{2+} ions at the Texas A&M University Ion Beam Laboratory, and the induced microstructural evolution and mechanical properties that resulted were investigated. Irradiated specimens were prepared for transmission electron microscopy (TEM) using a focused ion beam (FIB). Microstructural characterization of the irradiated specimens was performed using TEM, and mechanical properties were evaluated using the nanoindentation technique.

Accomplishments

Grade 92 steel specimens were irradiated to 10, 50, and 100 displacements per atom (dpa) using the IoneX 1.7 MV Tandetron accelerator. This effort was coordinated by Professor Lin Shao and graduate student Lloyd Price.

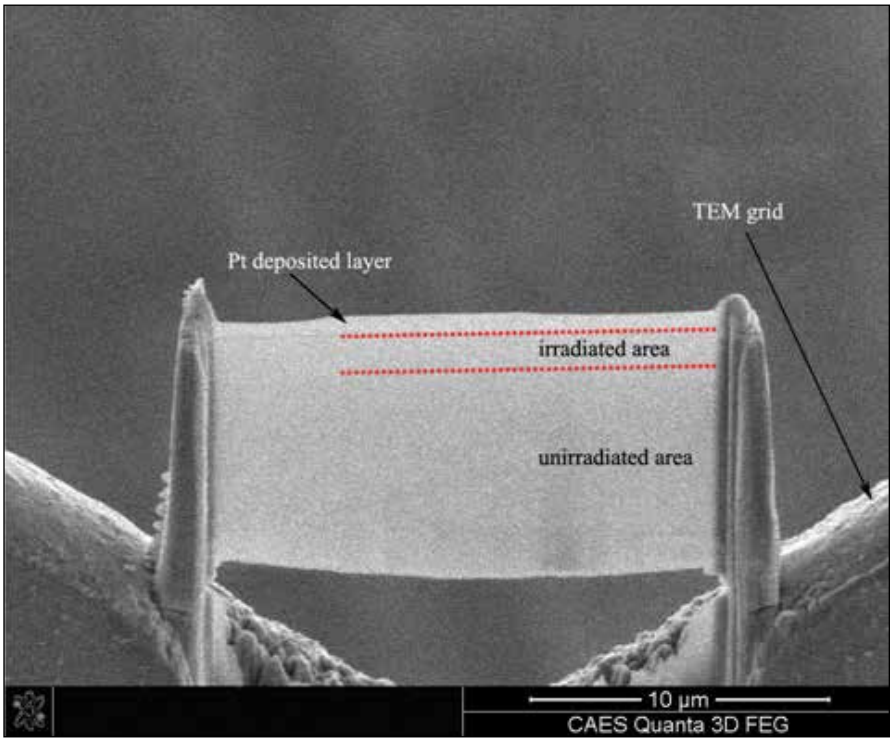


Figure 2. The FIB sample showing the Pt-deposited layer, irradiated and unirradiated areas.

The experiments were conducted at temperatures of 30°C and 500°C. The damage profile as calculated from Stopping and Range of Atoms in Matter (SRIM) is shown in Figure 1.

The irradiated samples were then prepared for TEM studies using a focused ion beam - scanning electron microscope (FIB-SEM) Quanta 3D field emission gun (FEG), in the CAES MaCS facility. Ms. Jatu Burns assisted graduate student Sultan Alsagabi in this effort. The FIB-SEM procedure started with protecting the irradiated surface with a platinum (Pt) deposition layer. Figure 2 shows the specimen after irradiation.

For detailed microstructural characterization, a TF30 FEG scanning TEM (STEM) was utilized, operating at an accelerating voltage of 300 kV. The FIB samples were attached to a copper grid before examination inside the TEM. The dislocation density of the prepared samples was estimated while they were oriented in a two-beam condition, and the electron energy loss spectrum (EELS) technique was applied to measure sample thickness.

TEM images of the irradiated samples were obtained from the irradiated area of the FIB sample. Samples from the unirradiated area were also examined for comparison purposes. Graduate student Alsagabi completed this task with the assistance of Dr. Yaqiao Wu, of the CAES MaCS staff.

The irradiation-induced damage evolution in the samples at 30°C as a function of damage dose (dpa) is shown in Figure 3. The irradiated samples did not show any distinct irradiation-induced precipitates, dislocation loops, or voids. However, they do show the presence of irradiation-

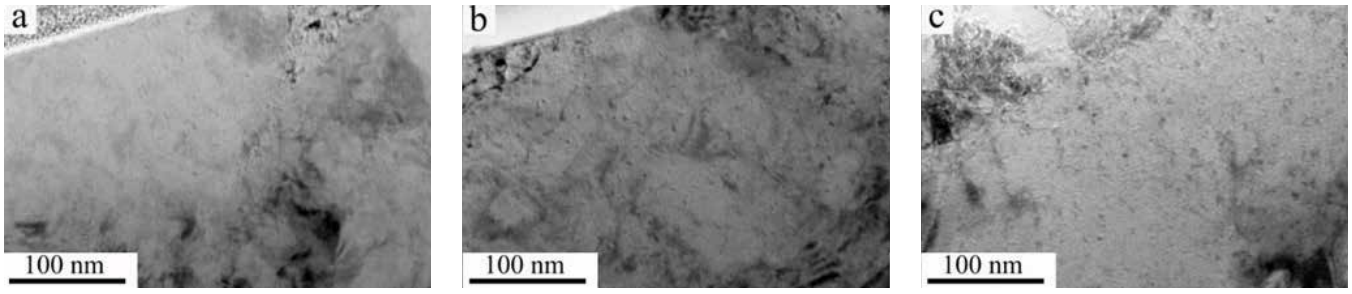


Figure 3. TEM images of irradiated Grade 92 specimens under two-beam bright field conditions at a) 10 dpa/30°C, b) 50 dpa/500°C, and c) 100 dpa/500°C.

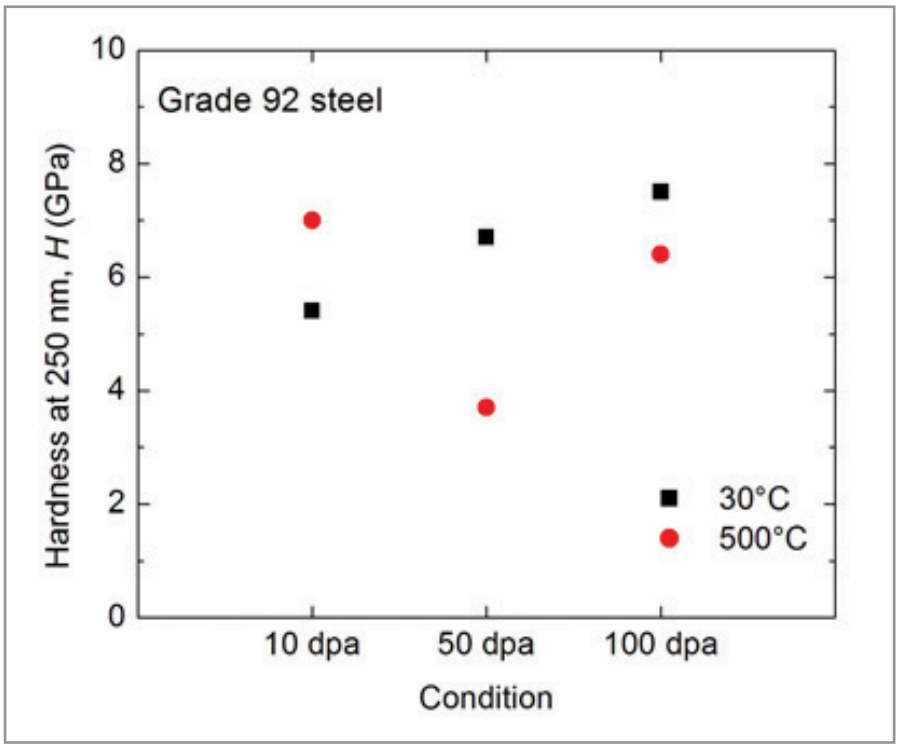


Figure 4. Hardness of Grade 92 steel as a function of irradiation dose at 30°C and 500°C.

induced defect clusters, appearing as 'black dots.' The overall concentration of these clusters increased as the dose was increased. The high density of induced clusters did not appear outside the irradiated areas.

The unirradiated area did not show any form of clustering at all, which confirms that these clusters were not induced by the FIB.

In samples irradiated at 10 dpa/30°C, these defect clusters were very small in size. They increased to around 3–4 nanometers (nm) in samples irradiated at 50 dpa and 100 dpa. The increased irradiation dose enhanced not only the formation of induced defect clusters, but also the number and density of these defects, which saturated the irradiated areas in later stages. This can be attributed to the limited mobility of clusters at 30°C.

Finally, a Hysitron Nanoindenter (model TI-950 TriboIndenter) was used to measure hardness as a function of depth. The indentation was made in a direction normal to the surface of the sample using a constant time segment to induce a penetration depth profile of 1000 nm. Samples irradiated at 30°C showed an evident hardening phenomenon, with the hardness increasing as the dose increased. However, at 500°C the irradiation hardening was less pronounced.

Clearly, the higher irradiation dose created greater hardness, especially in the near-surface region (less than 300 nm), so the monotonic increase of hardness with increased fluence was obtained. However, the material irradiated at 10 dpa/500°C exhibited higher hardness in the near-surface region than the material irradiated at 50 dpa/500°C. Figure 4 shows the hardness data plotted as a function of depth (at 250 nm), showing a gradual increase at 30°C. At 500°C the hardness gradually decreased, but then increased again during recovery. During the reported time under irradiation (417 min.), lower hardening was observed in the sample irradiated at 50 dpa/500°C, which indicates an enhanced recovery effect at 500°C.

The main results can be summarized as follows:

1. The irradiation-induced hardening was obvious, and increased as the dose was increased at 30°C, but was less definite at 500°C, as an annihilation mechanism becomes more pronounced. This was accompanied by a higher dislocation density of samples irradiated at 30°C.
2. The samples irradiated at 30°C showed irradiation-induced defect clusters appearing as dots in the irradiated area, and the density of these clusters increased as dose increased. Some dislocation loops were found in the sample irradiated at 50 dpa/500°C.
3. At 30°C the number and density of irradiation-induced clusters increased as dose increased, and at 50 dpa and 100 dpa they saturate to 3–4 nm.
4. The investigated samples did not show any cavities such as bubbles or voids under the testing parameters.

Future Activities

The Rapid Turnaround Experiment-Project (RTE) has been completed. No further work is planned at this time.

Publications and Presentations

1. S. Alsagabi, 2014, *High Temperature Deformation Behavior, Thermal Stability and Irradiation Performance in Grade 92 Steel*, Doctoral Dissertation: University of Idaho, Moscow, Idaho.

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
University of Idaho	Indrajit Charit (principal investigator), Sultan Alsagabi (collaborator)
Boise State University/ Center for Advanced Energy Studies	Jatu Burns (collaborator), Yaqiao Wu (collaborator)
Texas A&M University	Lloyd Price (collaborator), Lin Shao (collaborator)
Idaho National Laboratory	James Cole (principal investigator)

"The ATR-NSUF MaCs microstructural characterization facility provides easy access to state-of-the-art instruments. There is no doubt in my mind that this access has been very positive in our research and education."

— Indrajit Charit, Associate Professor, University of Idaho

Development of Advanced Crystallographic Analysis Technique for Localized Fission Product Transport in Irradiated Silicon-Carbide

Isabella van Rooyen – Idaho National Laboratory – isabella.vanrooyen@inl.gov

This study provides first-of-a-kind, nano-crystallographic information on neutron-irradiated TRISO fuels by the application of precession electron diffraction measurements.

The proposed study aims to perform compositional analysis to gain insight into a possible link between crystallographic information and fission products location in the silicon-carbide (SiC) layer of tristructural isotropic (TRISO) fuel. This information will help identify transport mechanisms for these fission products, especially silver (Ag), as they migrate through the SiC layer.

Recent work has shown that it is only possible to identify low concentrations of fission products in TRISO-coated particles, especially Ag, using transmission electron microscopy (TEM) with a field emission gun (FEG) [1]. The preliminary results of this study indicate that fission products are primarily associated with some, but not all, grain boundaries and triple junctions. Knowledge of the grain boundary crystallographic parameters will be required to explain the varied fission transport behaviors associated with individual grain boundaries.

Currently, no efficient way exists for measuring the grain boundary parameters while correlating them with fission product transport behavior. What is needed is a way of measuring both the chemistry at the grain boundary and the grain boundary parameters. Since the chemical analysis resolution has only been demonstrated in the FEG TEM, a technique of measuring the grain boundary parameters in the TEM is also required.

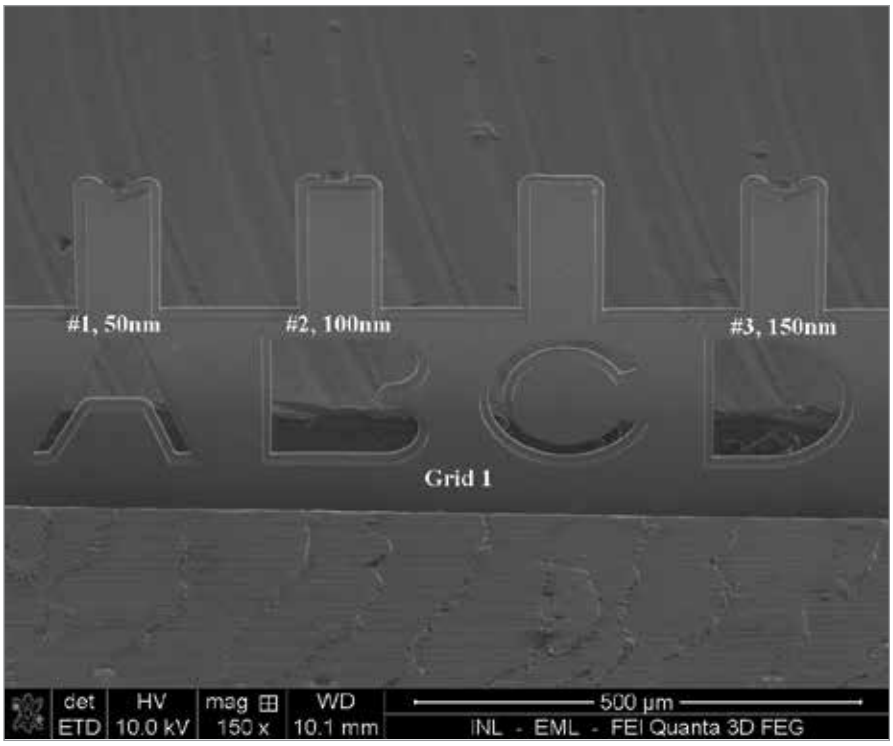
Attempts at measuring the grain boundary parameters using Kikuchi bands in TEM electron diffraction patterns under the Advanced Gas Reactor (AGR)-1 program have been unsuccessful. However, an emerging technique utilizing ASTAR software from NanoMEGAS SPRL, which examines the intensity of electron diffraction spots in precession electron diffraction (PED) patterns, shows potential for determining the necessary crystallographic parameters in the TEM,

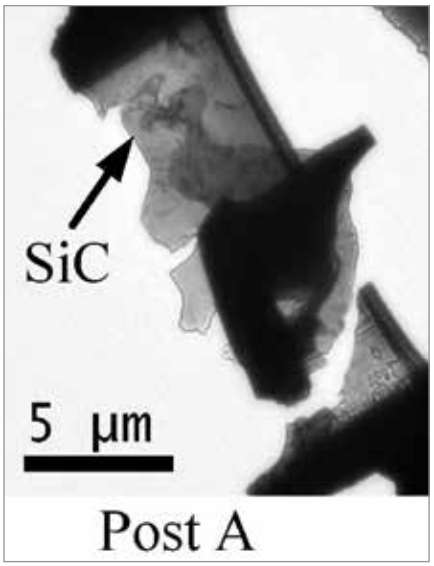
Project Description

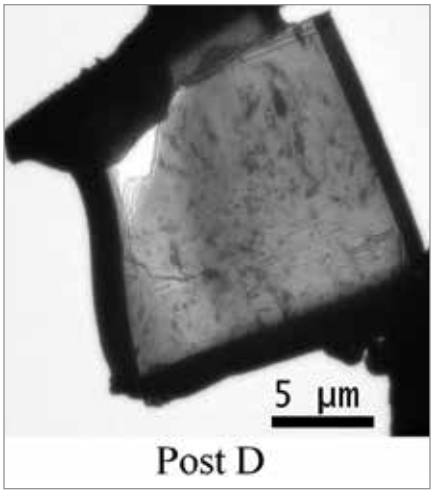
This project consists of two phases. In the first, researchers will develop the ASTAR technique and evaluate its resolution and reliability when examining SiC from an unirradiated TRISO fuel particle. The second phase, and the main focus of this project, will be to characterize the grain and grain boundary parameters in the SiC layer of irradiated TRISO fuel particles. Table 1 outlines the samples to be analyzed and the expected outcomes.

Phase 1: Research on unirriadiated SiC and sample optimization for ASTAR: Thicker samples increase the possibility of multiple grains being present in the focused ion beam (FIB) sample. Electron diffraction caused by these multiple grains degrades ASTAR's ability to identify the correct crystallographic orientation. Therefore, three TEM lamellae of unirradiated SiC with different thicknesses will be prepared using FIB techniques (50, 100 and 150 nm) at CAES or the Electron Microscopy Laboratory (EML). The quality of the ASTAR data will be evaluated for each thickness from the reliability map. The data obtained from these samples will also provide the baseline microstructure and crystallographic information (texture) of the as-fabricated SiC layer, which will then be compared to that of SiC from irradiated TRISO particles.

Phase 2: Research on irriadiated SiC The main objective is to demonstrate that ASTAR can be used to determine the crystallographic information of grains in irradiated SiC, determine the grain boundary parameters using the crystallographic orientations of adjacent SiC grains, and correlate that information with the transport of fission products. Samples have already







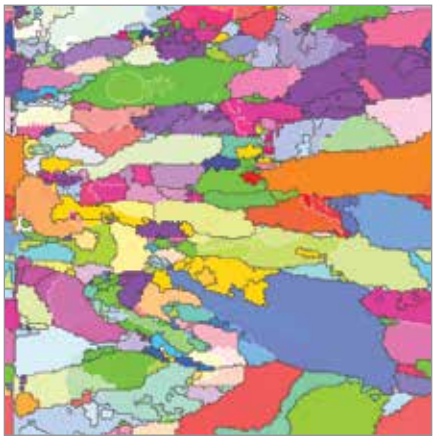


Figure 1. Three FIB-fabricated samples were prepared as shown in the micrograph left. The FIB lamellae at post B were lost due to handling and only lamellae from posts A (top right) and D (bottom right) were analyzed using ASTAR.

been fabricated using the Materials and Fuels Complex (MFC) EML-FIB from Particle 35 of Compact 6-3-2, which experienced 11.3% burnup during the AGR-1 experiment. Although, these samples may not be the optimum thickness, they will be sufficient to demonstrate ASTAR's ability to determine the orientation of individual, irradiated SiC grains and to reveal any other issues that may interfere with the accurate determination of the crystallographic orientation. The sample set includes one taken perpendicular to the growth direction of the SiC layer and three taken parallel. All these samples have already had a considerable amount of compositional analysis performed on them.

Ag has been identified in these samples, and several locations have been specifically identified for ASTAR analysis. Since the three parallel samples span the entire thickness of the SiC layer, the ASTAR data will show how the microstructure develops during the growth of the SiC layer. The data will also be used to determine the misorientation across specific grain boundaries, especially those that have previously been found to contain Ag. The misorientation information will be used to determine whether it is a high-angle, low-angle, random, or coincident site lattice (CSL)-related grain boundary. By assessing the crystallography of grain boundaries with and without fission products, it may be possible to make preliminary conclusions on the types of grain boundaries that are resistant to fission product transport. However, it is unlikely that a statistically relevant dataset will be collected during this RTE.

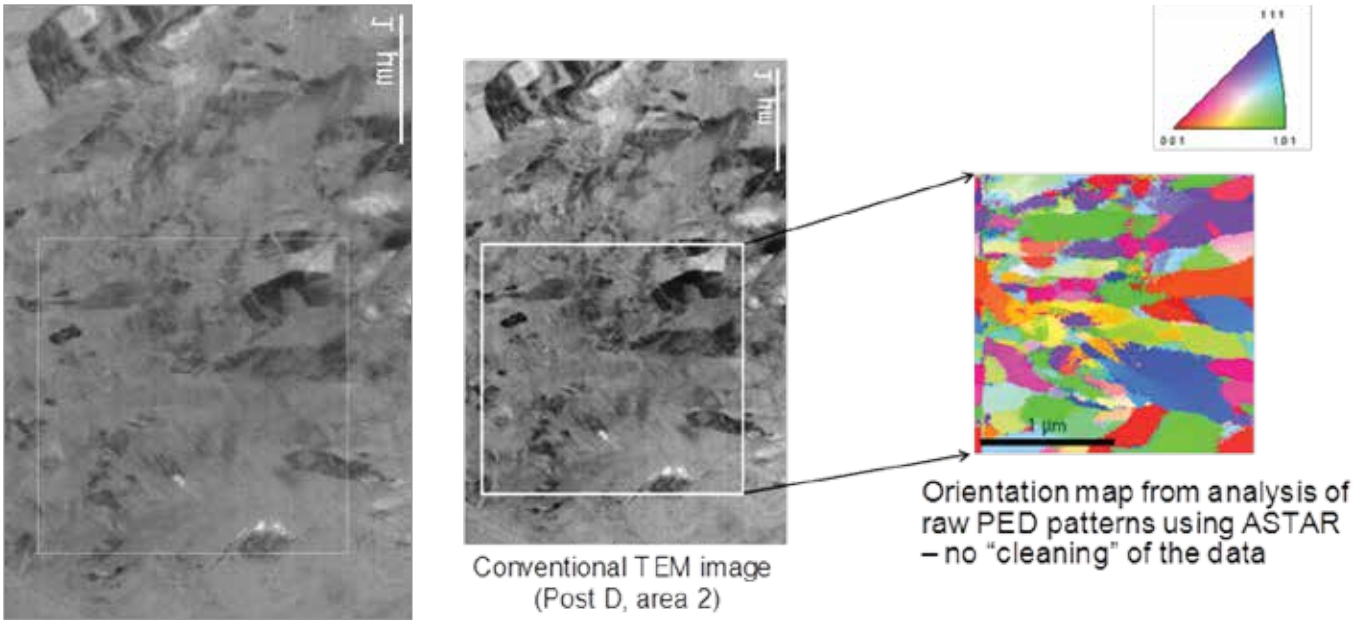


Figure 2. The crystallographic orientation of the area in the TEM image on the left is mapped on the right.

Accomplishments

The work was initiated in February 2014, resulting in the successful completion of all experimental work for the two phases, both on unirradiated and irradiated samples, as indicated in Table 1. The integration of all project results and papers is still to be performed, although some results have already been prepared for conference or workshop presentations. [2-4]

Summary of Phase 1 results: Research on unirriadiated SiC and sample optimization for ASTAR 2.3

Sample fabrication techniques as well as sample characteristics can critically influence the quality of the data collected. Researchers initially evaluated the influence of sample thickness on the quality of the orientation data generated by ASTAR. It is extremely difficult to prepare samples thinner than 80 nm from these materials due to the brittle nature of neutron-irradiated SiC. Although the original study plan included three FIB-prepared samples, only two samples of varying thickness (Figure 1) were analyzed. The thicknesses of five areas on these two samples were determined by electron energy loss spectroscopy (EELS), (e.g. location and height of the plasmon peak). Three areas were found to be approximately 80 nm thick, while two others were found to be approximately 120 nm thick. It is extremely difficult to prepare FIB lamellae thinner than the measured 80 nm due to the brittle nature of neutron-irradiated SiC, as demonstrated by the fractured regions shown in Figure 1 (center). The crystallographic information in the latter two areas was collected using the ASTAR system on the Tecnai TF30-FEG STwin at CAES.

The "Index" parameter calculated by ASTAR was taken to be the primary indicator of data quality, with higher values of the Index parameter indicating a higher confidence in the crystallographic orientation assigned by the software. An example of an area analyzed along with the orientation map is shown in Figure 2. Some boundaries between grains are slightly diffused due to the small grain size and grain overlap, but, in general, the individual grains are clearly visible. The distribution of the Index parameter for each pixel in the five areas analyzed is shown in Figure 3.

The frequency distributions of the thicker areas lie at higher values of the Index parameter compared to the thinner regions, which makes it appear that thicker samples generally produce higher quality orientation data than thinner samples. However, at some point grains overlap significantly, resulting in the degradation of the orientation data and more diffused zones between grains. The critical thickness of the overlap is likely grain size dependent, with that thickness decreasing with decreasing grain size. However, the samples appear to be of sufficient overall thicknesses to produce

Table 1. Samples and expected outcomes.

Sample Type	Sample ID	Description	Sample Status	Result to be Obtained
Unirradiated	Thickness 1	Effect of thickness on ASTAR (50mm)	To be FIB from unirradiated TRISO particle (fuel kernel removed) at CAES	Optimum sample thickness/baseline grain:grain boundary structure
	Thickness 2	Effect of thickness on ASTAR (100mm) to be FIB from unirradiated TRISO	To be FIB from unirradiated TRISO particle (fuel kernel removed) at CAES	Optimum sample thickness/baseline grain:grain boundary structure
	Thickness 3	Effect of thickness on ASTAR (150 mm) to be FIB from unirradiated TRISO	To be FIB from unirradiated TRISO particle (fuel kernel removed) at CAES	Optimum sample thickness/baseline grain:grain boundary structure
Irradiated	Compact 6-3-2			
	35-6B	Particle 35, 11% burnup, longitudinal orientation, low Ag release	Already prepared and analyzed for compositional information	Grain/grain boundary structure perpendicular to the SiC growth direction close to IPyC/SiC layer in low Ag release particle
	35-transverse 1	Particle 35, 11% burnup, transverse orientation (nearest kernel), low Ag release	Already prepared for compositional information*	Grain/grain boundary structure parallel to the SiC growth direction closest to IPyC/SiC interface in low Ag release particle
	35-transverse 2	Particle 35, 11% burnup, transverse orientation (middle SiC layer), low Ag release	Already prepared for compositional information*	Grain/grain boundary structure parallel to the SiC growth direction from the middle of the SiC layer in low Ag release particle
	35-transverse 3	Particle 35, 11% burnup, transverse orientation (furthest kernel), low Ag release	Already prepared for compositional information*	Grain/grain boundary structure parallel to the SiC growth direction closest to SiC/OPyC interface in low Ag release particle
	*Composition analysis is expected to be complete by the start of the RTE			

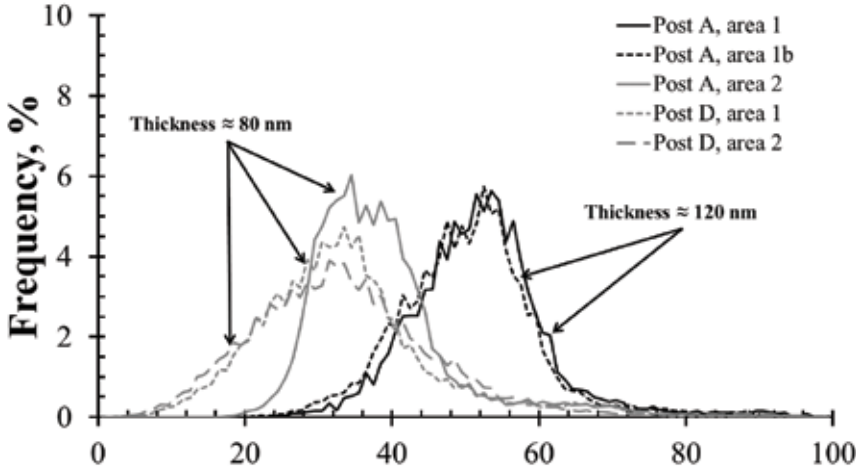


Figure 3. Frequency distribution of the Index parameter showing thicker samples yield higher quality orientation data.

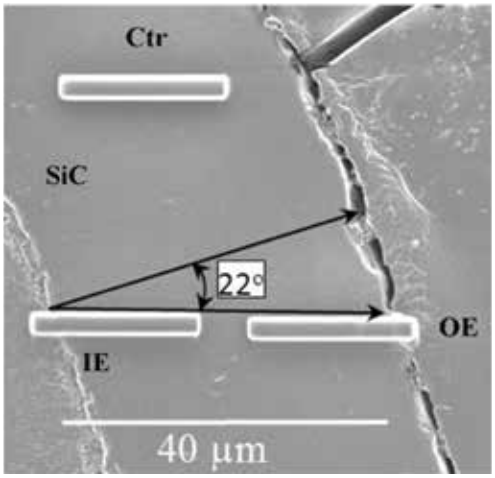


Figure 4. FIB lamellae positions on the SiC layer of sample AGRI-632-035.

high-quality orientation data. The effects of irradiation damage on the orientation data are not determined in this study, and may be an area of consideration for future research work. Irradiation damage can introduce significant numbers of defects into the SiC lattice, as well as generate residual stresses that may adversely affect the PED pattern and the resulting crystallographic orientation determination, (e.g. lowering the Index parameter).

Summary of Phase 2 results: Crystallographic characterization of grain boundaries and triple junctions in the SiC layer by PED utilizing ASTAR

ASTAR data has been collected in every area previously analyzed with STEM and energy dispersive x-ray spectroscopy (EDS) on FIB lamellae from coated particle AGR1-632-035, enabling examination of the crystallographic relationships on approximately 929 grain boundaries. Of these, only 179 boundaries and triple junctions contained fission products and transuranic elements. Analyses of these grain boundary characteristics are in the final stages of interpretation, and preliminary work already shows that using a high-angle annular dark-field scanning TEM (HAADF-STEM) is expedient to identify grain boundaries containing fission products. EDS was used to analyze, at least qualitatively, the composition of those fission products, and PED was used to evaluate the grain boundary parameters in the SiC layer of irradiated TRISO particles.

The final interpretation of these results and the integration of combined crystallographic and chemical analysis data still need to be completed to fully determine critical insight into the migration of fission products and transuranic elements through the SiC diffusion barrier layer. As an example of the level of information obtained, one specific area (Area B, Sample IE [4]) and an overview of the cumulative results from all areas are discussed in the following sections:

Area B, sample IE: A total of 446 grain boundaries were analyzed in the five areas (A to E) of the IE sample (Figure 4). However, only results specific to the analysis of the grain boundaries in Area B are shown. The characteristics of these 51 grain boundaries were determined and reported in a 2015 summary paper for the American Nuclear Society (ANS) [4].

The crystallographic parameters of the grains surrounding the grain boundary fission product were collected using ASTAR. PED patterns were collected in a square grid with a 10 nm step size and spot size of <1 nm. The grain orientation information was exported from ASTAR in the form of an American Standard Code for Information Interchange (ASCII) file containing the Euler angles in Bunge's notation as well as the corresponding x, y coordinates of each point in the scanned area. The orientation information was then analyzed using orientation imaging microscopy (OIM) to characterize the grain boundary misorientation. Brandon criteria were used to determine CSL-related grain boundary misorientations (Σ).

Most of the grain boundaries in this area are high-angle grain boundaries. The few low-angle grain boundaries present most likely arise from small

errors in orientation as determined by ASTAR. A plot of the misorientation angle distribution for grain boundaries in Area B is shown in Figure 5(a) while the pie-chart in Figure 5(b) provides the relative fractions of the various types of grain boundaries in the scanned area. As mentioned, the low-angle grain boundary fraction is very low. The misorientation distribution of the high-angle grain boundaries is centered around 35 degrees, with a large fraction observed at 60 degrees, which corresponds to the twin orientation.

The large fraction of CSL-related grain boundaries indicated in Figure 5 (b), consist mostly of twins and higher-order twins (e.g.Σ3 [69%],Σ9 [10%], and Σ27a and Σ27b [14%]). The remainder of the CSL-related boundaries consist of one Σ13a (3%) and one Σ35a (3%).

Only 17% of the grain boundaries in the analyzed area have fission products associated with them. A summary of these boundaries is provided in Table 2, where both the boundary crystallography and the associated fission products are detailed.

Only one CSL-related grain boundary exhibited the presence of a fission product that was exclusively palladium (Pd). The rest of the boundaries with fission products are random, high-angle grain boundaries containing only Ag, Pd+Ag, or Pd+uranium (U). However, the uniqueness of the CSL-related boundary containing fission products is questionable considering its high value (Σ35). Only one in 35 lattice sites between the two grains are coincident with a Σ35 CSL-related grain boundary. If this boundary lacks any special qualities relative to the other high-angle boundaries in Table 2, it can be concluded that fission products are only associated with random, high-angle grain boundaries.

Pd is the most prevalent fission product. It was found exclusively in five grain boundaries. Ag was found alone in only two grain boundaries, and with Pd in one. U was not found alone, and was associated with Pd in only one grain boundary. Only about 39% of the high-angle grain boundaries exhibited any presence of fission products at all. Thus, some other parameter would seem to strongly influence the precipitation of fission products, even in high-angle grain boundaries.

All grain boundaries analyzed in sample IE, Ctr and OE: The characteristics of 929 grain boundaries were determined in all the areas analyzed in samples IE, Ctr and OE (Figure 4). Full interpretation is in progress, with reports or papers being prepared. Figures 6 and 7 show some of the collective results on all grain boundaries analyzed.

It was found that 45.1% of all grain boundaries consist of high-angle boundaries that are not CSL boundaries, 49.2% are CSL boundaries, and only 5.7% are low-angle boundaries (Figure 6). Of all the boundaries analyzed, only 11.1% contain fission products. The grain boundary type distributions are presented in Figure 7. Only 6.0% of the boundaries containing fission products contain Ag and Pd, 14.3% contain only Ag, 17.9% contain Pd and U, and the remaining 61.9% contain only Pd. The 14.3% of grain boundaries containing only Ag are all high-angle grain boundaries while the grain boundaries containing

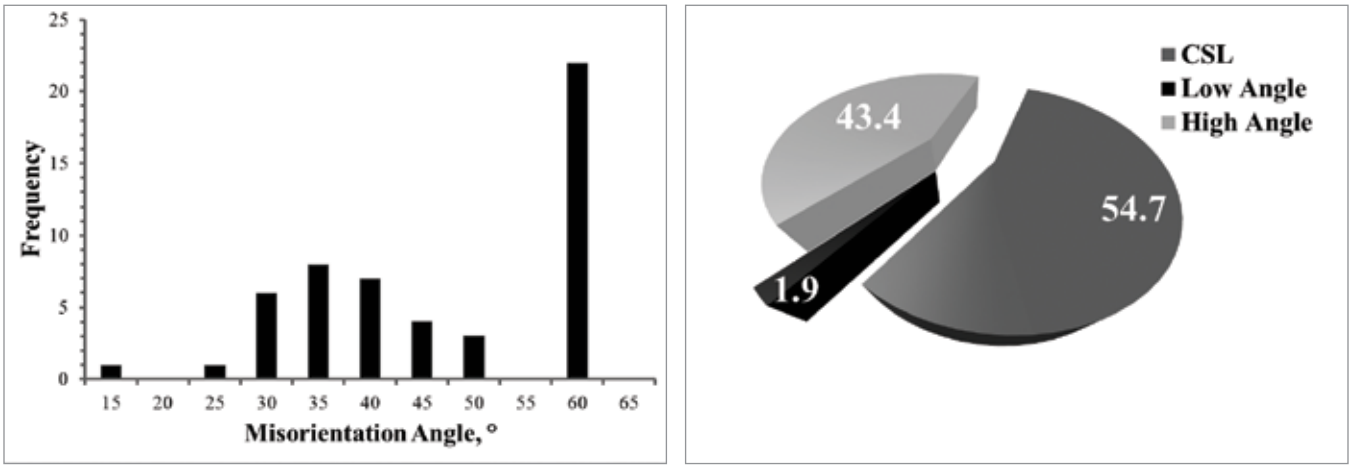


Figure 5. (a) Misorientation distribution of grain boundaries in Area B, sample IE, from particle AGR1-632-035. (b) A breakdown of grain boundary types [4].

Location	Misorientation	Axis			CSL, Σ	Elements		
	Angle, °	x	y	z		Pd	Ag	U
1	42.4	-7	12	-27	-	•		
2	29.4	7	-26	0	-	•		
3	49.5	1	-16	12	-	•		•
4	59.5	21	16	12	-	•		
5	27.0	-22	-17	11	-	•	•	
6	35.2	8	8	-5	-	•		
7	32.7	-25	11	10	35a	•		
8	39.7	-23	1	16	-		•	
9	31.2	5	2	-13	-		•	

Table 2. Details of boundaries with fission products.

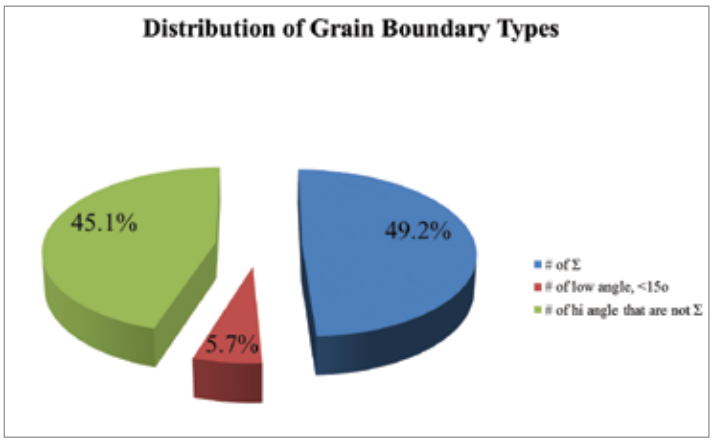


Figure 6. Summary of grain boundary types in the 929 grain boundaries analyzed.

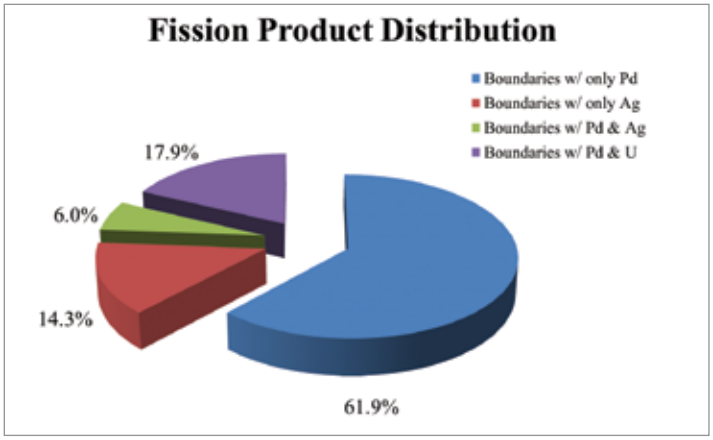


Figure 7. Summary of fission products contained in the 929 grain boundaries analyzed.

both Ag and Pd consist of four high-angle grain boundaries and one CSL grain boundary. This finding is now being pursued further as previous microstructural work does not show the presence of Ag on CSL boundaries. Understanding these phenomena could provide further clarity on the Pd-assisted Ag transport hypothesis proposed by Neethling et al. [5]

Future Activities

The goals of this research to continue are:

- Publish a journal paper on the ASTAR crystallographic results
- Interpret and integrate the results of the larger overarching project into a follow-up journal paper
- Continue work with the project collaborators to complete the larger project
- Submit a follow-up research proposal to expand the work on how irradiation damage effects the orientation data.

Staff Exchanges

The graduate intern and post-doctoral researcher were not directly involved in this particular study due to the timing of their appointments to INL: June and August 2014, respectively. However, this work formed the basis of the training that Dr Haiming Wen received on the application, collection, and interpretation of the ASTAR system, and he will continue his efforts on other samples as part of the AGR-1 experiment for 2015.

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[1.] I. J. van Rooyen, Y.Q. Wu, T. M. Lillo, T. L. Trowbridge, J. M. Madden, and D. Goran, 2013, "Advanced Electron Microscopic Techniques Applied to the Characterization of Irradiation Effects and Fission Product Identification of Irradiated TRISOCoated Particles from the AGR-1 Experiment," *Global 2013: Nuclear Energy at a Crossroads*, Salt Lake City, September 29–October 3, 2013.

[2.] I. J. van Rooyen, Y. Q. Wu, T. M. Lillo, J. Youngsman, J. H. Neethling, 2014, "Approach and Micro-Analysis Techniques Applied to Study Fission Product Transport Mechanisms in Neutron-Iirradiated SiC Layers," *XXIII International Materials Research Congress, Cancun, Mexico*, August 17-21, 2014.

[3.] I. J. van Rooyen, J. Youngsman, T. M. Lillo, Y. Q. Wu, D. Goran, M. E. Lee, W. E. Goosen, J. H. Neethling, T. L. Trowbridge, J. W. Madden, 2014, "Methods for Identification of Crystallographic Parameters of Irradiated SiC to Understand Fission Product Transport," *The 3rd Workshop on High Temperature Gas-Cooled Reactor SiC Material Properties*, Jeju Island, South Korea, Sept. 30–Oct. 1, 2014.

[4.] T. M. Lillo, I. J. van Rooyen, Y. Q. Wu, 2015, "Grain Boundary Character and Fission Product Precipitation in SiC," 2015 *American Nuclear Society Annual Meeting*, San Antonio, TX, June 7–11, 2015.

[5.] J. H. Neethling, J. H. O'Connell, and E. J. Olivier, 2012, "Palladium assisted silver transport in polycrystalline SiC," *Nuclear Engineering and Design*, Vol. 251, pp. 230-234.

Publications and Presentations

1. I. J. van Rooyen, Y. Q. Wu, T. M. Lillo, J. Youngsman, J. H. Neethling, 2014, "Approach and Micro-Analysis Techniques Applied to Study Fission Product Transport Mechanisms in Neutron- Iirradiated SiC Layers," *XXIII International Materials Research Congress, Cancun, Mexico, August 17-21, 2014.*
2. I. J. van Rooyen, J. Youngsman, T. M. Lillo, Y. Q. Wu, D. Goran, M. E. Lee, W. E. Goosen, J. H. Neethling, T. L. Trowbridge, J. W. Madden, 2014, "Methods for Identification of Crystallographic Parameters of Irradiated SiC to Understand Fission Product Transport," *The 3rd Workshop on High Temperature Gas-Cooled Reactor SiC Material Properties, Jeju Island, South Korea*, Sept. 30–Oct. 1, 2014.
3. T. M. Lillo, I. J. van Rooyen, Y. Q. Wu, 2015, "Grain Boundary Character and Fission Product Precipitation in SiC," 2015 *American Nuclear Society Annual Meeting, San Antonio, TX, June 7–11, 2015.*

"With its ability to quickly determine grain boundary parameters at the nanometer scale, we expect ASTAR will be a key tool in facilitating a full understanding of the fission product transport mechanism in the SiC layer of TRISO nuclear fuel, and ultimately allow the development of strategies to minimize or mitigate the release of fission products"

— Dr. Tom Lillo, Research Scientist/ Engineer, Idaho National Laboratory

Distributed Partnership at a Glance

ATR NSUF and Partners	Facilities and Capabilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Idaho National Laboratory	Materials and Fuels Complex, Electron Microscopy Laboratory
Collaborators	
Idaho National Laboratory	Isabella van Rooyen (principal investigator), Tomas Lillo (co-principal investigator), James Madden (collaborator), Haiming Wen (post-doctoral researcher, non U.S.)
Boise State University	Yaqiao Wu (co-principal investigator), John Youngsman (co-principal investigator)
Idaho State University	Connie Hill (graduate intern)

Microstructure Evolution in Ion-Irradiated Uranium Dioxide

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The first demonstration of micro-focused x-ray absorption fine-structure measurements on FIB UO_2 lamellae was successful, clearing the way for analysis of highly radioactive neutron-irradiated samples at synchrotron facilities.

Understanding irradiation damage evolution in uranium dioxide (UO_2) is crucial to protecting the world's fleet of current and future nuclear power plants. Since all of the phenomena caused by radiation damage originate at point defects, understanding the effects of irradiation at the atomic scale is crucial. However, because the irradiation defects are aperiodic, standard approaches, such as transmission electron microscopy (TEM) and x-ray diffraction (XRD) are ineffective, necessitating the use of techniques that are sensitive to short-range order. X-ray absorption fine-structure spectroscopy (XAFS) measures the population-weighted local structures and chemical speciation of the examined elements, making it perhaps the most incisive method for determining the local-range order in irradiated materials.

TEM measurements are crucial to relating the short-range changes observed using extended X-ray absorption fine-structure spectroscopy (EXAFS) to the complicated, long-range microstructures created through irradiation. Understanding these defects at various length scales is necessary for accurately predicting fuel degradation under reactor conditions.

Project Description

The goal of this experiment is to study irradiation-induced damage structure evolution in UO_2 on multiple-length scales. The main objective is to combine spatially resolved, synchrotron-based X-ray absorption spectroscopy (XAS) with TEM observations of ion-irradiated depleted UO_2.

This study uses XAFS observations combined with microscopy techniques performed at CAES to examine lamellae having spatially varied irradiation damage. Ion irradiations that produced spatially varied microstructures were performed at ~150°C on depleted UO_2 samples at the University of Wisconsin – Madison's Ion Beam Laboratory. The damage profiles in the helium (He)-implanted samples were about 10 μm, and were as flat as possible for the first 15 μm.

With the completion of these experiments, the targeted research has increased our understanding of fuel degradation and added to the knowledge base of the nation's nuclear infrastructure. The benefits of knowing how the local structure of ion-irradiated UO_2 evolves extend beyond nuclear fuel operations into long-term storage. Being able to predict the stability of reactor-irradiated UO_2 is crucial to determining what chemical reactions will occur while a fuel is being stored.

Accomplishments

All the milestones described in the ATR-NSUF rapid-turn-around proposal have been met. A technique to perform μ-EXAFS measurements has been developed at the Stanford Synchrotron Radiation Lab (SSRL), and a focused ion beam (FIB) lamella consisting of the damage layer created by injecting He ions into bulk UO_2 samples was extracted at CAES. Figure 1 compares the extracted FIB lamellae (Figure 1.1) to the overlay of the ion-damage profile created using Stopping Range of Ions in Matter (Figure 1.2).

Five such lamellae, two each from the two irradiated samples and one from the pristine sample, were mounted on an FIB grid. The grid was then inserted into a μ-EXAFS holder specially designed for the SSRL. Two of these samples are shown in Figure 1.2. They were measured at a 45-degree angle from the incident beam in fluorescence geometry. Further, bulk EXAFS analysis was performed on krypton-implanted samples at an angle of 10 degrees from the incident beam, which interrogated the irradiated region to a depth of 1.

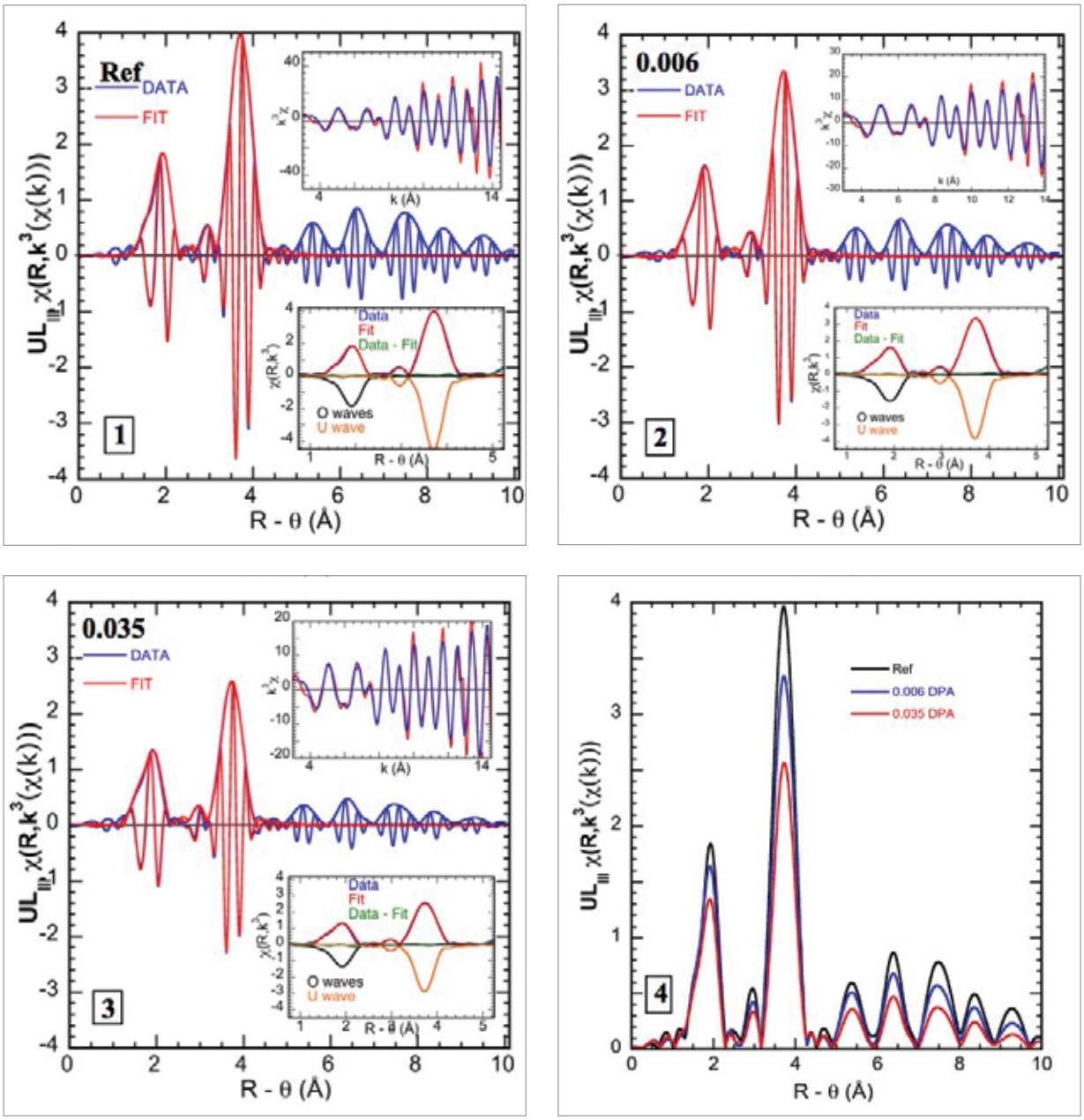


Figure 1. (1) The extracted FIB lamellae, (2) the overlay of the ion-damage profile created using Stopping Range of Ions in Matter.

"In this experiment, we're learning that identifying the differences between oxygen doping and irradiation isn't so simple. There may even be differences between the proton- and He-irradiated materials, and we want to make sure we get it right."

— Steve Conradson, Beamline Responsible, Synchrotron SOLEIL (current position)

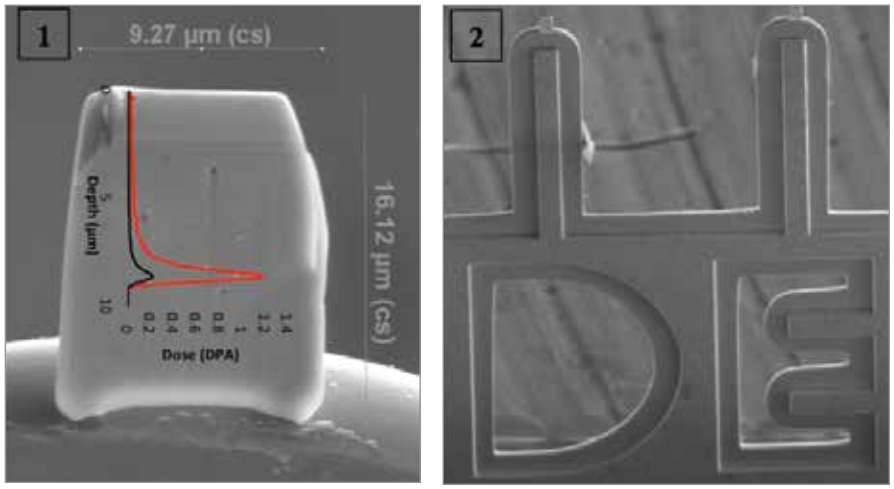


Figure 2. $k^3\chi(R)$ EXAFS of Ref UO_2, UO_2 irradiated to 0.006 dpa with He^{2+} ions and UO_2 irradiated to 0.035 dpa with He^{2+} ions. Plots 1-3: the modulus of the real part of the transform of both data and fit. Insets, (top) $k^3\chi$ spectra overlaid with curve-fit; (bottom) moduli of the data, fit, difference between data and fit, and the individual contributions to the fit (inverted for clarity). Plot 4: modulus of the Fourier transforms of the k^3-weighted EXAFS spectra of indicated samples. Transforms were performed over a range of 2.70 to 14.75 $Å^{-1}$.

1 µm. He-implanted samples irradiated to 0.006–0.035 displacements per atom (dpa) and proton-irradiated samples (0.01–0.5 dpa) were also successfully studied using EXAFS at SSRL. This was a first-of-its-kind demonstration of µ-EXAFS measurements on FIB lamellae and brought together high-energy x-ray studies from synchrotron sources and the microscopic analysis techniques used in material characterization.

EXAFS analysis has shown that irradiation disrupts local structure by giving rise to multisite oxygen distribution at approximately 1.9 angstroms (Å) from the absorbing atom, which is 0.2 Å farther than in documented UO_{2+x}. This is similar to oxygen interstitials created in UO_{2+x}, however, there is no observed oxidation of the material. The multisite distribution results from uranyl-type bonds that are ~1.8 Å in length and extremely stable due to their oblate geometry, which causes them to distort the original local structure.

The EXAFS data from the irradiated UO_2 shows a consistent decrease in amplitude in the crystallographic shells as irradiation doses increase, longer bond distances between near neighbors, and the appearance, and increase, of a shoulder on the low R side (at 1.9 Å) of the first crystallographic U-O shell. Random disorder in the material would result in a significant loss of amplitude and additional loss of overall structure, which is not seen in this set of samples. Due to the consistent amplitude reduction, as shown in Figure 2, it can be theorized that the defects are not random but instead cluster in such a way that the overall lattice retains its UO_2 structure even at higher irradiation doses. This is consistent with several studies that indicate fluorite ceramics in a reactor maintain their lattice structure even at high irradiation doses.

Future Activities

The most important future goal for the project is the full characterization of μ-EXAFS data from the FIB UO_2 samples. Processing this data has its own challenges due to the lower signal strength microscopic samples emit. Analysis is underway and is to be completed by March 2015.

A subsequent ATR NSUF rapid turnaround proposal to continue the research was submitted and accepted. Samples of depleted UO_2 were prepared using polishing techniques at CAES and were sent to the University of Wisconsin – Madison for proton implantation at its ion beam facility, which initiated radiation damage in the 0.2–0.3 dpa range. The irradiated samples were mounted on specially designed sample holders for local structure and speciation analysis at the Advanced Photon Source. These samples will be used to extract FIB lamellae from the irradiation damage layer, and their microstructure evolution will be studied using the TEM at the Microscopy and Characterization Suite at CAES.

The goal of this experiment is to bridge the gap between existing information on lattice structure and microstructure evolution under proton irradiation in UO_2. Preparation of samples for irradiation at ATR is currently under way. These irradiations will provide a detailed understanding of the evolution of the material structure in UO_2 under reactor irradiation.

Publications and Presentations

1. M. Gupta, 2014 "Damage Structure Evolution in Ion Irradiated UO_2," *TMS conference, San Diego, CA, February 16–20, 2014.*
2. M. Gupta, 2014, "Atomic Structure Evolution in Ion Irradiated UO_2," presentation for Dr. Franklin (Lynn) Orr, Undersecretary for Science and Energy, May 19th 2014.
3. M. Gupta, 2014, "Defects in Ion Irradiated UO_2 on Multiple Length Scales: A TEM and XAFS Study," presentation for Dr. Patricia Dehmer, Deputy Director for Science Programs in the Office of Science, Center for Advanced Energy Studies, August 12, 2014.
4. M. Gupta, 2014, "Defects in Ion Irradiated UO_2 on Multiple Length Scales: A TEM and XAFS Study", presentation for Dr. Ernest Moniz, US Energy Secretary, Center for Advanced Energy Studies, Idaho Falls, ID, August 19, 2014.
5. M. Gupta, S. Conradson, J. Pakarinen, and T. Allen, 2015, "Identificatio3n of Collective Effects in He2+ irradiated UO_2 via Extended X-ray Absorption Fine Structure Spectroscopy," submitted to the *Journal of Inorganic Chemistry*, March 2015.

Distributed Partnership at a Glance

ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	Advanced Test Reactor
Center for Advanced Energy Studies	Microscopy and Characterization Suite
University of Wisconsin – Madison	PIE facilities
Collaborators	
University of Wisconsin – Madison	Mahima Gupta (principal investigator), Janne Parkarinen (collaborator)
Idaho National Laboratory	Todd Allen (co-principal investigator), Jian Gan (collaborator)
Los Alamos National Laboratory	Steve Conradson (collaborator)

Radiation Induced Segregation in Nickel-Chromium Alloys

Janne Pakarinen – University of Wisconsin,Madison – jpakarin@sckcen.be

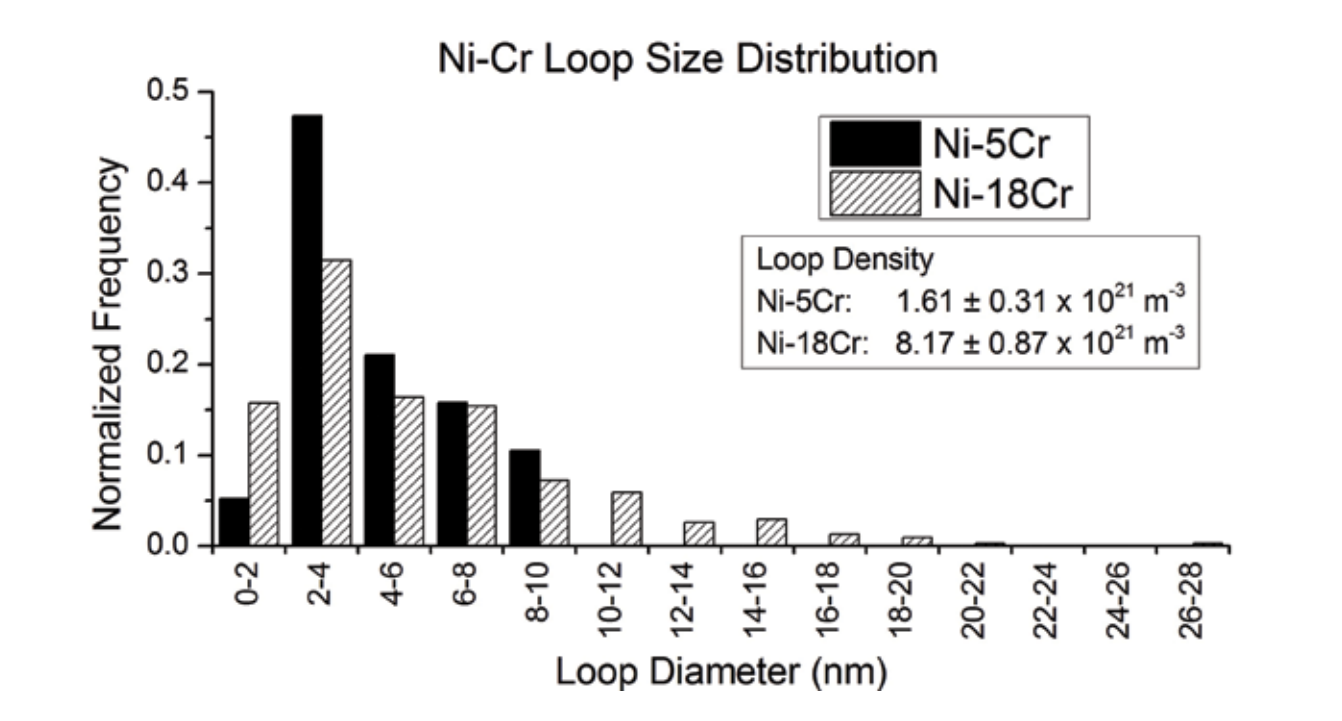


Figure 1. Dislocation loop density and size distribution in 500°C proton-irradiated Ni-Cr.

Understanding the radiation responses of Ni-based alloys is vital to developing reactor core components for next-generation molten salt technologies.

Nickel (Ni)-based alloys are employed in current light water reactors (LWR), though typically not as in-core components. They are also candidate materials for molten salt reactor applications due to their excellent corrosion resistance in fluoride salt systems. As such, understanding how radiation damage influences their microstructures and performance in these systems is crucial to the development of safe and reliable molten salt reactor technologies.

Project Description

This project will investigate the microstructural effects of ion irradiation in Ni-based alloys, with a focus on the manifestation of voids, dislocation loops, and radiation-induced segregation (RIS). The formation of these features in binary Ni-Chromium (Cr) alloys is being studied as a function of three experimental variables:

- Cr composition (5wt% Cr, 18wt% Cr, and 33wt% Cr)
- Irradiation temperature (400°C and 500°C)
- Irradiating species (proton vs. Ni-ion irradiation)

These variables are characterized using analytical transmission electron microscopy (TEM) techniques to show the expected radiation responses of these alloys in a nuclear reactor system, as well as to provide experimental data to be used as benchmarks in the development of predictive models for the formation of these features.

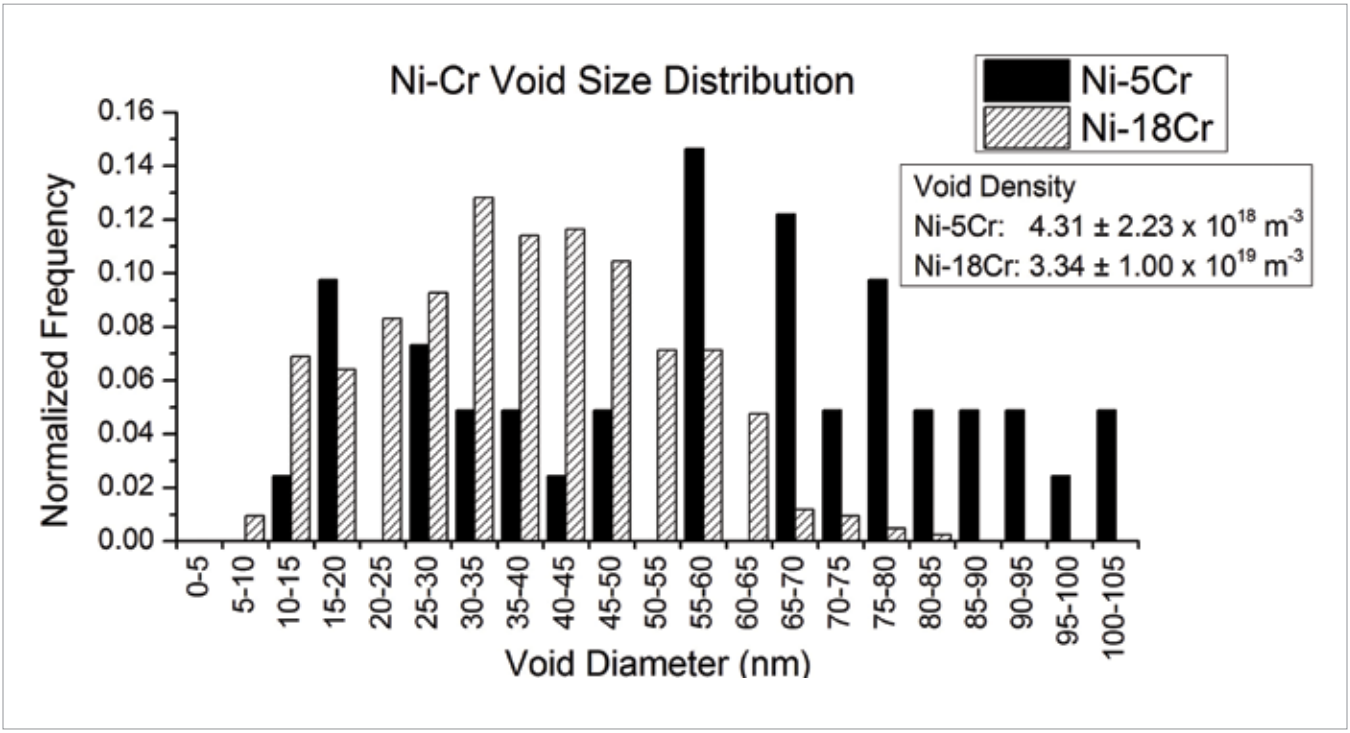


Figure 2. Void density and size distribution in 500°C proton-irradiated Ni-Cr.

Accomplishments

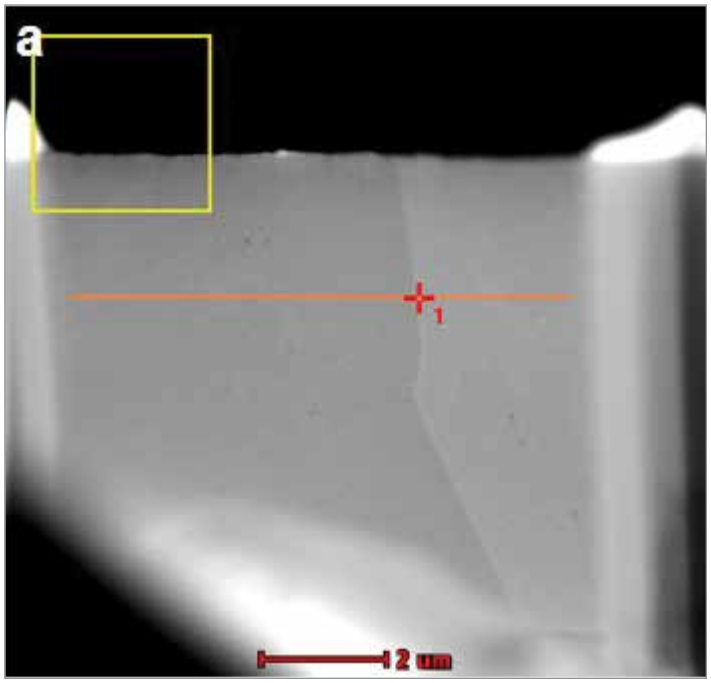
Prior to 2014, both Ni-5Cr and Ni-18Cr samples were irradiated with both protons and Ni-ions at 500°C, in the University of Wisconsin (UW) Tandem Accelerator Ion Beam. In early 2014, TEM sample preparation and preliminary analysis of the 500°C proton-irradiated materials was performed in the CAES Microscopy and Characterization Suite (MACS) using the Quanta 3D Focused Ion Beam (FIB) and Tecnai TF-30 TEM. Additional TEM characterization was also performed at the University of Wisconsin Materials Science Center (MSC) on the Tecnai TF-30 TEM and the FEI TITAN TEM.

These TEM observations characterized and compared the density and size distributions of voids and dislocation loops in the samples (Figures 1 and 2).

Abnormal RIS behavior was also observed at 500°C, and was thought to be due to grain boundary migration effects (Figure 3). This prompted the 400°C proton irradiation, in which the Ni-33Cr model alloy was also included. In addition to providing another composition with which to compare the formation of these microstructural features, Ni-33Cr was added to investigate whether or not irradiation could induce a low-temperature stable ordered phase, which is present in this material at that composition. The second proton irradiation was also performed at the UW Tandem Accelerator Ion Beam, and the sample preparation and preliminary analysis were again performed at CAES-MACS.

"The FIB and TEM capabilities at CAES are world class, and provide means for productive sample fabrication and analysis. The rapid turn-around system fit the needs of this project perfectly."

— Dr. Janne Pakarinen, Visiting Scientist, University of Wisconsin, Madison



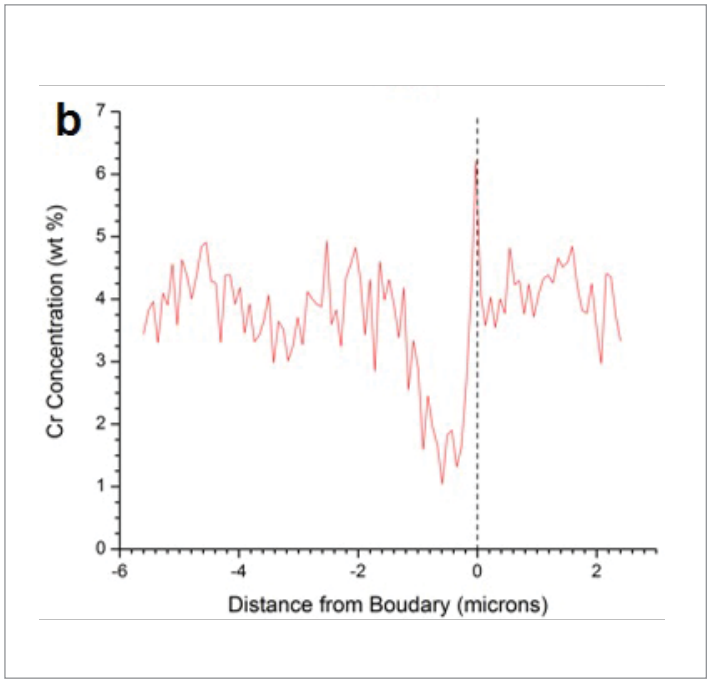


Figure 3. Typical RIS profile observed in 500°C proton-irradiated materials. A large Cr-depleted region is observed that is also denuded in voids and dislocations.

Simultaneously during 2014, Ni-ion irradiation and post-irradiation examination (PIE) were being performed by Christopher Barr at Drexel University. These samples were irradiated with 20 MeV Ni-ions at the Sandia National Laboratory Ion Beam Laboratory. Sample preparation and additional analysis was performed at the CAES-MACS facility.

Future Activities

TEM characterization of the 400°C proton-irradiated materials is ongoing. Additionally, characterization of dislocation loop density in the 500°C Ni-irradiated materials is still in progress. Completion of TEM analysis on these two alloy systems will allow us to compare the effects of composition, temperature, and irradiating species on the microstructure resulting from irradiation.

Three papers are currently planned for this project in 2015. The first will compare and contrast the resulting microstructures in the binary Ni-Cr system as functions of composition, irradiating temperature and irradiating species. The second will discuss the unusual RIS profiles observed in the 500°C irradiation. The third will discuss the viability of inducing low-temperature stable ordered phases using short ion irradiations as opposed to long-term thermal annealing experiments.

Publications and Presentations

1. S. Briggs, J. Pakarinen, L. Barnard, D. D. Morgan, T. R. Allen, and K. Sridharan, 2014, "Radiation-Induced Segregation and Other Radiation-Induced Effects in Ni–Cr Alloys, *Materials Science & Technology 2014, Pittsburgh, PA, Oct. 12–16, 2014.*
2. S. Briggs, J. Pakarinen, L. Barnard, D. D. Morgan, I. Szlufarska, T. R. Allen, and K Sridharan, 2014, "Study of Radiation-Induced Segreation Using Nickel-Chromium Binary Alloys," *TMS conference, San Diego, CA, February 16–20, 2014.*

Distributed Partnership at a Glance

ATR NSUF and Partners	Facilities and Capabilities
Center for Advanced Energy Studies	Microscopy and Characterization Suite
University of Wisconsin, Madison	Tandem Accelerator Ion Beam
Sandia National Laboratory	Ion Beam Laboratory
Drexel University	Central Research Facilities
Collaborators	
University of Wisconsin - Madison	Janne Pakarinen (principal investigator), Samuel Briggs (co-principal investigator), Leland Barnard (co-principal investigator), Kumar Sridharan (co-principal investigator), Dane Morgan (co-principal investigator), Mitra Taheri (co-principal investigator), Christopher Barr (co-principal investigator)

Ion Irradiation of Nuclear Grade NBG-18 and Highly Ordered Pyrolytic Graphites

K. Linga Murty – North Carolina State University – murty@ncsu.edu

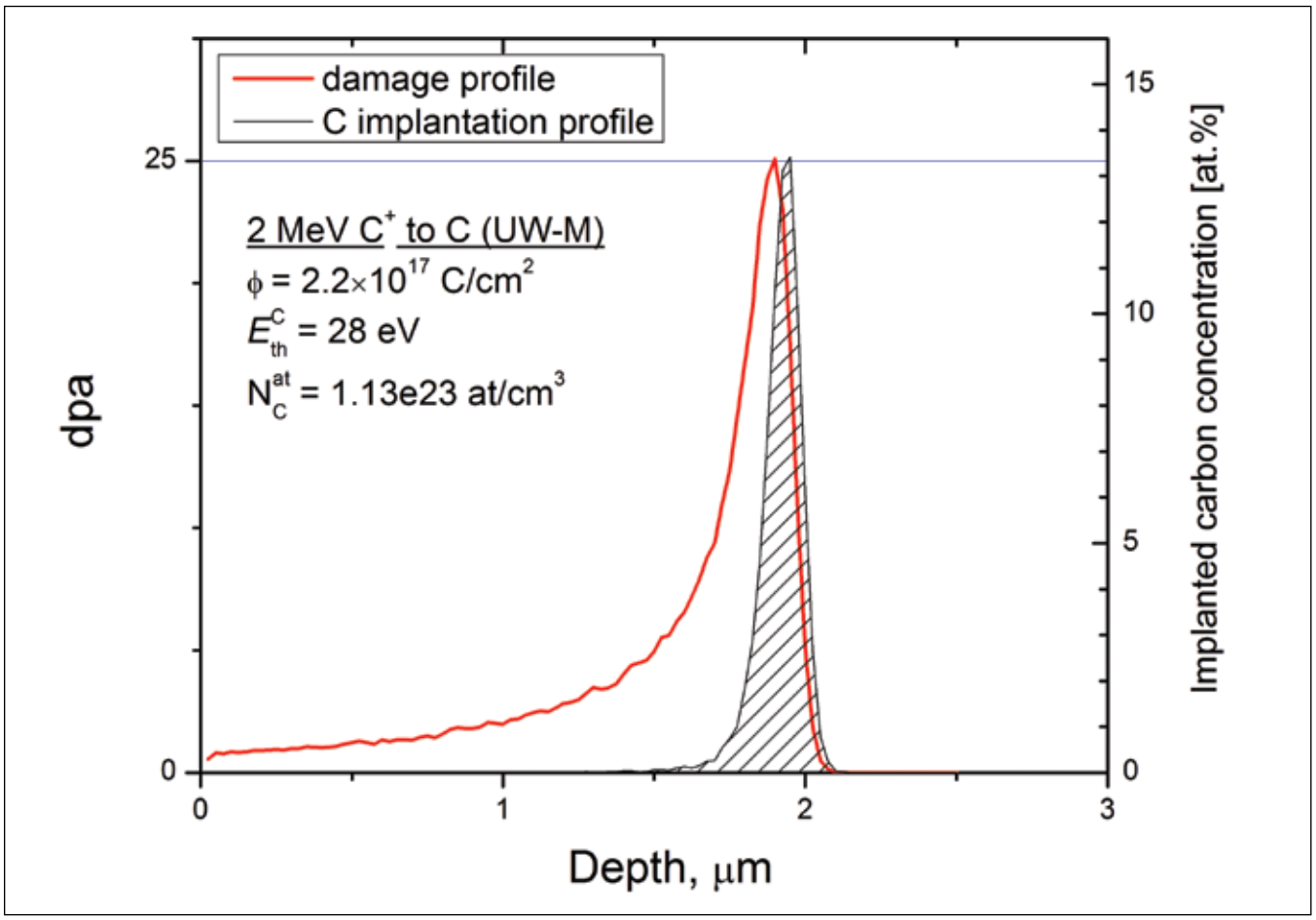


Figure 1. MeV carbon-implantation and damage profiles calculated using SRIM 2012.03 software.

By developing mechanistic, thermo-mechanical models for graphite that can be applied to next-generation, high-temperature reactors, our studies address the industry's general lack of clarity on the damage mechanisms of graphite. To this end, the project was conceived to investigate the radiation damage on highly oriented pyrolytic graphite (HOPG) and NBG-18 through ion irradiation at the University of Wisconsin – Madison's (UW) Tandem Accelerator Ion Beam.

Project Description

The general intent of the ion-irradiation experiments at UW was to characterize the early-to-late-stage damage mechanisms in graphite under irradiation. This follows the low-dose, neutron-irradiation studies conducted in the PULSTAR reactor at North Carolina State University (NCSU) and the high-dose studies carried out at the Oak Ridge National Laboratory (ORNL).

This project seeks to clarify our understanding of the damage mechanisms of graphite.

Table 1. Ion irradiation cases.

Temperature (K)	dpa	Flux (ions/cm^2.s)
300	1	1.3×10^{13}
600	1	1.1×10^{13}
900	1	1.3×10^{13}
600	25	1.4×10^{13}
300	25	1.2×10^{13}

Table 2. Raman analysis for ion-irradiated NBG-18 samples.

Ion Irradiation Condition	ID/IG	ID/ ID'
300 K/1dpa	0.87	2.54
600 K/1 dpa	0.58	4.47
900 K/1 dpa	0.84	2.18
600 K/25 dpa	1.38	1.79
900 K/25 dpa	0.80	2.43

Table 3. Raman analysis for ion-irradiated HOPG samples.

Ion Irradiation Condition	ID/IG
300 K/1dpa	0.02
600 K/1 dpa	0.01
900 K/1 dpa	0.06
600 K/25 dpa	0.58
900 K/25 dpa	0.25

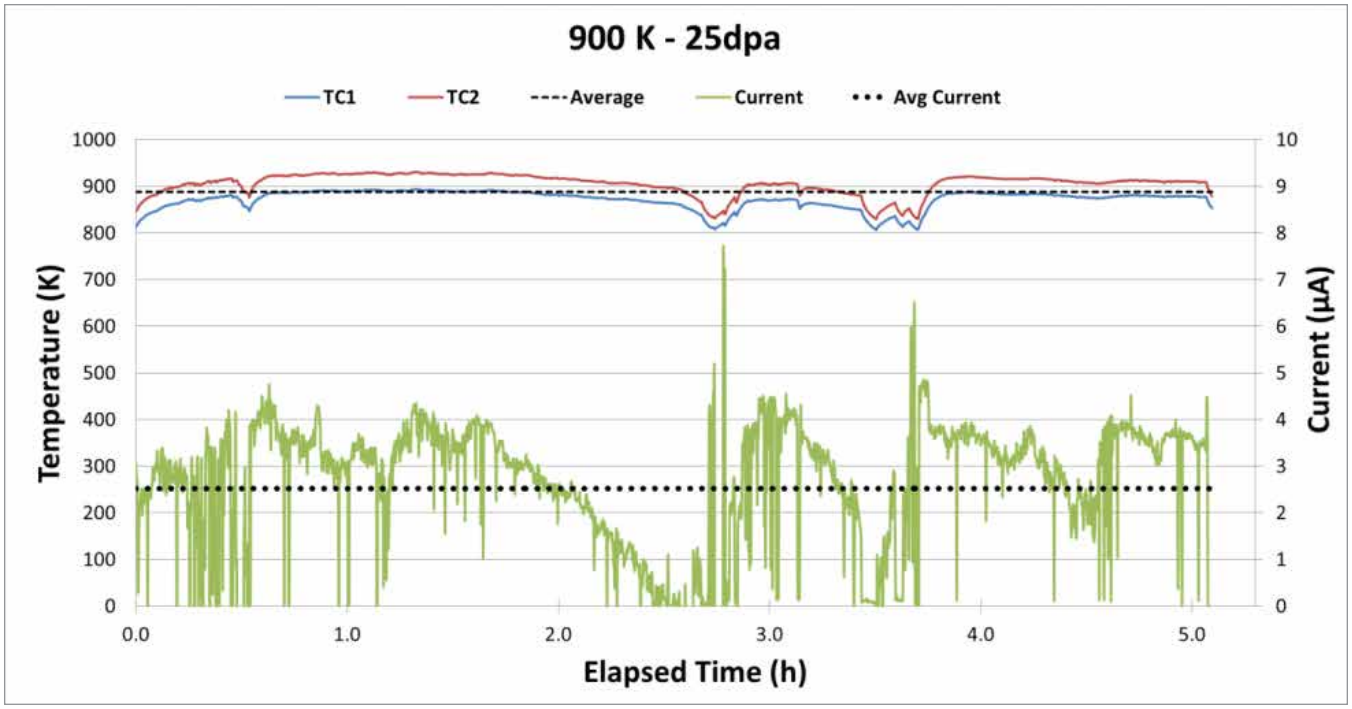


Figure 2. Temperature and beam-current profiles during irradiation at 900 K, 25 dpa, and 2.0 MeV C^+.

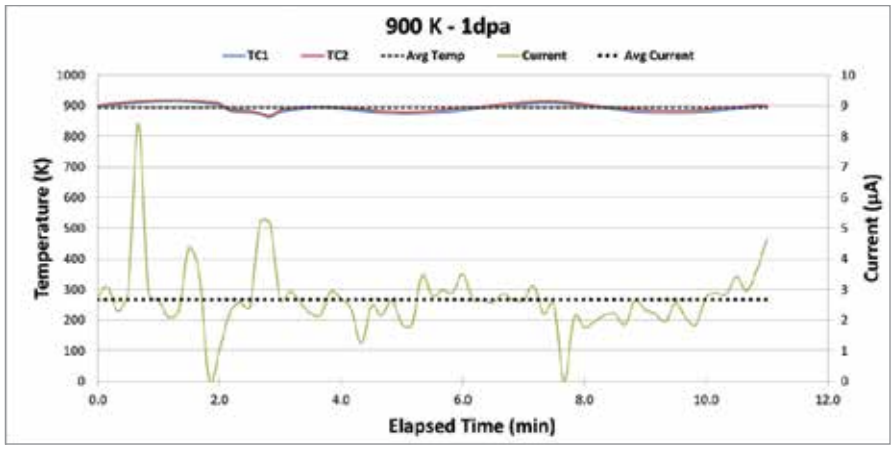


Figure 3. Temperature and beam-current profiles during irradiation at 900 K, 1 dpa, and 2.0 MeV C^+.

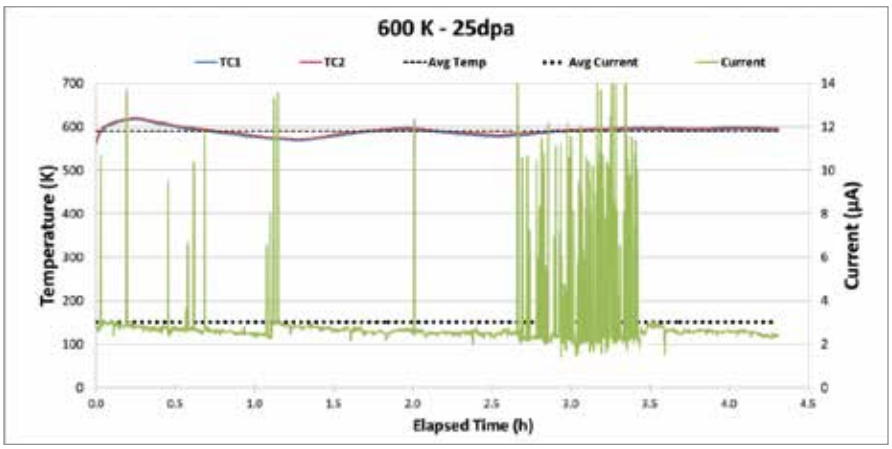


Figure 4. Temperature and beam-current profiles during irradiation at 600 K, 25 dpa, and 2.0 MeV C^+.

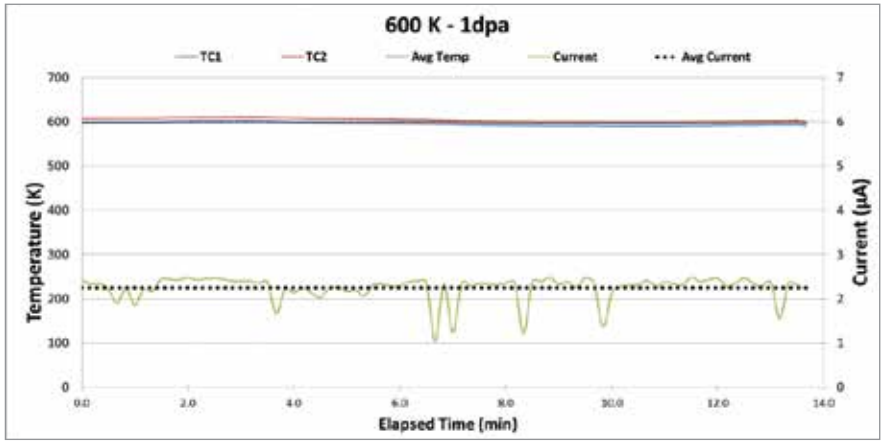


Figure 5. Temperature and beam-current profiles during irradiation at 600 K, 1 dpa, and 2.0 MeV C^+.

"The team at the University of Wisconsin has been methodical, insightful, and extremely professional throughout this project."

— Jacob Eapen, Assistant Professor, Associate Professor, Nuclear Engineering, North Carolina State University

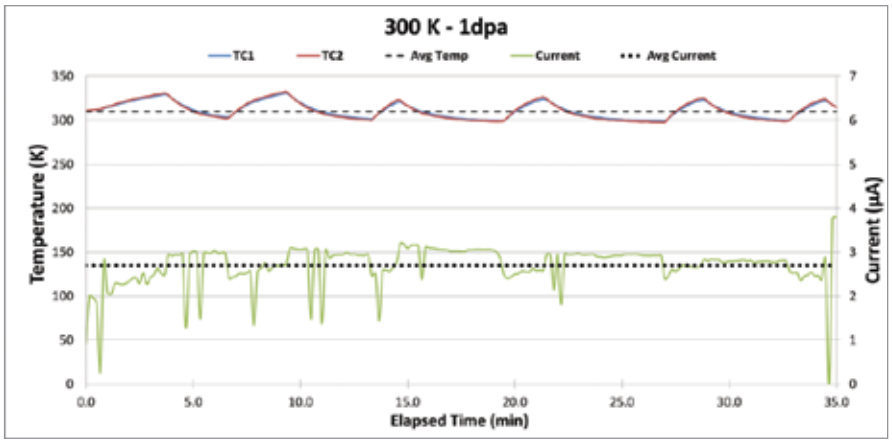


Figure 6. Temperature and beam-current profiles during irradiation at 300 K, 1 dpa, and 2.0 MeV C^+.

Accomplishments

Ion-irradiation experiments were conducted on the HOPG and NBG-18 samples at displacements per atom (dpas) of 1 to 25, and temperatures ranging from 300 to 900 K. Damage was initiated with C^+ ions at 2-MeV energy. The fluences at 25 dpa and 1 dpa are 2.2×10^{17} ions/cm^2 and 8.8×10^{15}, respectively. The test durations for the 25-dpa and 1-dpa samples were 4.5 hours and 12 minutes, respectively. The average displacement energy was 28 eV. Table 1 shows details of the irradiations at different temperatures and dpas.

Figure 1 shows the 2-MeV carbon-implantation and damage profiles calculated using Stopping and Range of Ions in Matter (SRIM) 2012.03 software and assuming a displacement-threshold energy of 28 eV and a mass density of 2.253 g/cm^3. As indicated in the plot, 25 dpa (peak) corresponds to a fluence of 2.2×10^{17} C/cm^2 (13 at.% of carbon in the peak). Temperature and beam-current profiles are shown in Figures 2 through 6. Tables 2 and 3 show the ratio of the peaks for each sample.

Publications and Presentations

1. J. Eapen, R. Krishna, T. D. Burchell, K. and L. Murty, 2014, "Early Damage Mechanisms in Nuclear Grade Graphite Under Irradiation", Materials Research Letters Vol. 2, pp. 43–50.

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
University of Wisconsin - Madison	Tandem Accelerator Ion Beam
Collaborators	
University of Wisconsin – Madison	K. Linga Murty (principal investigator), Jacob Eapen (co-principal investigator), Ram Krishna (co-principal investigator)

Transmission Electron Microscopy Study of the Microstructure Evolution in Kr-Irradiated UO_2

Lingfeng He – Idaho National Laboratory – lingfeng.he@inl.gov

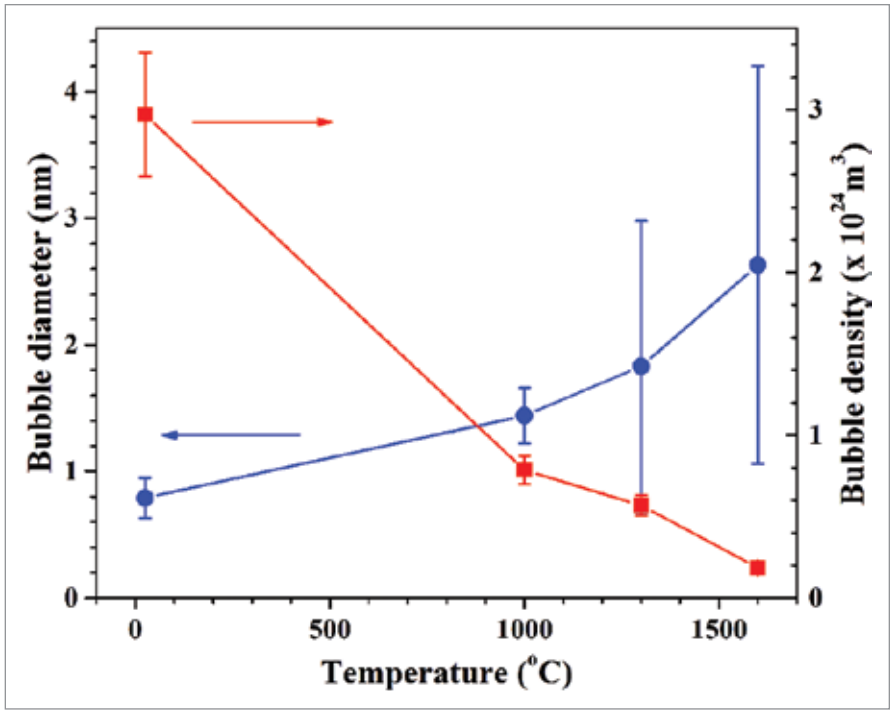


Figure 1. Intragranular bubble size and density as a function of the annealing temperature.

This experiment will improve our understanding of microstructure evolution in UO_2 under irradiation.

In-situ transmission electron microscope (TEM) observation of uranium dioxide (UO_2) irradiated with krypton (Kr) ions at the Intermediate Voltage Electron Microscope (IVEM)-Tandem facility at Argonne National Laboratory has shown the evolution of the basic radiation defects of dislocation loops and bubble formation. However, the TEM foil samples used for in situ observation were too thin and therefore unsuitable for thermal transport measurement.

Project Description

In this rapid turnaround work, we are investigating defect production and dislocation loop and bubble evolution in UO_2 under ex-situ, 1.8 MeV Kr irradiation. We are also providing theoretical modeling support that connects the material's microstructure to its thermal transport properties for a parallel project being conducted at INL.

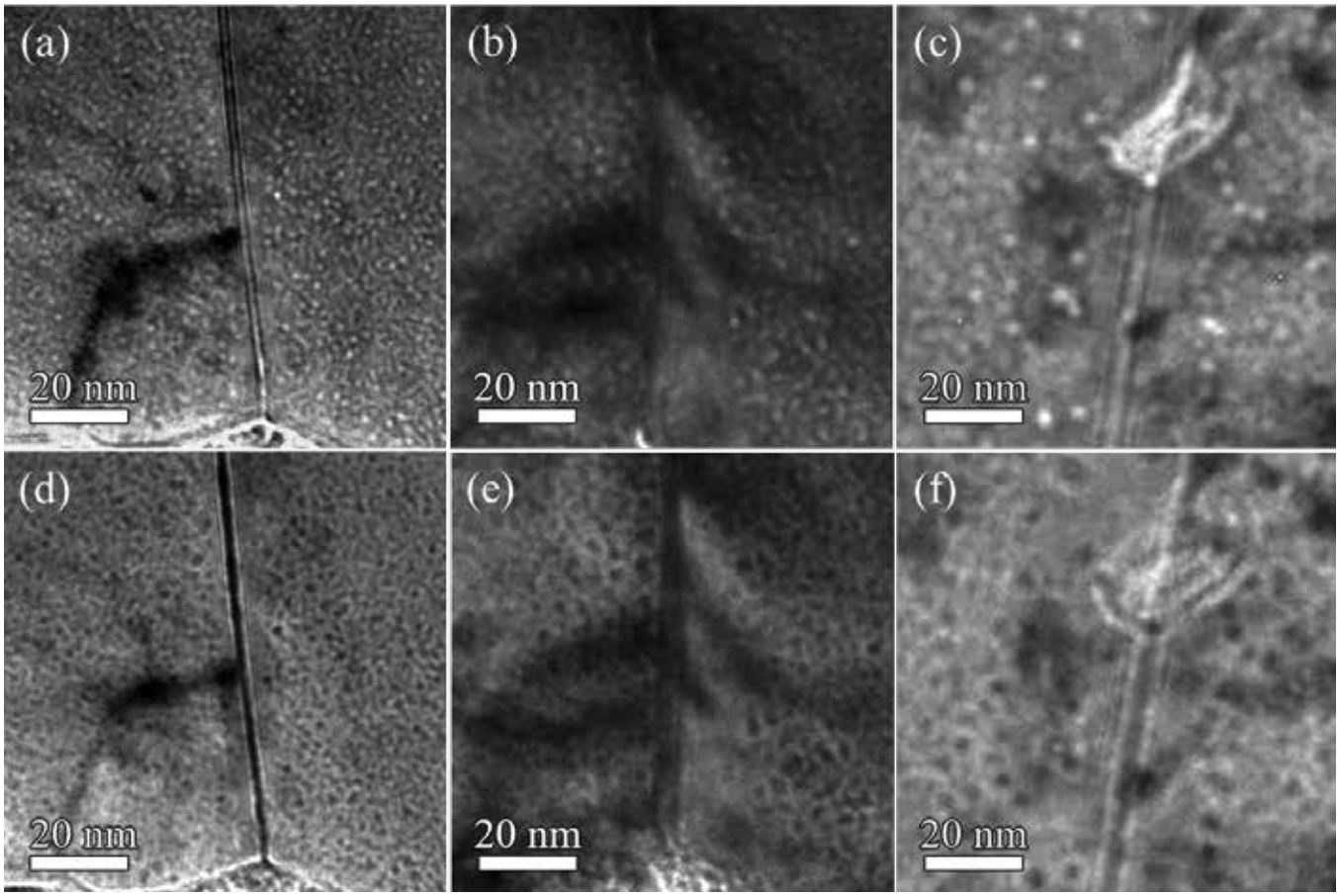


Figure 2. Cross-section TEM images showing bubbles near grain boundaries in Kr-irradiated, polycrystalline UO_2 after annealing at 1000°C (a and d), 1300°C (b and e), and 1600°C (c and f) for one hour. Figures a-c are under-focus images and Figures d-f are over-focus images.

Accomplishments

In this work, we investigated bubble and defect evolution in Kr-irradiated and high-temperature-annealed UO_2. The results showed bubble formation at room-temperature irradiation. Bubbles grew gradually at the annealing temperature, while their densities decreased gradually (Figure 1).

A bubble denuded zone in the vicinity of the grain boundaries was found (Figure 2), which indicates that the Kr diffused towards the grain boundaries during high-temperature annealing. Grain boundaries contained more vacancies than the bulk regions, and they proved to be good reservoirs for inert Kr gas, resulting in the rapid growth of intergranular bubbles at grain boundaries (Figure 3).

In addition, high-angle grain boundaries have more room for inert gas compared to low-angle grain boundaries, allowing the formation of larger

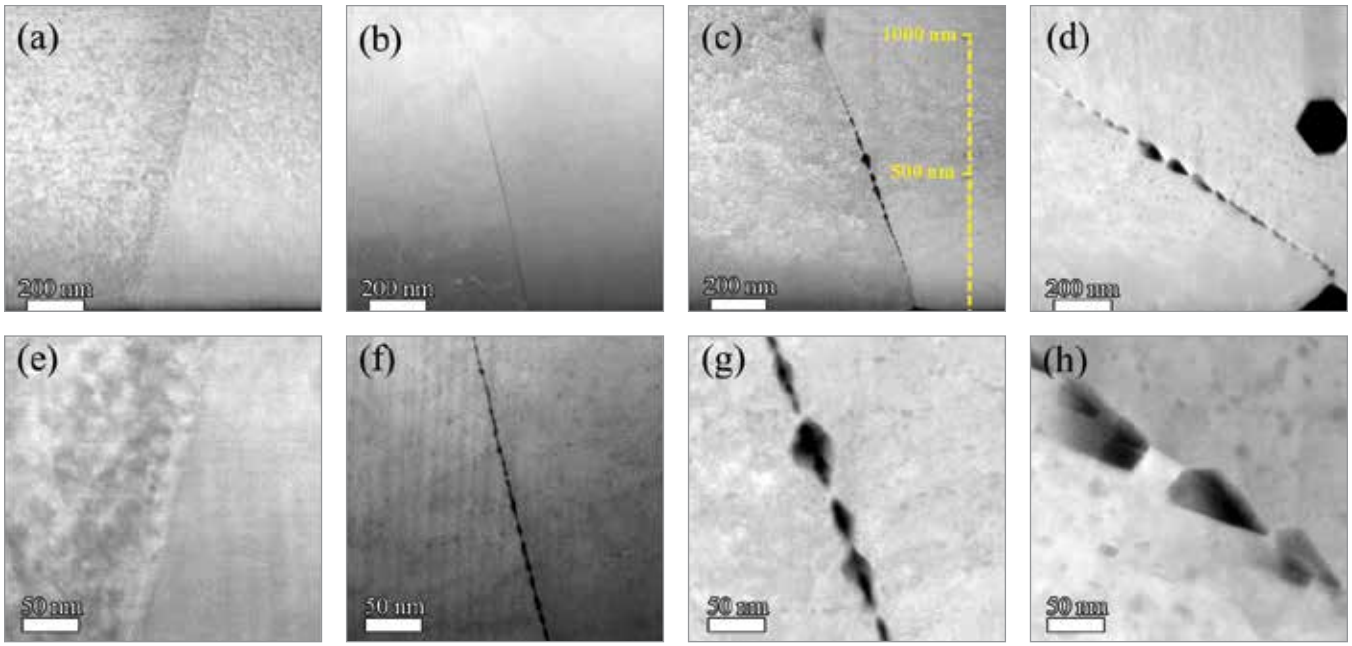


Figure 3. Cross-section scanning transmission electron microscope (STEM) images showing the evolution of intergranular bubbles during post-irradiation annealing at (a and e) 25°C, (b and f) 1000°C, (c and g) 1300°C, and (d and h) 1600°C. Figures (a-d) are low-magnification images, and figures (e-h) are high-magnification images. The dashed line in (c) indicates the bubbles' distance from the sample's surface.

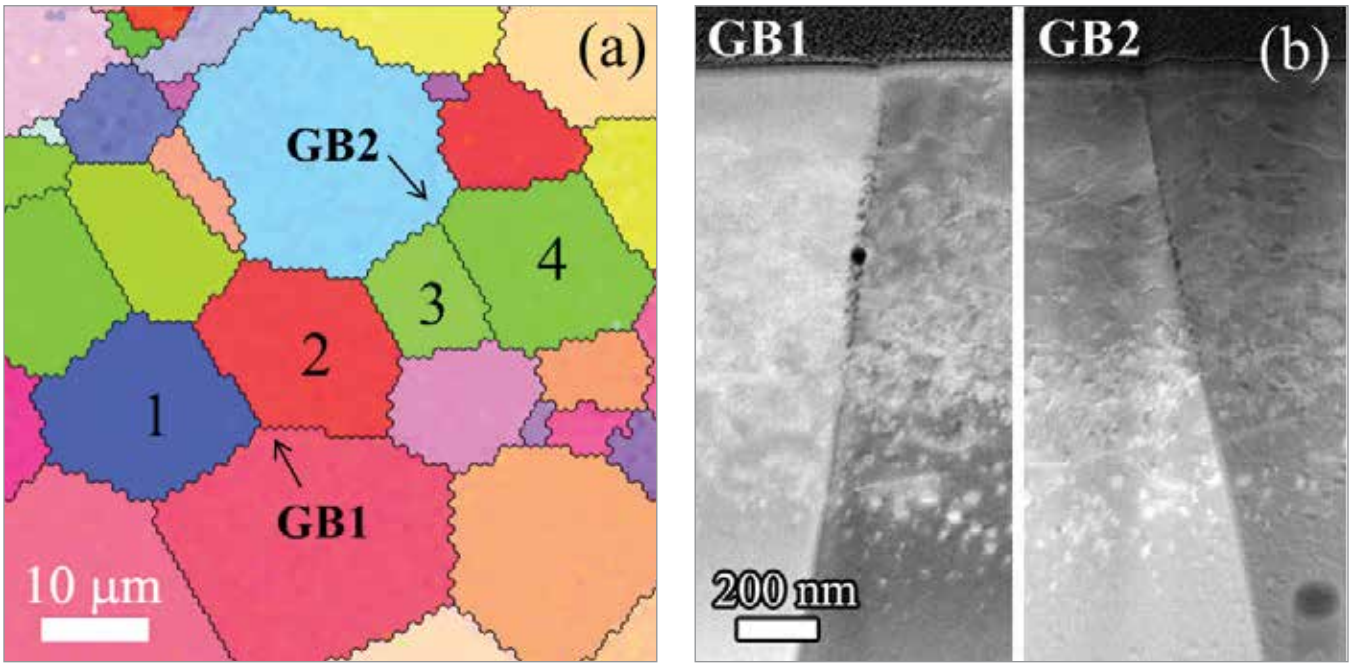


Figure 4. (a) Electronic backscatter diffraction (EBSD) map of Kr-irradiated, polycrystalline UO_2 annealed at 1300°C for one hour. Arrows mark a high-angle grain boundary between Grain 1 and Grain 2 (GB1) and a low-angle grain boundary between Grain 3 and Grain 4 (GB2). (b) Cross-section STEM images comparing the bubble distribution at GB1 and GB2.

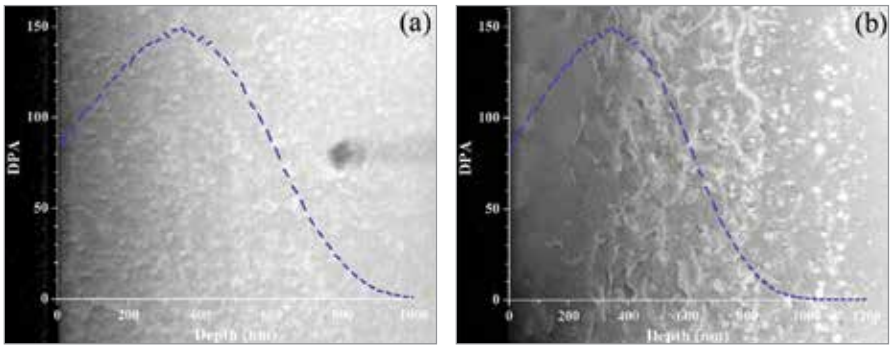


Figure 5. (a) Cross-section STEM images showing the dislocations in as-irradiated and annealed polycrystalline UO_2 at 1300°C for one hour. (b) Dashed lines show the damage profiles in displacements per atom (dpa) calculated by stopping-and-range-of-matter-in-ions (SRIM) software.

bubbles (Figure 4). The small defects induced by Kr-ion irradiation also grow at the annealing temperature. This results in the formation of three distinguished zones that appear at various distances from the surface of the sample (Figure 5): a defect denuded zone at the surface, a dislocation network zone farther from the surface and a dislocation loop zone at the deep region.

Future Activities

The project has revealed bubble evolution as a function of annealing temperature; however, single-bubble pressure in annealed samples of UO_2 has never been measured. Examining the relationship between bubble pressure, bubble size, and annealing temperature will be the primary focus of our work in 2015.

Publications and Presentations

1. B. Valderrama, L.F. He, H.B. Henderson, J. Pakarinen, B. Jaques, J. Gan, D.P. Butt, T.R. Allen, and M.V. Manuel, 2014, "Effect of Grain Boundaries on Krypton Segregation Behavior in Irradiated Uranium Dioxide," *JOM*, Vol. 66, pp. 2562–2568.
2. L.F. He, J. Pakarinen, J.Gan, A.T. Nelson, B. Jaques, D. Butt, A. El-Azab, and T.R. Allen, "Bubble Evolution in Kr-irradiated UO2 under Post-irradiation Annealing," *Journal of Nuclear Materials*, to be submitted.

"State-of-the-art techniques have led to cutting-edge research on nuclear materials."

— Lingfeng He, Assistant Scientist, University of Wisconsin-Madison (Currently Nuclear Fuels Engineer, Idaho National Laboratory)

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	Advanced Test Reactor, PIE facilities
Argonne National Laboratory	Intermediate Voltage Electron Microscope (IVEM)-Tandem facility
Collaborators	
University of Wisconsin - Madison	Lingfeng He (principal investigator), Janne Pakarinen (collaborator)
Idaho National Laboratory	Jian Gan (co-principal investigator)
Boise State University	Darrell Butt (collaborator)

Irradiation and Post-Irradiation Examination of Alloys X-750 and XM-19: Electric Power Research Institute Pilot Program Phase III, Cooperative Research and Development Agreement No. 12-CR-06

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This is the first project to utilize two newly-installed tools: Loop 2A in the ATR center flux trap and the irradiation stress corrosion cracking (IASCC) test systems.

As a means of establishing a basis for the development and execution of joint ATR NSUF/industry programs, the Electric Power Research Institute (EPRI) and ATR NSUF have developed a pilot program involving shared costs and responsibilities. In addition to providing data, this EPRI pilot project is designed to:

- Develop administrative protocols for the project's research, such as cooperative agreements and funding.
- Develop portions of the research capabilities and staffing required to address future research and development needs.
- Develop a protocol for validating data with industry; particularly stress corrosion crack (SCC) growth rate data.

This project is important for three reasons: first, it is the initial industry pilot project for ATR NSUF and establishes protocols for these types of projects; second, it is a full cradle-to-grave characterization of reactor internal material including baseline characterization, irradiation, and post-irradiation examination (PIE); third, it is the first project to utilize two newly installed tools: the controlled water chemistry Loop 2A in the ATR center flux trap and the irradiation-assisted stress corrosion crack (IASCC) test systems.

Project Description

Discussions between ATR NSUF and EPRI resulted in a decision to focus on investigation of the fracture toughness and IASCC growth rates of irradiated high-strength alloys used for boiling water reactor (BWR) repair hardware. Very little of this data exist for the nickel (Ni)-based alloy X-750 or for XM-19, a nitrogen (N)-strengthened austenitic stainless steel, at the exposure levels of interest (up to $1x10^{21}$ n/cm^2). The focus of this EPRI pilot project is on irradiation and characterization of these alloys in both un-irradiated (baseline) and irradiated states, and is being conducted in three phases.

In Phase I, researchers completed the fabrication of specimens from materials provided by EPRI and established the baseline fracture toughness and crack growth rates (CGR) of un-irradiated material. In Phase II, they completed the design and fabrication of specimen holders and performed a safety analysis on a test train in order to meet EPRI objectives for irradiation of tensile and compact tension specimens utilizing Loop 2A in the center flux trap of ATR (Figure 1). In the current and final Phase III, researchers are performing irradiation and PIE of the EPRI specimens previously tested in their unirradiated state.

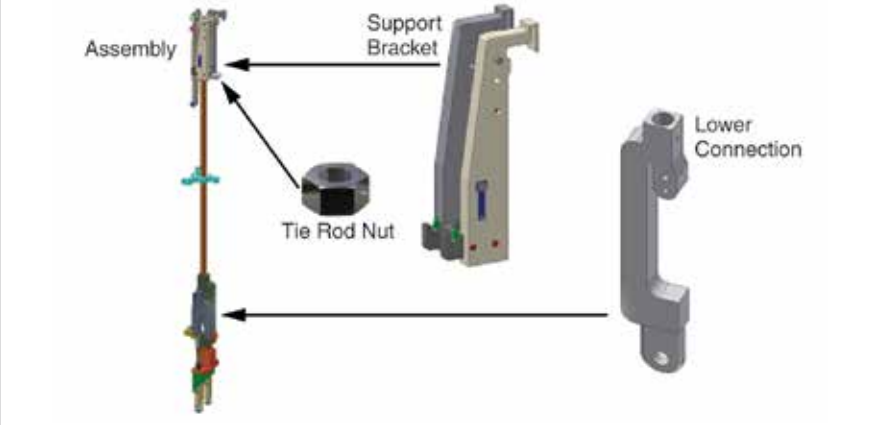

Figure 1. Repair hardware of interest for this study.

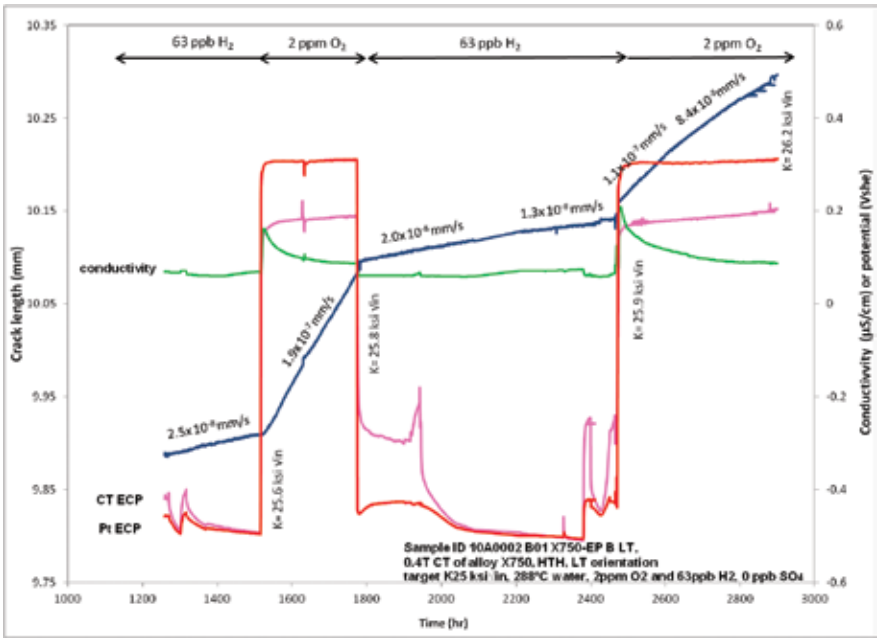

Figure 2. Typical baseline SCC test results for alloy X-750. CGR is the dark blue line.

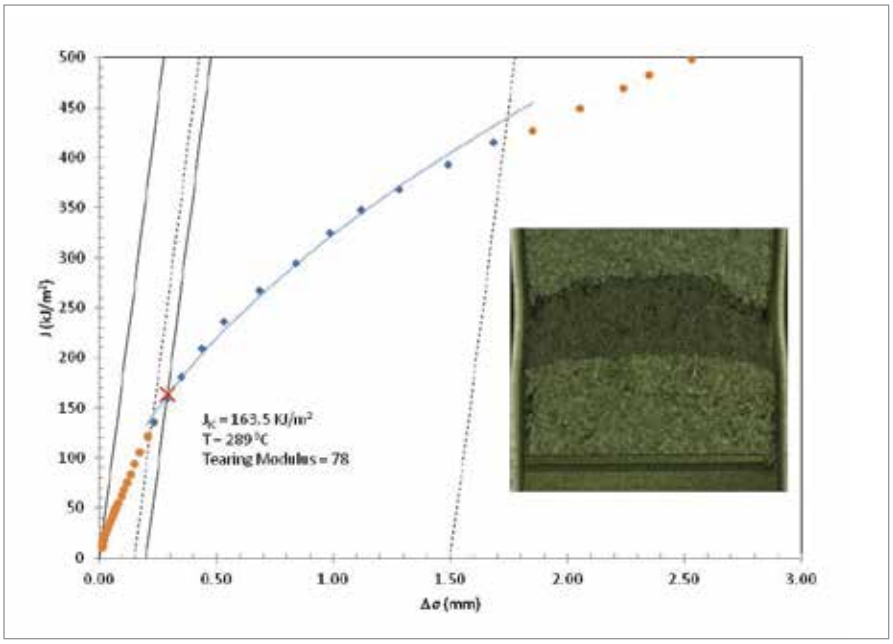

Figure 3. Typical fracture toughness test results for alloy X-750 at elevated (289°C) temperature.

"It is very exciting to see this program bearing fruit. The completion of the IASCC testing rigs and their use in determining crack growth rates of the irradiated materials marks a significant milestone and provides INL with a key capability for the future."

— Bob Carter, Technical Executive, EPRI

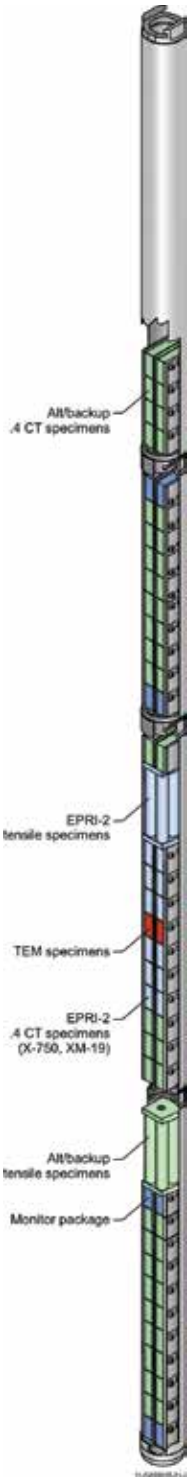

Figure 4. Typical irradiation test train containing CT, tensile, and TEM specimens.

Accomplishments

During this project, several baseline SCC tests were completed on alloys X-750 and XM-19. Cold-worked 9.3% XM-19 was utilized to study potentially similar material property effects between neutron embrittlement phenomena and embrittlement induced by cold-working the material. In addition to SCC tests, fracture toughness tests at temperature (not in environment) and tensile tests at temperature (not in environment) were conducted. Results of the SCC tests were compared to data produced at the General Electric Global Research Company as a means of benchmarking INL's capability to perform these highly specialized experiments. In all cases the measured crack growth rates from the INL tests compared favorably with those produced by the benchmark laboratory.

Figure 2 shows typical SCC test results for alloy X-750 and Figure 3 shows a typical fracture toughness test for alloy X-750. Results of the XM-19 tests are not included here for the sake of brevity, but may be found in INL Report INL/EXT-11-24173, Revision 1, or in the EPRI BWR Vessel Integrity Program Report 1025135 on the EPRI website.

Phase II of this project involved experiment physics and safety analysis as well as test train design for irradiating the specimens. A report was issued in February 2012 showing the results of these analyses and descriptions of the test train designs (Figure 4).

The first irradiation was completed during 2013 on the EPRI-2 capsule, with a target fluence of 2.0×10^{20} n/cm^2 (E>1MeV), in Loop 2A of the ATR center flux trap. Actual measured fluence for this capsule was 1.93×10^{20} n/cm^2. The capsule was shipped from ATR to the Hot Fuels Examination Facility (HFEF) following irradiation and cool down, and was disassembled in preparation for PIE.

In early 2014 the first of two irradiation cycles for the EPRI-3 capsule at a target fluence of 1.0×10^{21} n/cm^2, was completed, also using Loop 2A in the ATR center flux trap. Complications arising from a flow restriction have delayed completion of the second irradiation cycle, but it is expected to be completed in 2015, along with the lowest fluence (5.0×10^{19} n/cm^2) irradiation of the EPRI-1 capsule.

The very first test on the newly constructed IASCC test rig system was also conducted during 2014 on an X-750 specimen that was irradiated in the EPRI-2 capsule. This represented an exciting milestone for the project and for INL. Very stable crack propagation was measured and excellent chemistry control was exhibited in the new systems. The measured crack growth rates in this irradiated X-750, equal to nominally $3.1 - 6.2 \times 10^{-8}$ mm/s in hydrogen water chemistry (HWC) and $1.1 - 22 \times 10^{-7}$ mm/s in normal water chemistry (NWC), were comparable to CGRs that were previously measured in the same alloy in an un-irradiated state. Figure 5 shows a typical CGR plot for this very first IASCC test, demonstrating the system chemistry control in switching between HWC and NWC.

Future Activities

Fracture toughness tests are planned for the first part of 2015, and will be combined with the results of the initial IASCC tests in order to determine if there is any benefit to testing the EPRI-1 specimens at a lower fluence. The IASCC results suggest that even at the medium (EPRI-2) fluence, the material properties have not been affected. EPRI-1 will be irradiated to a target fluence of 5.0 x $10^{\wedge 19}$ n/$cm^{\wedge 2}$ in the ATR center flux trap as originally planned (5 day cycle), and then will be set aside pending fracture toughness test results from EPRI-2. EPRI-3, with a target fluence of 1.0 x $10^{\wedge 21}$ n/$cm^{\wedge 2}$, will continue an additional cycle of irradiation and should be ready for shipment to HFEF in late 2015. Specimens from these test capsules are scheduled for testing in 2015 and 2016.

Testing on the EPRI-2 specimens through the first part of 2015 will include fracture toughness and confirmatory IASCC tests for both X-750 and XM-19. If the EPRI-3 irradiation is completed during that time frame, those specimens may be tested as well.

Publications and Presentations*

1. C. R. Tyler, P. E. Murray, and J. W. Nielsen, 2014, *CRADA EPRI Phase II Design Report*, INL/LTD 12 24400, Rev. 1, February 2014.

**See additional publications from other years in the Media Library on the NSUF website.*

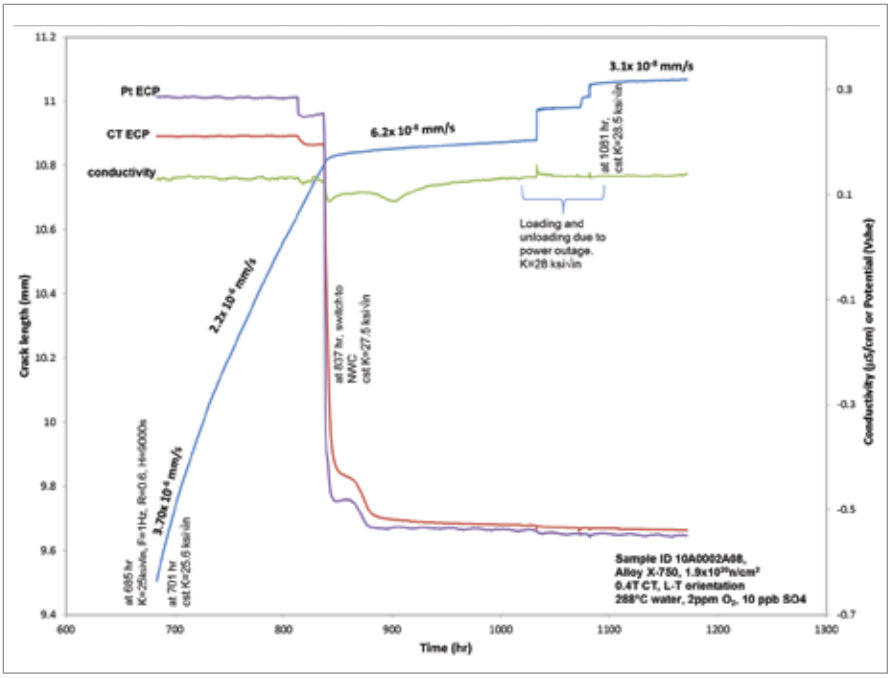

Figure 5. IASCC test results for X-750 irradiated to 1.93 x 10^{20} n/cm^2. CGR is the dark blue line.

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	Advanced Test Reactor, Hot Fuel Examination Facility
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
Idaho National Laboratory	John H. Jackson (principal investigator), Sebastien Teysseyre (co-principal investigator)
Electric Power Research Institute	Robert Carter (co-principal investigator), Peter Chou (co-principal investigator)

Irradiation and Post-Irradiation Examination to Investigate Hydrogen-Assisted Anomalous Growth in Zirconium Alloys

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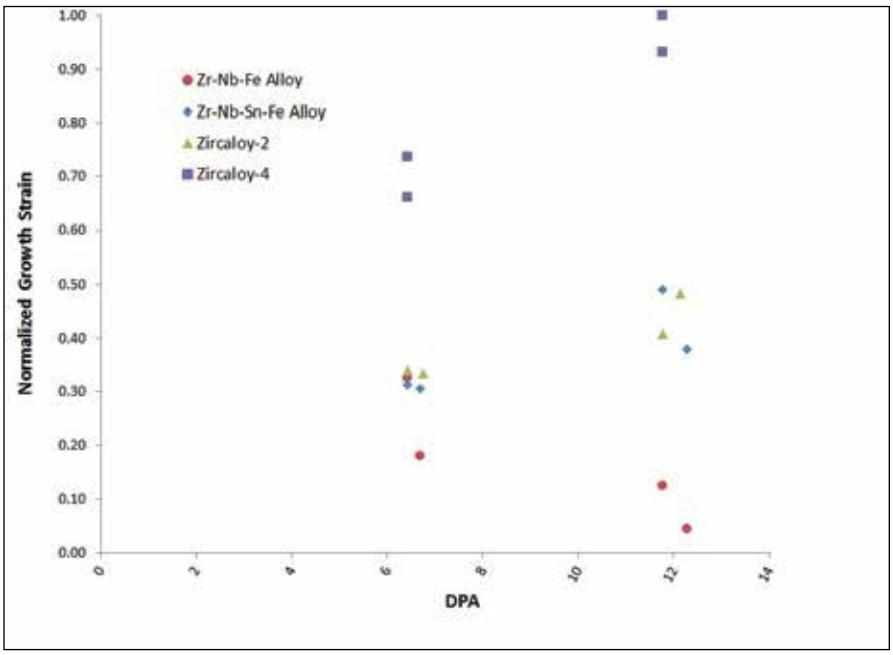


Figure 1. Measured growth strain of selected Zr alloys as a function of dpa, normalized using the maximum growth strain.

This experiment will provide the data needed to select Zr alloys that exhibit small growth strain, which will improve the reliability of commercial reactors by minimizing fuel channel distortion caused by irradiation growth.

Zirconium (Zr) alloy specimens are being irradiated in ATR to study the mechanisms of irradiation-induced growth and its dependence on hydrogen (H) content and neutron fluence. The H produced by corrosion and dissolved in Zr alloys during service in light water reactors (LWR) can form hydrides and may promote irradiation growth. Differential strain resulting from H-assisted irradiation growth is postulated to be partly responsible for fuel channel bowing observed in boiling water reactors (BWR).

Project Description

The objective of this project is to irradiate 200 specimens of various Zr alloys with various H concentrations in ATR up to four different neutron fluence levels. The change in length of the irradiated specimens will be measured to determine the irradiation-induced growth strain, and transmission electron microscopy (TEM) will be performed to study the mechanism of irradiation growth. The knowledge gained from this experiment will be used to select Zr alloys that exhibit small growth strain, which will improve the reliability of commercial reactors by minimizing fuel channel distortion caused by irradiation growth.

Accomplishments

The irradiation is taking place in an inert environment at a temperature of 285°C and at four neutron damage levels expressed in terms of displacements per atom (dpa). Four sets of 50 identical specimens were placed in four separate irradiation capsules identified as A, B, C, and D. Capsule A completed irradiation to 6.7 dpa in January 2013, Capsule B completed irradiation to 12.3 dpa in January 2014, and Capsules C and D are currently being irradiated with a goal of reaching approximately 20 and 30 dpa, respectively (Figure 1).

Capsules A and B were subsequently transferred to INL's Hot Fuel Examination Facility (HFEF) for post-irradiation length measurements. Those were completed for Capsule A in September 2013 and for Capsule B in August 2014.

An instrument was designed and built at INL to measure the length of irradiated specimens in HFEF using remote manipulators. It was tested and validated using specimens of known length. The geometric

profiles of the irradiated specimens from Capsules A and B were then measured and the results analyzed to determine the average growth strain of each specimen. In addition to strain measurements, other objectives included performing as-run reactor physics analysis to determine dpa, measuring neutron fluence using flux wires installed in the experiment, performing heat transfer calculations to determine irradiation temperature, and obtaining temperature indications from monitors (melt wires and silicon-carbide) installed in the experiment. These objectives have also been completed for Capsules A and B.

Future Activities

The growth strain data at 6.7 dpa (Capsule A) and 12.3 dpa (Capsule B) are being evaluated in 2015 to ascertain the effects of neutron fluence, alloy composition, and H content on growth strain. The results will be given to the Electric Power Research Institute (EPRI) and shared with private-sector fuel suppliers (AREVA, Global Nuclear Fuels, and Westinghouse) that provided some of the Zr alloys used in this experiment. TEM will be used to analyze selected irradiated specimens to study the mechanism of irradiation growth. Procedures for preparing specimens for TEM analysis are currently being developed at the irradiated materials characterization facilities at INL and include electropolishing and ion-polishing. Moreover, this project will continue to develop the research capability and staffing required to meet industry needs for research and development of nuclear materials.

Publications and Presentations*

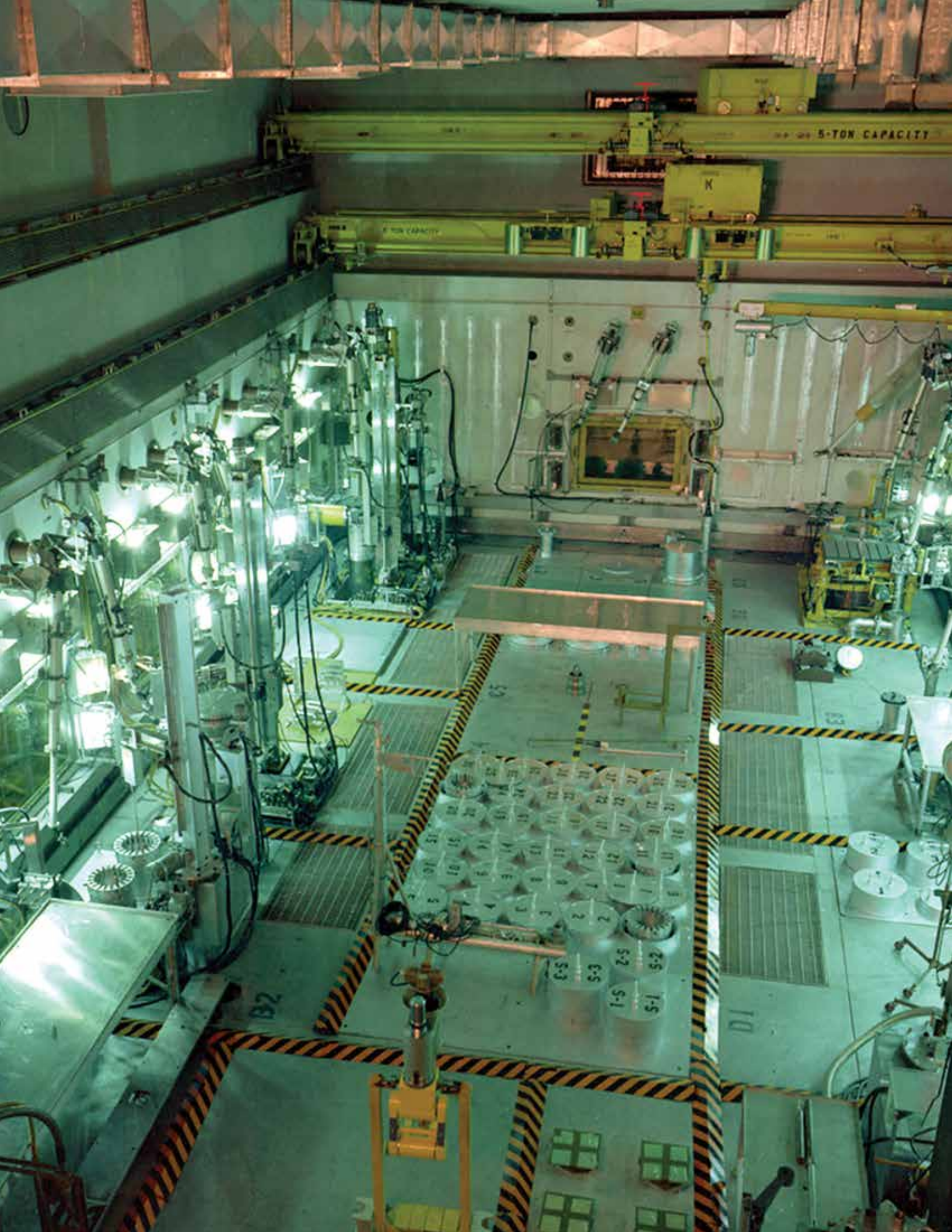
1. P. E. Murray, J. R. Parry, J. H. Jackson, 2014, *Interim Report on Capsule A of the Zirconium Alloy Irradiation Growth Experiment in the Advanced Test Reactor*. INL/LTD 14 31110, Rev. 1, June 2014.

2. P. E. Murray, J. R. Parry, J. Navarro, 2014, *Interim Report on Capsule B of the Zirconium Alloy Irradiation Growth Experiment in the Advanced Test Reactor*. INL/LTD 14 33859, December 2014.

**See additional publications from other years in the Media Library on the NSUF website.*

> *"This project demonstrates that the nuclear industry can benefit from INL's capability in nuclear materials testing."*
>
> **— Dr. Paul Murray, INL**

Distributed Partnership at a Glance	
ATR NSUF and Partners	Facilities and Capabilities
Idaho National Laboratory	Advanced Test Reactor, Hot Fuels Examination Facility
Center for Advanced Energy Studies	Microscopy and Characterization Suite
Collaborators	
Idaho National Laboratory	Paul Murray (principal investigator)
Electric Power Research Institute	Suresh Yagnik, (collaborator)


5-TON CAPACITY
B2
D1

NSUF LIST OF ACRONYMS

AES ..... auger electron spectroscopy
AFCI ..... Advanced Fuel Cycle Initiative
AGR ..... Advanced Gas Reactor
ANL ..... Argonne National Laboratory
ANS ..... American Nuclear Society
APS ..... Advanced Photon Source
APT ..... atom probe tomography
ASCII ..... American Standard Code for Information Interchange
ATLAS ..... Argonne Tandem Linac Accelerator System
ATR ..... Advanced Test Reactor
ATRC ..... Advanced Test Reactor Critical
BF ..... bright field
BWR ..... boiling water reactor
CAES ..... Center for Advanced Energy Studies
CASS ..... cast austenitic stainless steels
CG ..... conventional grain
CGR ..... crack growth rate
CSL ..... coincidence site lattice
CVD ..... chemical vapor deposition
DMSA ..... dimercaptosuccinic acid
DOE ..... Department of Energy
DOE-NE ..... Department of Energy Office of Nuclear Energy
dpa ..... displacements per atom
DSC ..... differential scanning calorimeter
EBR-II ..... Experimental Breeder Reactor II
EBSD ..... electron backscatter diffraction
ECAP ..... equal channel angular pressing

EDS ..... energy dispersive X-ray spectroscopy
EELS ..... electron energy loss spectroscopy
EML ..... Electron Microscopy Laboratory
ENDF ..... Evaluated Nuclear Data File
EPRI ..... Electric Power Research Institute
EUV ..... extreme ultraviolet
EUVR ..... extreme ultraviolet reflectometry
EXAFS ..... extended X-ray absorption fine structure
FCC ..... face-centered cubic
FEG ..... field-emission gun
FFT ..... fast fourier transform
FHR ..... Floride Salt-Cooled High-Temperature Reactor
FIB ..... focused ion beam
F-M ..... ferritic-martensitic
FP ..... fission products
HFEF ..... Hot Fuel Examination Facility
HFIR ..... High Flux Isotope Reactor
HIP ..... hot isostatic pressing
HOPG ..... highly oriented pyrolytic graphite
HRTEM ..... high resolution transmission electron microscopy
IASCC ..... irradiation-assisted stress corrosion cracking
ICPMS ..... inductively coupled plasma mass spectrometry
IFEL ..... Irradiated Fuels Examination Laboratory
IIT ..... Illinois Institute of Technology
IMET ..... Irradiated Materials Examination and Testing Facility
IMPACT ..... Interaction of Materials with Particles and Components Testing
INL ..... Idaho National Laboratory
ISU ..... Idaho State University
IVEM ..... intermediate voltage electron microscope
LAMDA ..... Low Activation Materials Development and Analysis
LEAP ..... local electrode atom probe
LEISS ..... low energy scattering spectroscopy

LWR.....light water reactor
MA..... mechanical alloying
MaCS .....Microscopy and Characterization Suite
MANTRA..... Measurement of Actinide Neutronic Transmutation Rates with Accelerator Mass Spectrometry
MCNP.....monte carlo n particle
MCOE.....Materials Center of Excellence Laboratories
MFC.....Materials and Fuels Complex
MIT ..... Massachusetts Institute of Technology
MITR.....Massachusetts Institute of Technology Reactor
MRCAT .....Materials Research Collaborative Access Team
MSA.....mercaptosuccinic acid
MSTL..... Materials Science and Technology Laboratory
NAA..... Neutron Activation Analysis
NASA ..... National Aeronautics and Space Administration
NCNR..... NIST Center for Neutron Research
NCSU ..... North Carolina State University
NDE.....nondestructive evaluation
NGA ..... Nuclear Energy Agency
NE..... Nuclear Energy
NEET .....Nuclear Energy Enabling Technologies
NEUP..... Nuclear Energy University Programs
NFS..... nanostructured ferritic steels
NGNP..... Next Generation Nuclear Plant
NIST .....National Institute of Standards and Technology
NMR.....Nuclear Magnetic Resonance
NRC.....Nuclear Regulatory Commission
NSUF ..... Nuclear Science User Facility
ODS .....oxide dispersion strengthened
OECD.....Organisaton for Economic Co-operation and Development's
ORNL .....Oak Ridge National Laboratory
OSU .....Oregon State University
PED.....precession electron diffraction

PI ..... Principal Investigator
PIE ..... post-irradiation examination
PNL ..... Pacific Northwest Laboratory
PNNL ..... Pacific Northwest National Laboratory
PSU ..... Pennsylvania State University
PWR ..... pressurized water reactor
REDC ..... Radiochemical Engineering Development Center
RERTR ..... Reduced Enrichment Research and Test Reactors
RF ..... radio frequency
RIS ..... radiation-induced segregation
RPL ..... Radiochemistry Processing Laboratory
RTE ..... rapid turnaround experiment
SCC ..... stress corrosion cracking
SEM ..... scanning electron microscopy
SPS ..... spark plasma sintering
SRB ..... Scientific Review Board
SRIM ..... Stopping and Range of Ions in Matter
SS ..... stainless steels
SSRL ..... Stanford Synchrotron Radiation Laboratory
STEM ..... scanning transmission electron microscopy
TAMU ..... Texas A&M University
TEM ..... transmission electron microscopy
TMS ..... The Minerals, Metals, & Materials Society
TRIGA ..... Training Research Isotope General Atomics
TRISO ..... tristructural isotropic
TRU ..... transuranic waste
UCB ..... University of California, Berkeley
UCF ..... University of Central Florida
UFG ..... ultra-fine grained
UM ..... University of Michigan
UNLV ..... University of Nevada, Las Vegas
USU ..... Utah State University

UW ..... University of Wisconsin
VHTR ..... very-high temperature reactor
XAFS ..... X-ray absorption fine-structure spectroscopy
XANES ..... X-ray absorption near-edge spectroscopy
XAS ..... X-ray absorption spectroscopy
XPS ..... X-ray photoelectron spectroscopy
XRD ..... X-ray diffraction
µm ..... micrometre

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