Office of Nuclear Energy

NEET-Reactor Materials Award Summaries

Nuclear Energy Enabling Technologies-Reactor Materials May 2016

Material science plays a pivotal role in the extension of the life of the existing fleet of nuclear reactors; in the deployment of new modern light water reactors, advanced reactors with non-water coolants, and small modular reactors and in the storage, recycling and disposal of used nuclear fuel. Understanding and overcoming material degradation in an extreme environment is essential for safe and efficient operation. Deployment of new, advanced materials may make construction of new plants more economical. Materials research is featured in all of the major research thrusts within Department of Energy Office of Nuclear Energy (DOE-NE) research portfolio. The Nuclear Energy Enabling Technologies–Reactor Materials Crosscut (NEET-RM) is designed to provide support and coordination amongst these programs by enabling the development of innovative and revolutionary materials and provide broad-based modern materials science support to the materials research within all of the DOE-NE programs and providing coordination of research with the five other DOE-NE R&D programs (Light Water Reactor Sustainability, Small Modular Reactors, Very High Temperature Reactor, Advanced Reactor Technologies, and Fuel Cycle Research and Development). This provides an opportunity to coordinate responses and provide additional gains from joint research nationally and internationally by sharing research with all relevant programs. There are ongoing needs for new research tools, improved infrastructure, and coordination efforts to improve research efficiency.

The needs can be categorized into three major categories:

- *Research and development needs:* The major DOE-NE programs are making strides in a number of key areas. However, developing new characterization tools and fabrication and testing techniques may make research more efficient and enable new discoveries. For example, the development of computational thermodynamic tools may enable accelerated aging tests and reduce experimental burden during alloy development. Ion irradiation may be used to complement neutron irradiation at a reduced cost. Advanced welding or joining techniques may overcome traditional component limitations but will require dedicated research.

- *Infrastructure needs:* Available tools limit some of the research. New tools and facilities may enable research in multiple programs. For example, there is currently no fast reactor irradiation capability in the United States.

- *Coordination and collaboration needs:* Finally, in some areas, formal collaboration and discussion between the programs within DOE-NE and other relevant efforts may promote more efficient research and eliminate overlap. Code qualification and alloy development are examples of common research topics that may benefit from additional, broad discussion.

The Reactor Materials Crosscut effort will enable the development of innovative and revolutionary materials and provide broad-based modern materials science support to the materials research within all of the DOE-NE programs. This will be accomplished through innovative materials development; promoting the use of modern materials science; and establishing new, shared research partnerships. Today, the NEET-RM Crosscut is pursuing all of these areas actively via a competitive process. Four rounds of competition for three year research awards have been completed. Also, one round of competition exclusively for universities was completed in 2011 through the Nuclear Energy University Program (NEUP).

In 2011, four awards, totaling $2,011,730, were granted through NEUP in advanced materials development. The research for these awards has been completed and was documented in the FY15 Annual Report. Concepts that were awarded under this solicitation include steel foams for lightweight shielding applications, austenitic oxide dispersion strengthened steels, MAX phases and high entropy alloys.

In 2012, nine awards, totaling $7,954,651, were given for advanced materials development concepts. These included Fe-based steels, metal-ceramic composites, ceramic composites, amorphous coatings, nanocrystalline SiC, and radiation-tolerant cable insulation.

In 2013, seven awards, totaling $6,898,673, were granted for the development of advanced characterization techniques. These projects include developing advanced synchrotron diffraction techniques, spherical nano-indentation techniques, and microscale and mesoscale simulation tools with advanced microscopy techniques.

The 2014 competition was focused on the development of advanced joining methods for advanced materials. Updates on the three awards, totaling $3,000,000 are provided. The projects include developing advanced techniques to build functional gradient welds for joining dissimilar metals, fusion welds for FeCrAl alloys, and low-energy solid state welds for steel.

The 2015 competition centered on advanced material development concepts as in 2012. Two projects were funded totaling $1,994,292. These projects will develop metal-ceramic composites and nanoprecipitate-strengthened ferritic steels.

Since FY 2012, NEET-RM has funded 21 projects for a total investment of $19,847,616.

Overall, the open competition has been very successful with high quality proposals being submitted from a very diverse set of institutions. Participation and partnerships have grown with each solicitation. As shown in the following sections, the technical quality and innovation have been very high, consistent with the expectations and goals of the NEET program.

2012 NEET-RM Open Award Research Summaries

In 2012, the NEET Crosscutting Reactor Materials program sought applications for advanced materials discovery and development. Successful completion of awards has provided structural and clad materials that dramatically improve performance over traditional materials used in terrestrial and space reactors and in the nuclear fuel cycle.

Specific goals may include:

- Improvement in mechanical performance by a factor of 5-10 over traditional materials
- Increase in maximum operating temperature of greater than 200 C over an 80 year lifetime
- Increased radiation tolerance to beyond 300 dpa

Such performance would enable significantly improved safety, performance and reliability for future advanced reactor and fuel cycle designs. However, such improved performance cannot be at the expense of other properties or performance.

Applications were requested that describe innovative materials concepts, concept advantages, concept limitations, and key development needs. Successful applications described innovative materials that offer the potential for revolutionary gains in reactor and fuel cycle performance. Materials that could be applied to multiple reactor designs, components, and concepts were given preference over materials restricted to a single reactor concept, component, or coolant.

Radiation Tolerance and Mechanical Properties of Nanostructured Ceramic/Metal Composites

Michael Nastasi, University of Nebraska-Lincoln
Michael Demkowicz, Massachusetts Institute of Technology
Lin Shao, Texas A&M University
Funding: $979,978 (10/1/2012-9/30/2015)

Description of project: The objective of this proposal was to explore the development of advanced metal/ceramic composites with greatly improved radiation tolerance, stability above 500 °C, and improved mechanical performance combining the good properties of glasses (high strength and elastic limit, corrosion resistance) with those of crystals (high toughness, strain hardening). The ceramic component of the composite consists of a high crystallization temperature amorphous material composed of SiOC, while the metal component is Fe, chosen as a model material for steel. We hypothesized that the combination of the composite constituents as well as the interfaces between them will provide significantly enhanced radiation tolerance, similar to or superior to those observed in metallic nanolayered structures, but in a more engineering-relevant material system.

Impact and value to reactor applications: The need to develop advanced cladding that does not react to form hydrogen is urgent considering past accidents at Fukushima. The project will aim to develop super tough and ultra-high temperature resistant materials that are in critical need for nuclear applications under extreme conditions where in-core materials have to withstand neutron damage and high temperature. The potential impact will be the development of a new class of ceramic/metal composites that can be adapted for engineering applications, resulting in dramatically improved materials performance for advanced reactors.

Recent results and highlights: Studies have shown that the amorphous SiOC material possess good thermal and irradiation stability over a wide range of composition, irradiation dose and irradiation temperature. Specifically, the material showed no evidence of crystallization up to a temperature of 1200 °C and an annealing time of 2.0 hours. In addition, amorphous SiOC films remain amorphous after both light ion (He) and heavy ion (Kr) irradiation within a wide envelope of irradiation conditions. Fig. 1 presents the typical TEM micrographs of amorphous SiOC films (a) before, and after (b) 20 dpa He irradiation at 600 °C, (c) 5 dpa Kr irradiation at 300 °C. Both high resolution TEM micrographs and corresponding selective area diffraction (SAD) patterns clearly show that SiOC films retain their amorphous structure without any crystallization, void formation or element segregation.

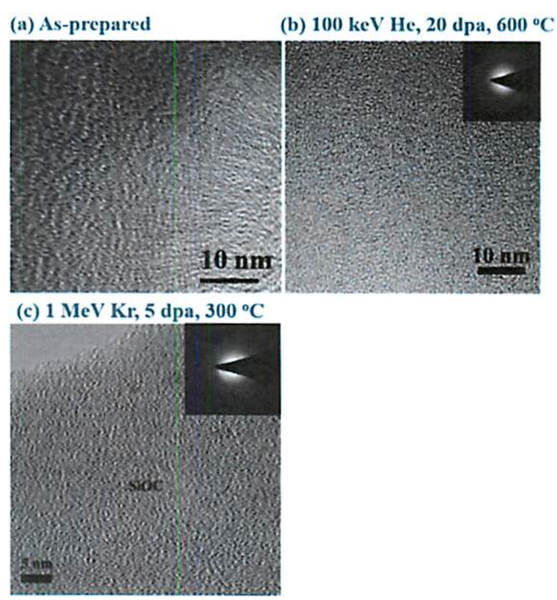


Fig. 1.Typical TEM micrographs of SiOC films (a) before, and after (b) 20 dpa He irradiation at 600 °C, (c) 5 dpa Kr irradiation at 300 °C. The inset is the corresponding SAD pattern of each micrograph.

The highlights of results suggest that neither local atomic perturbations nor large damage cascade zones lead to crystallization, element segregation or void formation of this material. These important observations support the hypothesis that some amorphous alloys are radiation-indifferent; within an envelope of irradiation conditions, radiation-induced damage anneals out as fast as it is created, allowing these alloys to persist indefinitely in an externally driven steady-state, with time-invariant structure and properties.

Accelerated Development of Zr-Containing New Generation Ferritic Steels for Advanced Nuclear Reactors

Lizhen Tan, Ying Yang, Oak Ridge National Laboratory
Kumar Sridharan, Beata Tyburska-Pueschel, and Lingfeng He, University of Wisconsin-Madison
Funding: $849,000 (10/1/2012-9/30/2015)

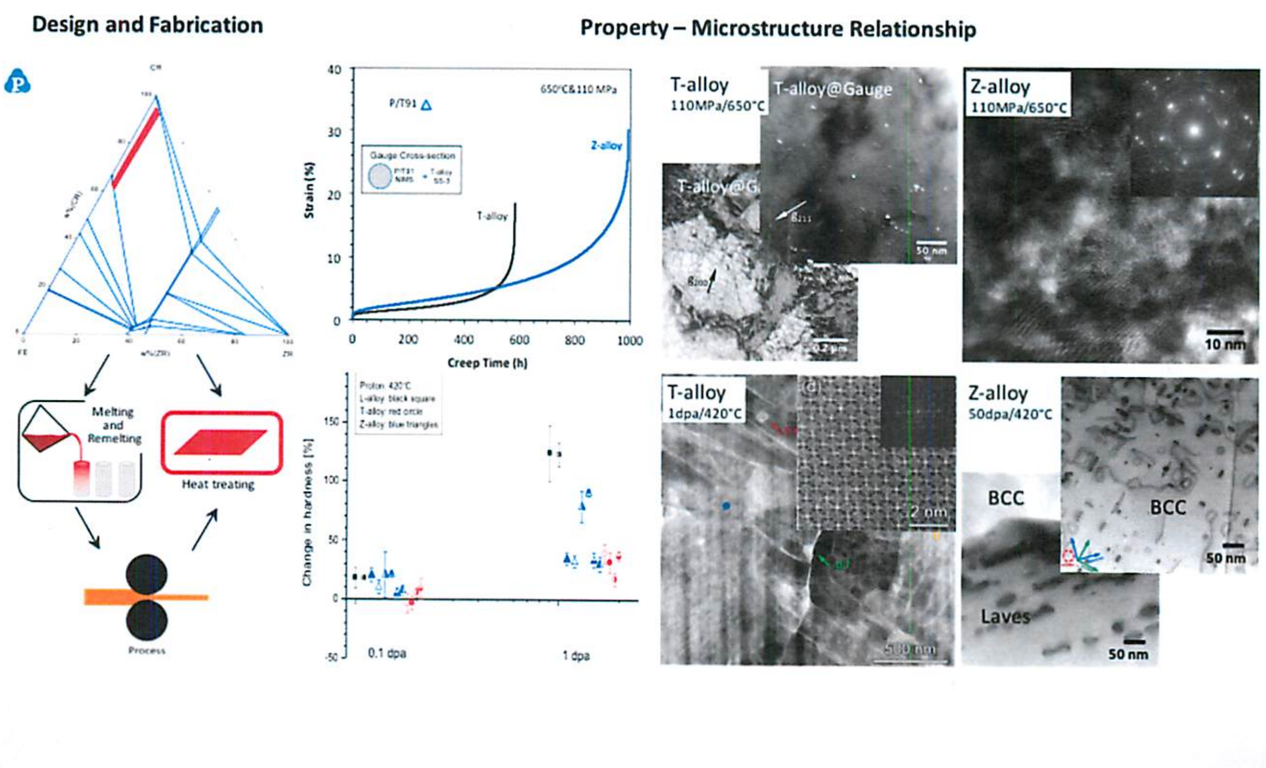
Description of project: The project developed a new generation zirconium-containing ferritic steels, with the aid of modern computational microstructural modeling tools, to enhance high temperature creep resistance and improve radiation resistance.

Impact and value to reactor applications: Development of inherently low-swelling ferritic steels with enhanced creep resistance, which are produced by conventional economic steelmaking techniques, would represent a significant step towards development of components for high temperature, high dose reactors. The thermodynamic property database of the Fe-Cr-Zr-X system enables broader applications in nuclear power plants for understanding the interaction between the Zr-alloy fuel cladding and the structural steel core components.

Results and highlights:

- A thermodynamic database containing Fe, Cr, Zr, W, Mo, Nb, Ti, C was developed and used to design alloy chemistry and heat treatment scheme for alloy fabrication using conventional steelmaking methods (Fig. 1).
- Three series of Zr-containing ferritic alloys, denoted as Z- (high Zr > 4 wt.%), T- (9Cr), and L- (15Cr) alloys, were developed and tested. The Z- and T-alloys demonstrated superior high temperature mechanical properties and lower or comparable ion-irradiation hardening compared with conventional ferritic/martensitic steels, e.g., Grade 91.
- Microstructural characterization using advanced techniques including X-ray diffraction, SEM, and high resolution TEM/STEM-EDS established property-microstructure relationship.

Figure 1



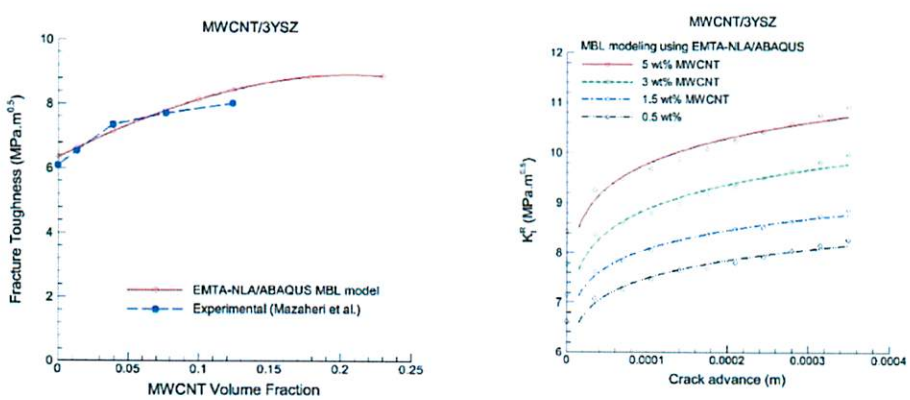
Nanocrystalline SiC and Ti_3SiC_2 Alloys for Reactor Materials

C. H. Henager, Jr., Pacific Northwest National Laboratory
Funding: \$977,577 (10/1/2012-9/30/2015)

Description of Project: This project synthesized, characterized, and modeled SiC/Ti_3SiC_2 dual-phase composites formed by simultaneous polycarbosilane pyrolysis that forms nanocrystalline SiC and Si/TiC displacement reactions that form SiC/Ti_3SiC_2 interpenetrating phase composites. To the best of our knowledge these are the first such dual phase composites made by combining these two methods.

Impact and value to reactor applications: The value of this material for reactor applications is that it can be made denser with improved fracture toughness and improved thermal conductivity compared to SiC/SiC composites, and polymer processing allows near net shape fabrication. A caution for using Ti_3SiC_2, and other MAX phases, in reactor applications is that our data show that fission produced diffusion is much higher than SiC. This may have serious implications for the use of this type of material for fuel cladding.

Recent Results and Highlights: Important experimental results from a diffusion study of fission product surrogates (Ag and Cs) and a noble metal (Au) in MAX phase Ti_3SiC_2, cubic/hexagonal SiC, and a dual-phase nanocomposite of SiC/Ti_3SiC_2 showed that all implanted species were mobile in MAX phase Ti_3SiC_2 and diffused to the surface at moderately high temperatures (873 to 973 K). However, Ag in SiC was observed to be immobile at the highest temperature (1273 K) applied in this study. These results suggest caution in using Ti_3SiC_2 as a fuel cladding material for advanced nuclear reactors operating at high temperatures.
A new modeling method (Fig. 1) was developed during this project to model thermal conductivity of composites, specifically multi-phase composites such as SiCf/SiC composites, accounting for radiation damage degradation. The use of atomistic data is integrated into the methodology, which makes it particularly powerful. This project also developed a new micromechanics model of carbon nanotube (CNT) toughening that demonstrated the limitations of CNTs in increasing the fracture toughness of SiC. Toughness data, along with model data for CNT toughening, shows that homogeneously adding or dispersing CNTs in SiC cannot achieve significant increases in toughness. This result was anticipated but, prior to our work, had not been proven to be the general case. Our model clearly shows that dispersed CNTs cannot achieve toughness values of 15 to 20 MPa$\sqrt{m}$ for SiC. Unfortunately, this project was unsuccessful in synthesizing an alternative CNT-based architecture that could be incorporated into the composites being studied that would increase toughness.



(a) (b)

Figure 1. Results from the PNNL toughness modeling. Shown in (a) is predicted and experimental Mode I fracture toughness vs. Multi-Walled (MW)CNT volume fraction for tested MWCNT/3YSZ composites and (b) predicted stress intensity factor vs. crack advance for MWCNT/3YSZ composites as a function of MWCNT content. Note the peak in toughness at about 0.2 MWCNT volume fraction in (a).

Study of Intermetallic Nanostructures for Light-Water Reactors

Prof. Niels Gronbech-Jensen and Dr. Benjamin Beeler, University of California-Davis
Profs. Mark Asta and Peter Hosemann, University of California-Berkeley
Dr. Stuart Maloy, Los Alamos National Laboratory
Funding: $749,940 (10/1/2012-9/30/2015)

Description of project: This project aimed to study and utilize self-forming precipitates in structural steels for nuclear applications. We utilized the concept of interfaces as sinks for radiation induced defects, as has been studied for oxide dispersion strengthened (ODS) alloys, and expanded it to intermetallic precipitates. Certain commercially available tool steels, commonly known as MarAging steels, form NiAl based nanoclusters by a simple heat treatment. The question is if these clusters provide radiation tolerance benefits comparable to oxide clusters in ODS alloys. These precipitates could act as defect sinks while yielding radiation self-hardening, since more clusters precipitate under radiation and therefore provide more defect sinks. Atomistic computational modeling and experiments address this basic concept and evaluate its feasibility.

Impact and value to reactor applications: This work can potentially lead a path towards a new class of radiation tolerant alloys, as well as provide important insight into the fundamentals of radiation precipitation behavior and the role of coherent interfaces as defect sinks. These insights are likely to be relevant to a wide variety of materials.

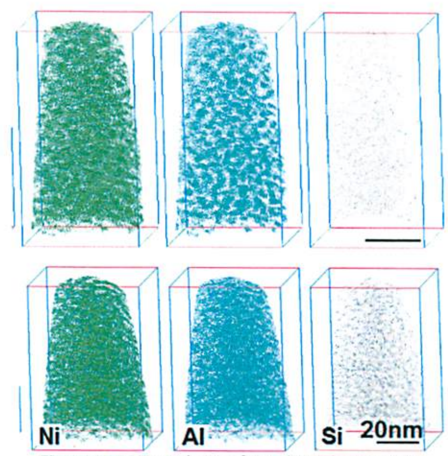


Figure 1 APT analysis of irradiated materials

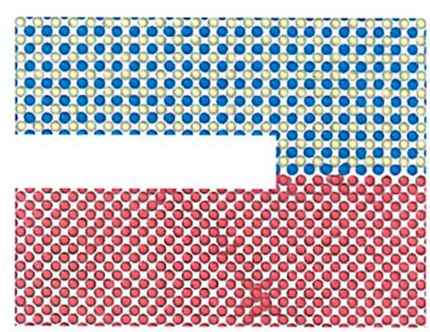


Figure 2- Modeling the BCC-Fe/ B2-NiAl interface.

Recent results and highlights: Our recent experimental results on the material with and without precipitates show that low temperature irradiations (RT) do cause the development of cluster precursors in a material, which has no clusters prior to irradiation. In a material with preexisting clusters the Ni-Al precipitates stay of similar dimension, and a small amount of Si enrichment was found in the clusters after radiation. Figure 1 shows the APT data of the material before and after radiation. We will evaluate if severe mechanical property changes occur at long times or the radiation causes significant changes in the microstructure. Our recent modeling efforts have produced the first interatomic potential capable of reasonably describing the ternary FeNiAl system. Systems with (100), (110) and (111) BCC-Fe/B2-NiAl interfaces are being investigated with and without radiation to determine the potential role of such interfaces as defect sinks. Radiation damage is varied in depth and angle with respect to the interface, as well as energy of the impinging particle. In Figure 2, an example (100) interface of BCC-Fe with B2-NiAl is displayed.

Nanoscale Stable Precipitation-Strengthened Steels for Nuclear Applications

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Ömer N. Doğan, Paul D. Jablonski: National Energy Technology Laboratory
Kristin Tippey, Kip O. Findley, John G. Speer: Colorado School of Mines
Semyon Vaynman, Morris E. Fine, Yip-Wah Chung: Northwestern University
Jonathan Almer, Argonne National Laboratory
Funding: $880,000 (10/1/2012-5/12/2016)

Description of project: Modern alloy development strategies have been implemented to produce thermally stable precipitation hardened steels that are ***manufactured by conventional methods*** for nuclear reactor structural applications. These strategies have shown potential to produce desirable nano-precipitate dispersions, which have the potential to substantially improve mechanical properties, increase thermal service duration, and improve irradiation resistance, while reducing cost and increasing manufacturability. Two classes of materials are being designed: Tailored Precipitate Ferritic (TPF) steels and Advanced High-Cr Ferritic-Martensitic (AHCr-FM) steels.

Impact and value to reactor applications: Optimization of current alloys and development of novel alloys will produce materials with improved mechanical properties, thermal service capabilities, and irradiation resistance. These modern materials will allow reactor designers greater flexibility to improve performance, durability, and safety.

Recent results and highlights: AHCr-FM steel alloys have been produced to exploit the benefits of low-carbon, high-nitrogen compositions to produce a high density of Nb or V nitrides. Figure 1 shows tempering and aging studies comparing the baseline P92 alloy with newly developed low carbon (LC) and Niobium free (0Nb) alloys, which have achieved increased precipitate volume fraction in the nano-scale size range desired for irradiation resistance and mechanical property optimization. TPF alloy design strategies resulting from computational and experimental understanding have produced microstructures with nanoscale Al-Ni precipitates and nanoscale Nb or V carbides. The homogenously distributed nanoprecipitates have been subjected to extended high-temperature aging experiments and the resulting nanoscale characterization shows excellent thermal stability. Controlled thermomechanical treatments of both TPF and AHCr-FM alloys are underway to link nanoscale precipitate distributions with stable dislocation sinks and further optimize mechanical properties, thermal performance, and irradiation resistance. Initial irradiation testing has shown improvement relative to standard alloys.

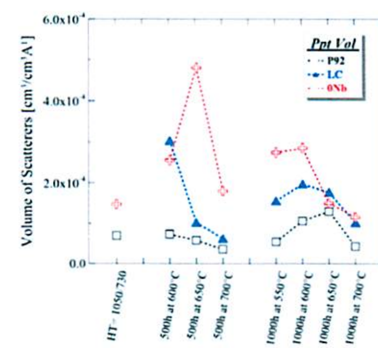

Figure 1 – Volume fraction of nanoscale precipitates after tempering and aging, showing AHCr-FM alloying strategies increase precipitate volume fraction relative to standard P92 material.

Nanostructured Fe-Cr Alloys for Advanced Nuclear Energy Applications

R. O. Scattergood, North Carolina State University
Funding: $788,156 (10/1/2012-9/30/2015)

Description of the project: This research addresses the development of Fe14Cr base alloys that have improved performance in terms of reduced swelling due to He bubble formation. Alloy additions are made to Fe14Cr base alloys to develop nanocrystalline alloys that can mitigate He bubble formation by a combination of nano-oxides and nanoscale grain boundaries that trap He atoms before large He bubbles are produced. The alloys were synthesized using SPEX ball milling. The selected non-equilibrium alloy elements introduced can segregate to grain boundaries to stabilize nano-scale grain size at high temperatures (thermodynamic stabilization), and facilitate He ion trapping. He ion irradiation to evaluate the performance of the candidate alloys was done in conjunction with facilities at Los Alamos National Laboratory.

Impact and value to reactor applications: The mitigation of the formation of large He bubbles is essential to avoid swelling of reactor components during the intended lifetime of fission reactors.

Research highlight 1: The effect of different nano-oxide particles on the resistance of He irradiation in Fe-14CrxZr/Hf base alloys was investigated. High densities of nano-size ZrO_2 particles are found in the ferritic matrix with one or more He bubbles existing near-by, showing a strong He trapping effect by nano ZrO_2 particles. This is coupled with good stability of the nano ZrO_2 particles when subjected to 200 keV He irradiation. He bubbles and nano HfO_2 particles evolve into core-shell structures (yellow arrow) and thus reduce void swelling by suppressing the growth of He bubbles as shown in Figure 1.

Research highlight 2: A new software program was developed for analysis of void swelling, bubble size and density from a series of High Resolution Transmission Electron Microscope (HRTEM) depth-profile images. This was applied to a range of irradiated microstructures after 200 keV irradiation at several temperature in order to evaluate contributions from nano-oxide he trapping and grain-boundary trapping. As shown in Figure 2, with decreasing oxide particle size, swelling percentage drops. A slight decrement of nano-oxide particle size of about 1 nm was shown to result in over 30% reduction of the swelling percentage. Effective oxide particles sizes must be less than 3-4 nm. A grain-boundary He trapping effect was also observed when the average grain size becomes sufficiently small.

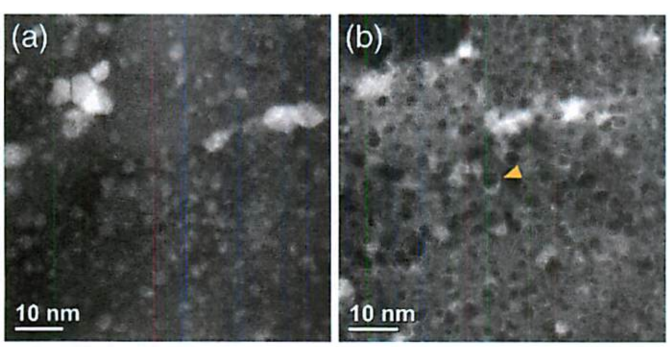

Figure 1. The structure of HfO_2 particles in Fe14Cr2Hf_ subject to 200 keV He^+ irradiation at 500°C. HAADF-STEM image in (a) non-irradiated area and (b) irradiated area at the depth of 550-600 nm. As marked by the yellow arrow, the morphology of HfO_2 particle evolves to a shell structure around the trapped He bubble.

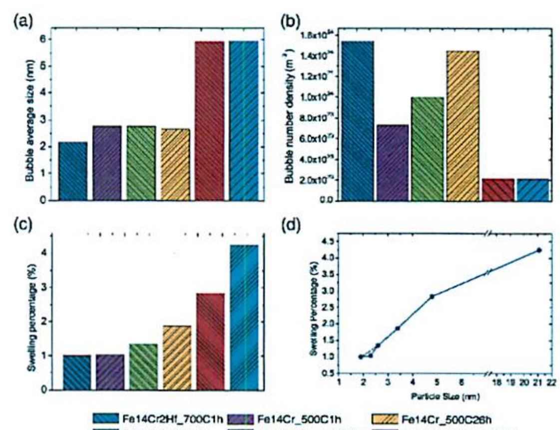

Figure 2. Comparison of the maximum value of (a) void swelling, (b) He bubble size and (c) bubble density for Fe14CrHf/Zr alloys. (d) the relationship between nano-oxide particle size and void swelling. Nano grain size is notably effective for the trapping of 2-3 nm He bubbles.

SiC Composite for BWR Channel Application

Ken Yueh, Electric Power Research Institute
In Partnership with AREVA, GNF, INL, MIT, ORNL and WEC
Funding: $800,000 (10/1/2012-9/30/2016)

Description of Project: The commercial LWR industry has experienced a number of severe boiling water reactor (BWR) channel bowing issues that required rechanneling of fuel assemblies. The issue is caused by fast flux gradients across the channel faces and less understood shadow corrosion. SiC is known to be irradiation stable after saturation and thus could provide a total solution to the problem. After the Fukushima accident, the benefits of SiC high temperature performance in steam environments have gained more prominence. The goals of the projects are to (1) evaluate SiC properties against design requirements, (2) resolve any issues identified, and (3) generate physical property data to support the commercial demonstration of a SiC composite BWR channel.

Impact and Value to Reactor Applications: Successful commercialization of the technology is expected to solve the channel bowing issue and provide margin in design basis and severe accidents. Replacing zirconium with SiC based channels significantly reduces exothermic heat generation in a severe accident and thus provides much needed response time and reduced emergency core cooling system requirements. The lower neutron capture cross-section of SiC could translate to financial savings in terms of reduced enrichment requirements.

Recent Results and Highlights: Majority of the issues identified by the team have been evaluated and the results indicate SiC composite could meet the mechanical design requirements of a BWR channel. Corrosion under BWR oxidizing environment remains the last issue to be resolved. In-core testing of a sister sample under pressurized water reactor (PWR) reducing conditions conclusively showed the high weight loss reported earlier was due to the BWR oxidizing condition. Research is in progress to modify the surface to provide a barrier to separate the SiC from the coolant. An approach taken has been to introduce a stabilizing element near the end of the SiC infiltration process so that the combined corrosion products form a stable and protective layer. Initial evaluations included doping with titanium and zirconium to form a layer of protective water insoluble $TiSiO_2$ or $ZrSiO_2$ oxide. Preliminary results indicate that a titanium doped surface does not form a stable and protective $TiSiO_2$ layer. The apparatus and procedure used was not able to produce a surface layer of desired ZrC/SiC ratio and quality (surface layer cracked). However, after corrosion testing areas of continuous oxide, matching the composition of $ZrSiO_2$ was detected, see Figure 1. Additional process development will be needed to improve the ZrC+SiC coating quality and composition to determine if the oxide layer is protective.

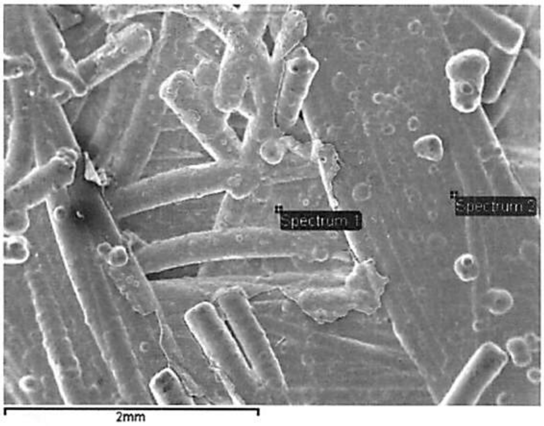

Figure 1 – SEM image of zirconium doped sample showing a layer with $ZrSiO_2$ composition.

Radiation Resistant Electrical Insulation Materials for Nuclear Reactors Using Novel Nanocomposite Dielectrics

Robert Duckworth, Oak Ridge National Laboratory
Funding: $940,000 (10/1/2012-6/30/2016)

Description of project: The primary objective of this project is to initiate the development of a transformative materials system using nanocomposite dielectric technology to increase the radiation and thermal resistance (>150 ºC) of electrical insulating materials for nuclear environments. Recently there has been renewed interest in nuclear reactor safety especially as it relates to cable insulation from commercial reactors approaching and surpassing their original 40 years' service. While the current materials that are deployed in nuclear reactors have been effective, the next generation of nuclear reactors may push these materials beyond these limits where the combined environmental effects of radiation, temperature and moisture, or operation during abnormal conditions may become more important.

Impact and value to reactor applications:
Based on the results to date, we believe that feasibility of nanocomposite dielectrics has been demonstrated. While there is additional study needed to determine the best concentration for a given set of conditions, the formulations of these dielectrics has been tailored to allow with some optimization for manufacturers to replace their base resin system with this new mixture and improvements could be realized without a significant increase in manufacturing costs. Given the nature of the change in electrical properties with respect to nano-composite additions, deployment of these nano-dielectrics in power and instrumentation cables could result in improvements in in-situ electrical spectroscopic methods that could more effectively show cable aging due to accidents and/or long-term cable aging mechanisms.

Recent results and highlights:
Three different in-situ methods for three different base resin materials have been developed for the inclusion and production of nanoparticles in dielectrics. These base resin materials, polyvinyl alcohol (PVA/Cross-Linked (XL) PVA), polyethylene (PE, XLPE), and polyimide (PI) have been paired with either MgO, SiO_2, Al_2O_3 to produce nanodielectrics whose electrical, mechanical, & chemical properties with and without irradiation have been impacted by these additions. An example is shown in Figure 1 where the breakdown performance improved for XLPE SiO_2 nano composites. When gamma irradiation was applied up to 18 MRad, concentrations above 3wt% caused parameters such as permittivity and dissipation factor to change especially at low frequencies. Current work aims to optimize and understand nanoparticle concentration for the combination of thermal aging and higher accumulated doses of gamma irradiation.

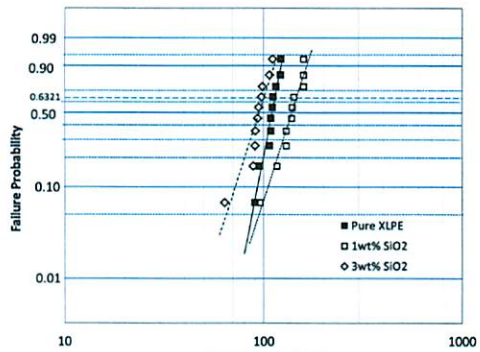

Figure 1. Weibull plot for the dielectric breakdown strength of XLPE SiO_2 nano-composites with different percentage of SiO_2 nanoparticles

Radiation-Induced Ductility Enhancement in Amorphous Fe and Al_2O_3+TiO_2 Nano-Structured Coatings in Fast Neutron and High Temperature Environments of Next Generation Reactors

Nikolaos Simos, Brookhaven National Laboratory (BNL)
Simerjeet Gill, BNL
T. Tsakalakos and K. Akdogan, Rutgers University
Funding: $990,000 (10/1/2012-8/31/2016)

<u>Description of project:</u> Amorphous Fe-based and ceramic coatings Al_2O_3+TiO_2 and Al_2O_3 are being explored for the enhancement and protection of next generation reactor materials expected to operate under severe neutron irradiation conditions and high temperatures. Nanostructured coatings deposited on steel and other metal alloys and consequently their ability to enhance the mechanical and physical properties of the coating-substrate structure under the combined extremes of neutron irradiation and high temperatures needs to be experimentally verified.

<u>Impact and value to reactor applications:</u> Oxidation and ductility loss resistance of Fe-based nanocoatings on steels can revolutionize the operating temperature regime of reactor materials provided that amorphous-to-crystalline transformations can be shifted upward. Understanding and controllability of phase diffusion in Al_2O_3+TiO_2 and Al_2O_3 coatings may have significant impact for materials to withstand extreme temperatures. The unique X-ray diffraction technique implemented at the synchrotron beam line for nano-structured coatings opened the way to an extremely capable approach for radiation damage characterization in next generation reactor materials.

<u>Recent results and highlights:</u> Amorphous Fe-based nanostructures under modest neutron irradiation (~10^{18} n/cm^2) showed remarkable resistance to ductility loss and oxidation/corrosion, behaviour attributed to the nature of their micro-structure. However, further experimental evidence and in particular at higher fluences were needed to qualify these nano-structures for nuclear reactor applications and nuclear steel protection. To that end, a series of irradiation experiments using spallation-produced fast neutrons combined with (a) macroscopic post-irradiation evaluation of ductility and oxidation assessment, (b) X-ray diffraction using high energy (200 keV) white beams and EDXRD techniques and (c) high temperature annealing combined with electron microscopy were conducted. These irradiation and post-irradiation studies at higher fluences indicated the onset of ductility loss above a fluence threshold but also the enhanced resistance to crystallization of the amorphous nano-structure (Fig. 1)

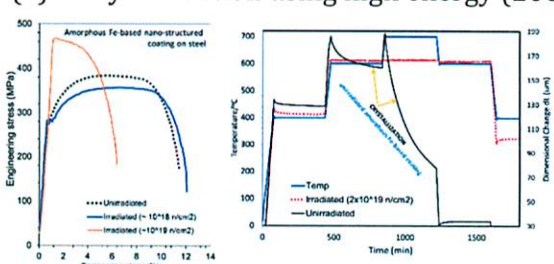

Figure 1 - Stress/Strain Curves measured on amorphous Fe-Based coatings after irradiation and the irradiation conditions.

Irradiation and high temperature studies showed that phase transformations and diffusion in the ceramic coatings Al_2O_3-TiO_2 and Al_2O_3 coatings on Ti-6Al-4V and steels influence the behaviour of the composite structure at high temperatures where a metastable, α-alumina layer tends to form leading to degradation and micro-cracking at the coating-substrate interface regardless of the environment (Fig.2). On-going irradiation experiments with 130 MeV protons on amorphous Fe-based nanocoated steel to higher fluence (~10^{20} p/cm^2) to be followed by X-ray diffraction at the new synchrotron at BNL is expected to shed more light on its potential applicability as nuclear material structure.

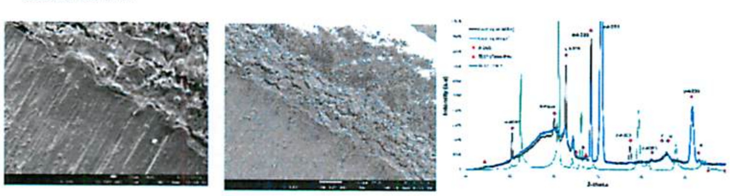

Figure 2- SEM images and X-ray diffraction patterns showing degradation of ceramic coatings under irradiation

2013 NEET-RM Open Award Research Summaries

In 2013, the NEET Crosscutting Reactor Materials program sought applications for advanced reactor materials characterization techniques and tools. Successful applications proposed advanced methods for sample preparation and new tools and techniques for examining and understanding material microstructures in a variety of conditions ranging from as-received to treated or irradiated.

Developing an extensive understanding of reactor material behavior in extreme environments is vital to the development of new materials for service in advanced nuclear reactors. This understanding is also needed for the extension of the operating lifetimes of the current fleet of nuclear reactors. Advanced characterization methods utilizing advanced tools and techniques, coupled with modeling simulation and advanced sample preparation tools will further the understanding of the effects of irradiation, temperature, pressure and corrosive environments on material microstructures and mechanical behavior. Modern sample fabrication tools could also allow for more efficient use of existing irradiated materials and enable fabrication of smaller specimens from previously examined materials.

Developing Microstructure-Property Correlation in Reactor Materials Using In Situ High-Energy X-Rays

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Jonathan D. Almer, Argonne National Laboratory
Yong Yang, University of Florida
Lizhen Tan, Oak Ridge National Laboratory
Funding: $1,000,000 (10/1/2013-9/30/2016)

Description of project:
The objective of this project is to advance our understanding of microstructure-property relationships in reactor materials through the use of high-energy synchrotron X-ray measurements during in situ thermal-mechanical loading. The gained knowledge is expected to enable accurate predictions of mechanical performance of nuclear reactor materials subjected to extreme environments, and to further facilitate design and development of new materials.

Impact and value to reactor applications:
Developing this new capability of in situ thermo-mechanical deformation, in concert with multiple high-energy x-ray modalities on activated specimens is essential to fundamentally understand irradiated microstructure, their interaction with grown-in defects and second-phase precipitates, and how nano- and micro-scale structures behave collectively to yield the observed (and desired) macroscopic behavior.

Recent results and highlights:

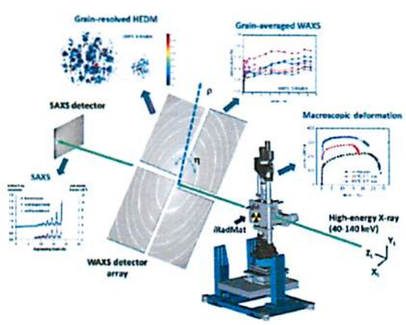

Fig. 1 A schematic of an *in situ* X-ray Radiated Materials (*i*RadMat) apparatus capable of applying thermo-mechanical loads on bulk-scale (mm-sized) neutron-irradiated specimens placed in the 1-ID- E beamline of the Advanced Photon Source. A suite of detectors are employed to understand the material behavior at multiple length scales under temperature and stress using grain-averaged wide-angle X-ray scattering (WAXS), grain-resolved high-energy diffraction microscopy (HEDM), and small-angle X-ray scattering (SAXS) techniques.

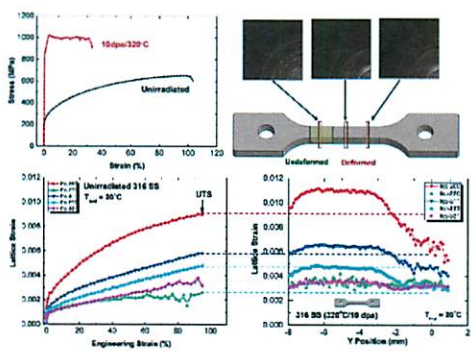

Fig. 2 In situ high-energy X-ray characterization during tensile deformation at 20°C (Top right) stress-strain curves recorded during *in situ* tensile tests of unirradiated 316 SS and neutron-irradiated 316 SS (10 dpa/320°C), (Top left) Diffraction patterns taken during deformation revealed Luders band propagation and strain-induced martensite transformation in irradiated 316 SS, (Bottom) comparison of lattice strain developed in unirradiated and neutron-irradiated 316 SS.

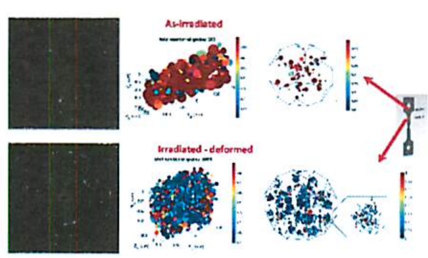

Fig. 3 Far-field HEDM gain mapping of a neutron-irradiated Fe-9Cr model alloy after an *in situ* tensile test showing substructure formation under tensile deformation.

Determining the Stress-Strain Response of Irradiated Metallic Materials via Spherical Nanoindentation

Nathan A. Mara, Los Alamos National Laboratory
Surya Kalidindi, Georgia Institute of Technology
Lance Kuhn, Hysitron, Inc.
Funding: $1,000,000 (10/1/2013-9/30/2016)

<u>Description of project:</u> Ion beam irradiation is a novel technique to impart large amounts of irradiation damage (several displacements per atom (dpa) or more), without activating the material, in a relatively short time spans of hours or days. This would require weeks or months to achieve in reactor conditions. However, the volume of irradiated material is limited by the beam energy to depths of microns or less, making the investigation of bulk mechanical properties very difficult. We utilize a novel approach for extracting indentation stress-strain curves from spherical nanoindentation datasets in order to study the material behavior at such length scales. Due to their ability of reliably quantifying many of the important aspects of the local mechanical constitutive behavior in the samples – such as the loading and unloading elastic moduli, the indentation yield points, as well as some of the post-yield characteristics from shallow indentation depths of ~50-100 nanometers– these indentation stress-strain curves are far more versatile than other nanomechanical test techniques which provide data for hardness and modulus values, or require extensive sample preparation.

<u>Impact and value to reactor applications:</u> This project can potentially revolutionize mechanical testing of irradiated nuclear materials. It will provide: 1.) Benchmarked measurement of the stress-strain response from small volumes of material for direct mechanical characterization of ion-irradiated materials (W, Zr, and stainless steels) with little surface preparation (only a polished surface prior to ion irradiation is needed). 2.) Since such small quantities of material (less than 0.5 mm^3) are needed for this technique, it can be expanded to indentation of radioactive materials outside of a hot cell environment. We have installed a nanoindenter in a radiological area that can test such materials.

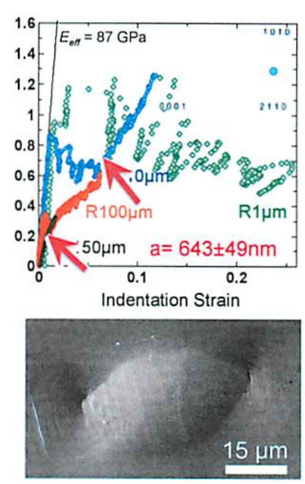

Figure 1. The indentation stress-strain responses of an off-c axis grain of Zr (top) and an SEM image of a representative indent. Our approach has revealed the onset of steady-state work hardening at a critical indent radius of ~700 nm, which may correspond to the onset of critical dislocation cell sizes or the onset of deformation twinning.

<u>Recent results and highlights:</u> In our recent paper in Scripta Materialia, we present the elastic and plastic anisotropy of Zirconium as a function of crystal orientation using our high-throughput approach (spherical nanoindentation). Zr is a highly anisotropic material, so this work underpins a larger effort to deconstruct the separate effects of mechanical anisotropy and ion irradiation. We demonstrated the differences in indentation moduli, yield strengths and post-elastic hardening rates over multiple grain orientations. These results are validated against bulk single crystal measurements, as well as data from cubic materials. By varying the indenter size, we demonstrated indentation size effects in Zr, including possible signatures of strain hardening due to twin formation in the nanoindentation stress–strain curves.

Predictive Characterization of Aging and Degradation of Reactor Materials in Extreme Environments

Jianmin Qu, Northwestern University
Rémi Dingreville and Khalid Hatta, Sandia National Laboratories
Funding: $999,812 (12/20/2013-12/19/2016)

Description of project: Understanding of reactor material behavior in extreme environments is vital not only to the development of new materials for the next generation of nuclear reactors, but also to the extension of the operating lifetimes of the current fleet of nuclear reactors. To this end, we propose a suite of unique experimental techniques, augmented by a mesoscale computational framework to understand the long-term effects of irradiation, temperature, pressure and corrosive environments on material microstructures and mechanical behavior. The experimental techniques and computational tools will be demonstrated on two distinctive types of reactor materials, namely, Zr alloys and high-chromium (9-12wt%) ferritic/martensitic steels. These materials are chosen as the test bed because they are the archetypes of high-performance reactor materials (cladding, wrappers, ducts, pressure vessel, piping, etc.).

Impact and value to reactor applications: The micro scale simulations tools and in situ transmission electron microscopy (TEM) techniques proposed here are not only unique and innovative, but also potentially groundbreaking in those new phenomena and new mechanisms will be discovered when the samples are subjected to multiple driving forces (heat, stress, ion radiation and He implantation).

Recent results and highlights: TEM samples for irradiation experiments have been prepared of nanocrystalline and coarse grained nickel and iron, chosen as model systems, as well as more complicated alloys including 316, 304 and HT9 steels and zircalloy. In-situ ion irradiation and gaseous implantation TEM experiments have demonstrated that the sequence of irradiation is essential in understanding defect evolution [1]. Annealing and high temperature implantation experiments have revealed different defect formation mechanisms and evolution pathways depending on thermal conditions. Key electron beam effects have been observed and mitigation plans developed [2-3]. The insight gained so far from experiments on model systems will be applied to relevant cladding materials.

Modeling efforts have also been exploring radiation damage evolution. A spatially resolved stochastic cluster dynamics (SRSCD) model has been used to successfully reproduce published experimental results of defect accumulation in neutron-irradiated iron. This information was then used as an input into a novel multi-scale crystal plasticity framework for estimating the change in initial yield strength of a metal due to the presence of both voids and dislocation loops [4]. Complimentary experiments are underway to characterize hardening as a function of dose and dose rate. Currently, modeling efforts are focused on studying damage accumulation in nanocrystalline metals both within grain boundaries and in the grain interior using SRSCD as well as defect clustering based on mean field rate theory. Successful ion irradiation and TEM orientation mapping of nanograined materials have been performed which will be used to validate models.

[1] B. Muntifering, S. J. Blair, C. G., A. Dunn, R. Dingreville, J. Qu, K. Hattar. Cavity Evolution at Grain Boundaries as a Function of Radiation Damage and Thermal Conditions in Nanocrystalline Nickel. Materials Research Letters. Vol. 4, pp. 96-103.

[2] B. Muntifering, R. Dingreville, K. Hattar and J. Qu, 2015. Electron Beam Effects during In-Situ Annealing of Self-Ion Irradiated Nanocrystalline Nickel. MRS Proceedings, 1809, mrss15-2136630 doi:10.1557/opl.2015.499.

[3] Muntifering, B., Dunn, A., Dingreville, R., Qu, J., and Hattar, K., 2016, "In-Situ TEM He+ Implantation and Thermal Aging of Nanocrystalline Fe," *Microscopy and Microanalysis*, Vol. 21(Suppl. 3), pp 113-114.

[4] A. Dunn, R. Dingreville, L. Capolungo, 2015, "Multi-scale simulation of radiation damage accumulation and subsequent hardening in neutron-irradiated α-Fe. Modelling and Simulation in Materials Science and Engineering, Vol. 24, pp. 015005.

Advanced 3D Characterization and Reconstruction of Reactor Materials

Assel Aitkaliyeva, PI, Idaho National Laboratory
Michael Tonks, Penn State University; David Field, Bradley Fromm, Washington State University;
Kumar Sridharan, University of Wisconsin
Funding: $1,000,000 (10/1/2013-9/30/2016)

Description of the project: The goal of the proposed research is to develop a critical line of inquiry that integrates advanced materials characterization techniques developed for reactor materials with state-of-the-art mesoscale modeling and simulation tools. The key objectives that will push the limits of present technology are:

1) Explore new sample preparation techniques for reactor materials using broad beam ion-etching methods to decrease sample preparation time, improve scan quality, increase scan size, and reduce radiation exposure/waste.

2) Implement an advanced characterization technique known as High-Resolution Electron Back Scatter Detection EBSD (HR-EBSD) to enable estimation of critical material properties like dislocation density and residual strain from reactor materials.

3) Develop necessary post-processing tools and procedures to utilize the HR-EBSD data obtained in Objective 2 for microstructure reconstruction into MARMOT and perform model validation based on the HR-EBSD data.

Impact and value to reactor applications: A coordinated effort to link advanced materials characterization methods and computational modeling approaches is critical to future success for understanding and predicting the behavior of reactor materials that operate at extreme conditions. The objectives described in this project push the limit of current experimental capabilities and innovate means to bring new characterization and simulation techniques to bear on both irradiated and unirradiated reactor materials. These advances will result in improved understanding of microstructure evolution and its impact on pertinent material properties. They will also aid in initializing, calibrating, and validating current phase field models, used to predict microstructure evolution at the mesoscale.

Recent results and highlights: The team conducted HR-EBSD work on irradiated HT-9 cladding from the Fast Flux Test Facility (FFTF) prepared using broad ion-etching technique (Figure 1) and has conducted serial sectioning of unirradiated HT-9 cladding. Obtained HT-9 serial sections were reconstructed into a digital microstructure for use in MARMOT. The serial sectioning of irradiated material will be conducted by the end of the fiscal year.

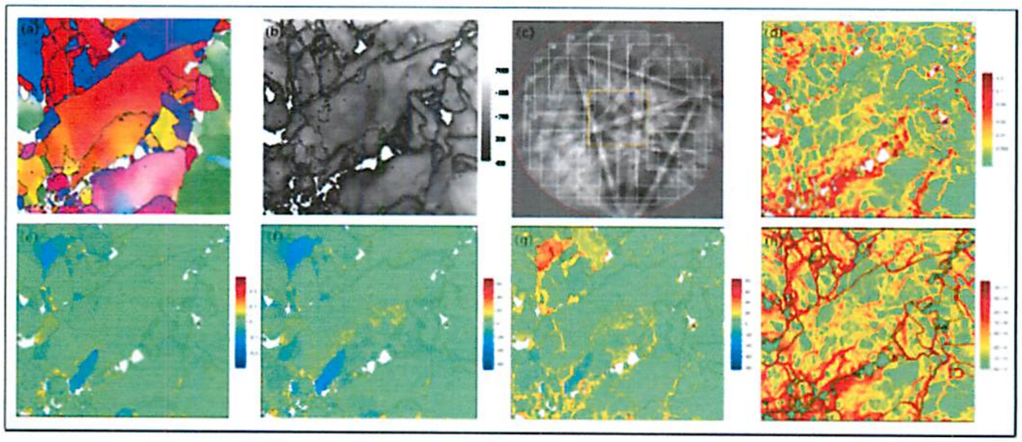

Figure 1. Results from HR-EBSD analysis of irradiated HT9 cladding: (a) inverse pole figure map, (b) image quality map, (c) reference electron backscatter pattern with 20 regions of interest overlaid onto pattern, (d) high resolution kernel average misorientation map, (e) ε_{22} strain map, (f) σ_{22} stress map, (g) Von Mises stress plot, (h) geometric necessary dislocation map.

Unraveling Dynamics of Radiation Damage Formation Via Pulsed-Ion-Beam Irradiation

S. O. Kucheyev, Lawrence Livermore National Laboratory
S. J. Shin, Lawrence Livermore National Laboratory
Funding: $1,000,000 (10/1/2013-9/30/2016)

Description of project: The primary cause of radiation damage to many nuclear materials is high-energy neutron recoils. Such recoils create collision cascades whose formation and thermalization are believed to be understood. In contrast, our understanding of the dynamic evolution of defects after cascade thermalization is still very limited. In this project, we are developing an experimental method to study radiation damage dynamics. Our method is based on pulsed-ion beam irradiation and aims to provide direct measurements of time constants, diffusion lengths, and activation energies of defect interaction processes in nuclear reactor materials.

Impact and value to reactor applications: This project offers an excellent opportunity to pioneer a new direction of advanced reactor materials characterization techniques. It could establish the pulsed ion beam method as the primary approach to study defect interaction dynamics in nuclear materials.

Recent results and highlights: We have been developing the pulsed-ion-beam methodology with the simplest and best studied material system, high-purity single-crystalline Si, and applying it to an inherently more complex nuclear ceramic material, SiC. In our method, the defect diffusion length is revealed by the dependence of damage on the active part of the beam duty cycle (t_{on}), while the time constant of defect interaction (τ) is measured directly by studying the dependence of damage on the passive part of the beam cycle (t_{off}), as shown in Figure 1. Our results have demonstrated the robustness of the pulsed-ion-beam method for studying radiation defect interaction dynamics. We have found that, for SiC, while dynamic defect annealing efficiency monotonically increases with increasing temperature, the temperature dependence of τ is complex and non-monotonic. For both Si and SiC, we have found that τ non-trivially depends on the density of collision cascades and on the level of pre-existing lattice disorder. These results demonstrate that the pulsed ion beam method provides a wealth of information about defect interaction dynamics.

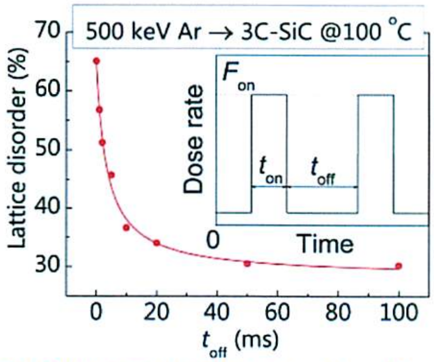

Figure 1. Bulk disorder in SiC as a function of the passive part of the beam duty cycle and all the other irradiation conditions kept constant.

Automated Synchrotron X-Ray Diffraction of Irradiated Materials

Lynne E. Ecker, Eric Dooryhee, and Sanjit Ghose, Brookhaven National Laboratory
G. Robert Odette, University of California Santa Barbara
Funding: $979,200 (10/1/2013-9/30/2016)

Description of project: This project developed an automated system with a robot to rapidly and safely acquire data on radioactive samples at the X-ray Powder Diffraction (XPD) beamline at the National Synchrotron Light Source-II (NSLS-II). Software to analyze the large amounts of data generated by the robot was also written and implemented. The system has been used to characterize neutron irradiated Reactor Pressure Vessel (RPV) steels, silicon carbide, nano-ferritic alloys, ferritic-martensitic steels and thermally aged FeCrAl, alloy 690 and low-fluence irradiated metal fuel alloys (UMo and UZr).

Impact and value to reactor applications: The robot in Figure 1 addresses a gap in nuclear materials research by providing greater access to synchrotron data to the entire nuclear community. It allows researchers to leverage large databases of irradiated materials that currently exist at facilities, such as test reactors. Obtaining additional structural data with state-of-the-art characterization techniques available at synchrotrons including high resolution x-ray diffraction (XRD), pair distribution function analysis and small angle x-ray scattering (SAXS) is an ideal way to maximize the investment in these expensive samples that require years of reactor exposure. This research has also provided a direct contribution to the understanding of the effects of radiation and thermal aging in structural materials through diffraction and scattering experiments. For reactor pressure vessel steels, the data generated can inform thermodynamic models *and* be included in existing material databases that can be used to model the life extension of existing light water reactors.

Figure 1 (a) The robot installed at XPD. (b) Radioactive sample holder with magnetic closure ring for rapid loading

Recent results and highlights: The robot for sample manipulation and software for batch data processing for high-throughput SAXS and XRD have been commissioned on active materials and are available for use.

XRD and SAXS were used to characterize the highly embrittling nm-scale Mn-Ni-Si precipitates that develop in the irradiated steels at high fluence. Application of the complementary techniques has, for the very first time, successfully identified the crystal structures of the nanoprecipitates, while also yielding self-consistent compositions, volume fractions and size distributions. Figure 2 shows the SAXS and XRD results for a selected sample. The nanoprecipitate phase selection in the refinement of the XRD is consistent with Thermocalc II database thermodynamic predictions. However, an alternative thermodynamic database predicts the formation of different intermetallic phases in some alloys. Thus our combined SAXS/APT and refined XRD data provide a basis to test and improve the parameterization of the thermodynamic models in a way that was not possible using either technique alone.

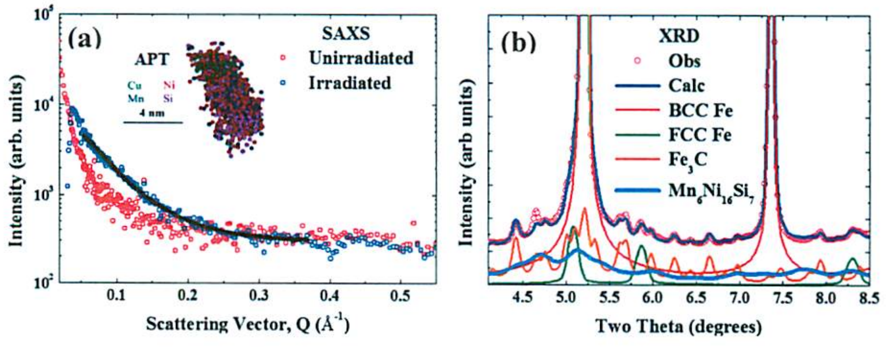

Figure 2. (a) SAXS and (b) XRD results for the irradiated RPV samples. The inset of (a) shows the Ni-Mn-Si precipitate as determined from APT. The phases are overlaid in (b) for reference.

Use of Micro- and Meso-scale Magnetic Characterization Methods to Study Degradation of Nuclear Reactor Structural Material

Pradeep Ramuhalli (PNNL), John S. McCloy (WSU-Pullman), Bradley Johnson (PNNL), Jon Suter (PNNL), Weilin Jiang (PNNL), Ke Xu (PNNL), Dan Schreiber (PNNL), Shenyang Hu (PNNL), Yulan Li (PNNL)

Funding: $919,661 (10/1/2013-9/30/2016)

Description of project: The goal of the project is to improve the fundamental understanding of reactor structural material performance under irradiation. This is being done by integrating magnetic signatures and microstructural characterization with phase-field models at the same length scale. The intent is to develop meso-scale models that will provide an interpretive understanding of the state of degradation in a material, based on meso-scale and bulk magnetic signatures. Predictive and interpretive computational models that correlate bulk non-destructive evaluation (NDE) data to the state of microstructural damage in a material will be developed and applied for condition monitoring of reactor structural materials.

Impact and value to reactor applications: The approach to understanding magnetic behavior in irradiated materials will provide insights into interpreting NDE measurements for quantifying damage in irradiated materials such as reactor pressure vessel (RPV) structural steel. Benefits will include the ability to ensure safe long-term operations of the existing U.S. nuclear fleet and the development and qualification of advanced materials for next generation nuclear power reactors.

Recent results and highlights: Initial studies have been completed with single and poly-crystalline Fe thin films to develop correlations between microstructural features and magnetic domain wall motion. Meso-scale phase field models of magnetic behavior have been created that allow simulation of domain wall movement with applied magnetic fields. These show wall-pinning behavior from defects such as those created by radiation damage. A focused ion beam (FIB) mill on a scanning electron microscope (SEM) has been used to create specific structures in the Fe thin films with boundaries that are analogous to edges in phase field models. Magnetic force microscopy (MFM) measurements show domain wall motion as a function of applied field. Phase field model simulations have been successfully compared to MFM measurements (Fig. 1).

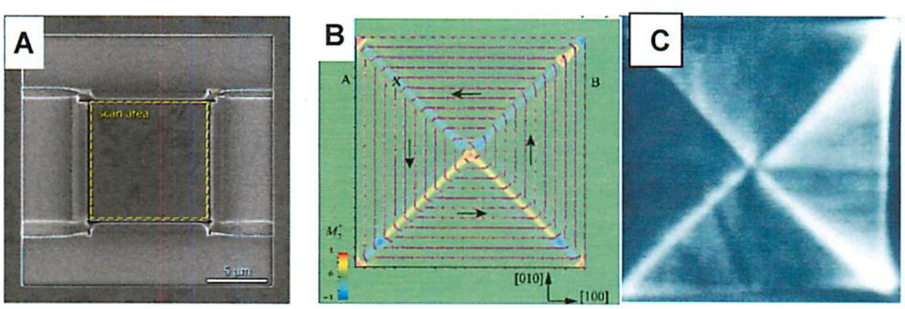

Fig. 1. SEM micrograph of FIB structure in Fe thin film (A); phase field model of domain wall structure (B); MFM measurement of domain wall structure.

The ability to correlate phase field model simulations to MFM measurements on Fe thin films provides evidence that this approach could be extended to more complex systems. Our goal is to expand this technique to develop tools that can eventually interpret magnetic NDE data, and estimate the state of damage in that material and safe operational limits.

Ongoing research is examining the changes in these properties due to irradiation damage, and correlating the findings from laboratory-scale experiments to simulation model predictions.

2014 NEET-RM Open Award Research Summaries

In 2014, the NEET Crosscutting Reactor Materials program sought applications the development of advanced joining techniques for materials for nuclear fission reactor applications. As advanced materials are developed to increase the energy efficiency, cost efficiency, safety and security of the operation of nuclear reactors, advanced joining techniques must also be developed. Advanced welding or joining techniques will overcome traditional component limitations as well as allow for the use of more advanced materials in nuclear reactor applications.

These advanced joining techniques must maintain or improve properties at the joint, such as strength, irradiation resistance, corrosion resistance, and creep. Innovative methods to control and understand residual stress, heat affected zones, and/or phase stability during joining are also of interest.

Functionally Gradient Transition Joint for Dissimilar Metal Welding using Plasma Arc Lamps

Evelina Vogli, Joshua Caris, Anupam Ghildyal MesoCoat Inc.
Zhili Feng, Xinghua Yu, Oak Ridge National Laboratory
Funding: $1,000,000 (10/1/2014-9/30/2017)

Description of project: The primary objective of this research is to develop functionally gradient transition joints between carbon steel and austenitic stainless steel for nuclear reactors. The research directly addresses the need to develop advanced joining techniques for materials for nuclear fission reactor applications. Mesocoat Inc. and Oak Ridge National Laboratory collaborate capitalizing on recent advances made by each organization in the field of dissimilar metal joining and application of high-energy density plasma arc lamp processing (PALP). Plasma arc lamps generate a high density infrared to build a gradient transition joint for dissimilar metal welding (DMW). The tasks include: transition joint composition design, transition joint fabrication microstructure characterization, residual stress measurement and mechanical testing. The results of this research will provide: 1) design approach and processing parameters for manufacturing gradient transition joints; 2) validation of controlled microstructure and composition in the gradient transition joint; and 3) validation of reduction of residual stress and improved stress corrosion cracking resistance in gradient transition joints.

Impact and value to reactor applications: To resolve DMW service failures in nuclear reactor applications, this work posits an innovative manufacturing technology using reflected light from high density plasma as a heat source to produce functional gradient transition joints, thus eliminating DMW during installation. This novel process is expected to improve transition area strength and phase stability while reducing residual stress and the tendency for stress corrosion cracking. In addition, PALP is cheaper and consumes much less power than conventional arc welding. This joining technique will be not only beneficial to DMWs in existing boiling water reactors and pressurized water reactors, but also beneficial to DMWs in high temperature gas cooled reactors and small modular reactors which are under development.

Recent results and highlights: In order to minimize elemental diffusion in the DMW during service conditions, a suitable composition gradient of the DMW has been designed and simulated under service conditions using thermodynamic (ThermoCalc) and kinetic (Dictra) softwares. The carbon content was graded from 0.2 wt.% (in SA508) to 0.04 wt.% (in SS 316L) and the simulation results showed that this composition remains both thermodynamically and kinetically stable at service temperatures (290 to 360 °C). The simulation of C diffusion using Dictra proved that C segregation can be suppressed by proper design of composition gradient in functionally graded joints. Multilayer fused clads with parameters based on experimental and simulation results have been demonstrated on weld joint composition relevant substrates. Demonstration/Validation coupons of Alloy 22 and Alloy 800H materials were produced on thick/narrow substrates 0.5" in width which simulate cladding on a pipe edge. These coupons are currently undergoing microstructural analysis, welding, and elevated temperature stress rupture testing, corresponding to process demonstration, at ORNL.

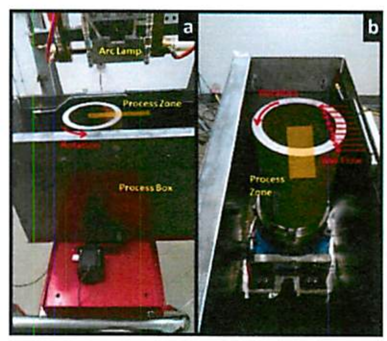

Figure 1. a-b) Photographs of the pipe rotation equipment and inert atmosphere process box. The zone of processing is highlighted in yellow.

Pipe rotation equipment and a corresponding inert atmosphere process box have been designed, built and validated for multilayer pipe edge cladding, Figure 1. Initial attempts at cladding a pipe edge showed a uniform clad and following a similar sample translation geometry.

Radiation Tolerance of Controlled Fusion Welds in High Temperature Oxidation Resistant FeCrAl Alloys for Enhanced Accident Tolerant Fuel Cladding Applications

Kevin G. Field, Maxim N. Gussev, Yukinori Yamamoto, Xunxiang Hu, and Richard Howard
Oak Ridge National Laboratory
Funding: $1,000,000 (10/1/2014-9/30/2017)

Description of project: Current and future nuclear power plant (NPP) designs call for materials to be able to withstand extreme environments including elevated temperatures, acute and chronic corrosive media attack, low to high dose radiation, and potential severe accidents like loss of coolant accidents. The FeCrAl alloy class is becoming an attractive material class due to their performance within these environments including high temperature oxidation and long-term aqueous corrosion. However, FeCrAl alloys may have critical degradation issues including severe radiation hardening and embrittlement and/or cracking during welding. Even more, these two degradation modes could be synergistic leading to exacerbated degradation while in service at a NPP. This project uses a varying range of alloy refinement techniques to mitigate such issues including alloy composition refinement and precipitate tailoring. Of primary interest is decreasing the susceptibility of the alloys to weld-induced cracking, typically associated with hydrogen embrittlement and the reduction or complete elimination of a Cr-rich nanometer-scaled precipitate phase found to occur in aged and neutron irradiated FeCrAl alloys. The viability of the deployed alloy refinement techniques is currently being evaluated in the welded state, irradiated state, and the welded-irradiated state. Irradiations are being completed using Oak Ridge National Laboratory's High Flux Isotope Reactor (HFIR) to provide specimens capable of nano and small scale mechanical testing. Assessments of specimens in the varying states include use of advanced and/or novel mechanical and microstructural characterization techniques.

Impact and value to reactor applications: Successful alloy refinement techniques that mitigate or reduce the propensity for degradation of FeCrAl weldments could have far reaching implications for nuclear power production. For example, FeCrAl alloys are currently under examination as an accident tolerant fuel cladding option. These designs require hermetic seals formed through fusion-welding techniques. A strong, radiation tolerant weldment could ultimately lead to a fuel-cladding technology with increased accident tolerance and hence better overall safety of the current U.S. reactor fleet. Furthermore, increasing the radiation tolerance could have implications for future designs such as advanced fast reactor technologies.

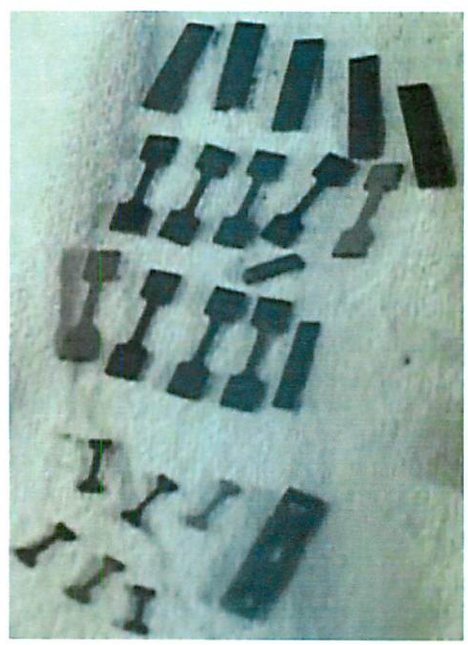

Fig. 1 Image of irradiated specimens being sorted in hot cell facility including SS-J type and SS-2E type tensile specimens and SiC thermometry.

Recent results and highlights: Several key advances have been made within the second year of funding, including: (1) development and refinement of digital image correlation algorithms for systematic determination of mechanical properties on objects with gradient properties, (2) full database on mechanical properties including engineering and true stress-strain curves for all alloys in the as-received and welded state, (3) extensive microstructural examination of all alloys in the as-received and welded state including electron-backscattered diffraction mapping, (4) the irradiation of selected alloys in the as-received and welded state up to nominally 1.8 dpa at target temperatures of 200°C, 330°C, and 550°C in HFIR, and (5) shipment of irradiated capsules to hot cell facility for sample removal, sorting, and identification, see Figure 1.

Extending the In-Service Life of Welded Assemblies Through Low-Energy Solid State Joining

Glenn Grant, Ken Ross, Nicole Overman, Mychailo Toloczko, Pacific Northwest National Laboratory
Gary Cannell, Fluor Corporation; Daniel Ingersoll, NuScale Power;
George Young, Knolls Atomic Power Laboratory; Scott Rose, MTI, Inc.;
Greg Fredrick, Electric Power Research Institute
Funding: $1,000,000 (10/1/2014-9/30/2017)

Description of Project: For a wide range of alloy systems being considered in the Advanced Reactor and the Small Modular Reactor programs, welded joints are the weak link in the life of a fabricated assembly. High heat input and melting associated with arc welding degrades the properties of the base metal resulting in poor performance in and around the welded joint. Solid state processes that neither melt, nor excessively heat the base material, provide potential to preserve the original microstructure and properties. This project will develop a variant of friction stir welding where energy input is held at very low levels. The project will develop the weld process, produce coupon weldments and quantify the improvement in residual stress, creep, creep/fatigue, stress corrosion cracking (SCC) susceptibility relative to arc welding for alloys of interest to current and advanced reactor designs and specifically those of interest to small modular reactor designs.

Impact and value to reactor applications:
Low thermal input has the potential to produce welded assemblies that could approach the performance of the parent materials and contribute to, rather than degrade, the benefits of high performance reactor materials. The project will focus on development and testing of low energy friction stir welds in two alloy classes, ferritic/martensitic and austenitic stainless steels, and one dissimilar ferritic to austenitic steel joint. Weld strength reduction factors can be as high as 50% in some ferritic alloy weldments and pressurized water SCC issues with welded austenitics can limit the service life of these materials. Low thermal input, solid-state welding may be able to limit the material degradation processes in these alloy classes. This project will develop weldments and the environmental performance data that will enable engineers to design reactor components around weldments with longer service life. This will help realize the benefits of increased energy efficiency, cost efficiency, safety and durability that new advanced reactor materials can provide.

Recent results and highlights:
Process parameters for defect free friction stir welds of stainless steel 304L and P-92 were developed. These experimental trials involved an exploration of process parameter space with the preliminary goal of weldments that were free of volumetric defect and passed cross-weld tension testing by failing in the parent material. The second goal of the reporting period was to develop algorithms to control weld temperature through a direct feedback mechanism during welding. The ability to hold tool temperature within +/-5°C of a commanded set point in both 304L and P-92 was developed and demonstrated. Fig. 1 shows a cross section and Fig. 2 shows process data from a portion of weld in 304L. Now, with the ability to control temperature throughout the length of the weld the project team is well positioned to optimize the process and drive energy input to a minimum. Environmental testing will begin in FY 16 to quantify performance benefits against established fusion welding practice.

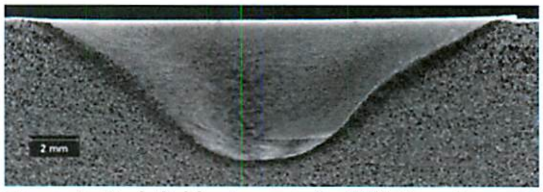

Figure 1: Dark field micrograph of friction stir weld in SS 304L

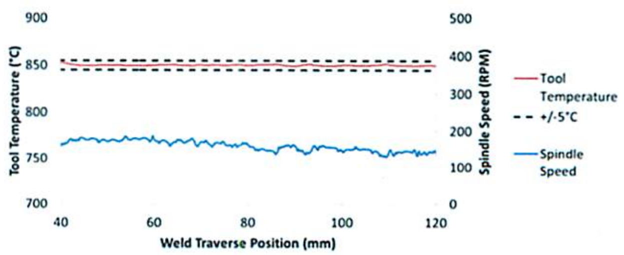

Figure 2: Temperature controlled friction stir weld in SS 304L

2015 NEET-RM Open Award Abstracts

In 2015, the NEET Crosscutting Reactor Materials program sought applications for advanced materials discovery and development similar to the call in FY12. Successful completion of awards will provide piping, structural, or clad materials that dramatically improve performance over traditional materials used in terrestrial and space reactors and in the nuclear fuel cycle.

Specific goals may include:

- Improvement in mechanical performance by a factor of 5-10 over traditional materials
- Increase in maximum operating temperature of greater than 200 C over an 80 year lifetime
- Increased radiation tolerance to beyond 300 dpa

Such performance would enable significantly improved safety, performance and reliability for future advanced reactor and fuel cycle designs. However, such improved performance cannot be at the expense of other properties or performance.

Applications were requested that describe innovative materials concepts, concept advantages, concept limitations, and key development needs. Successful applications described innovative materials that offer the potential for revolutionary gains in reactor and fuel cycle performance. Materials that could be applied to multiple reactor designs, components, and concepts were given preference over materials restricted to a single reactor concept, component, or coolant.

Radiation Tolerance and Mechanical Properties of Nanostructured Ceramic/Metal Composites

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Michael Demkowicz, Lin Shao, Texas A&M University
Don A. Lacca, Oklahoma State University
Funding: $994,292 (10/1/2015 – 9/30/2018)

Description of project: The objective of this proposal will be to explore the development of advanced metal/ceramic composites with greatly improved radiation tolerance, stability above 500 °C, and improved mechanical performance combining the good properties of glasses (high strength and elastic limit, corrosion resistance) with those of crystals (high toughness, strain hardening). The ceramic component of the composite will consist of a high crystallization temperature amorphous material composed of SiOC, while the metal component will be Fe or Fe(Cr), chosen as a model material for steel. We hypothesize that the combination of the composite constituents as well as the interfaces between them will provide significantly enhanced radiation tolerance, similar to or superior to those observed in metallic nanolayered structures, but in a more engineering-relevant material system.

Impact and value to reactor applications: The need to develop advanced cladding that does not react to form hydrogen is urgent considering past accidents at Fukushima. The project will aim to develop super tough and ultra-high temperature resistant materials that are in critical need for nuclear applications under extreme conditions where in-core materials have to withstand neutron damage and high temperature. The potential impact will be the development of a new class of ceramic/metal composites that can be adapted for engineering applications, resulting in dramatically improved materials performance for advanced reactors.

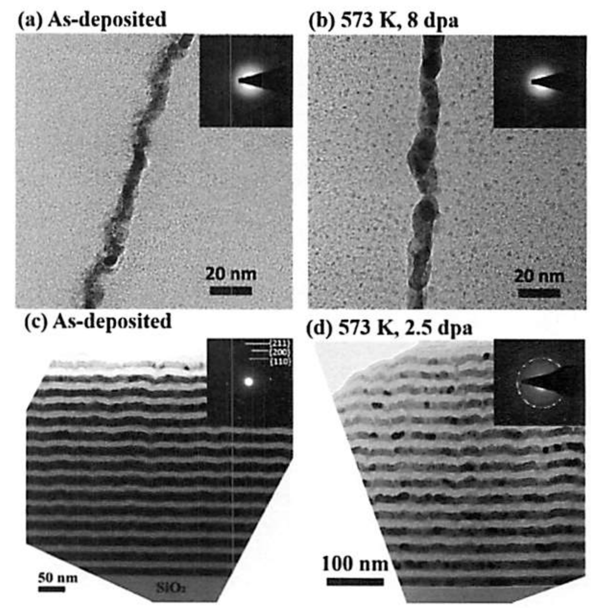

Fig. 1. Typical TEM micrographs of marker specimen (SiOC/Fe/SiOC, 200/7/200 nm) (a) before, and (b) after 8 dpa Kr irradiation at 573 K. Similar results are observed for Fe/SiOC multilayers (c) before and (d) after irradiation. The inset is the corresponding SAD pattern of each micrograph.

Recent results and highlights: Studies have shown that the Fe/SiOC nanocomposite is thermodynamically stable and radiation tolerant at elevated temperatures. Fig. 1 shows the typical TEM micrographs of marker specimen (SiOC/Fe/SiOC, 200/7/200 nm) (a) and Fe/SiOC multilayers (c) before and after 573 K Kr irradiation with (b) 8 dpa and (d) 2.5 dpa. Although irradiation tend to mix Fe and SiOC constituents due to ballistic effect, the Fe/SiOC interfaces are all clearly observed after the 573 K irradiations. In addition, the corresponding select area diffraction (SAD) pattern demonstrates no irradiation-induced mixing or secondary phase formation between the Fe layers and the amorphous SiOC layers. The Fe/SiOC results suggest that the heat of mixing for this system is positive and a demixing process takes place during irradiation at elevated temperatures. The findings deliver a new understanding of how SiOC/Fe amorphous/crystal interfaces effect, influence, and enhance irradiation tolerance. This information can be used to aid in the potential design of amorphous-ceramic/metal composites for service in extreme irradiation environments with significantly improved safety, performance and reliability.

Nanoprecipitate-Strengthened Advanced Ferritic Steels for Nuclear Reactor Application

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Kumar Sridharan and Beata Tyburska-Püeschel, University of Wisconsin-Madison
Funding: $1,000,000 (10/1/2015–9/30/2018)

Description of project: The project seeks to develop novel precipitates with a strong propensity for amorphization in ferritic steels, with the aid of modern computational microstructural modeling tools, favoring improved radiation resistance and enhanced high temperature creep resistance.

Impact and value to reactor applications: Development of inherently radiation resistant ferritic steels with enhanced creep resistance, through conventional economic steelmaking techniques, represents a significant step towards development of components for high temperature, high dose reactors. Advancement on understanding the relationship between the irradiation resistance and amorphization of intermetallic precipitates can provide guidance on the design of future irradiation resistant metal/amorphous composite materials

Recent results and highlights:

- Nickel has been identified to have the potential to suppress the liquid surface of Fe–Cr–Zr to lower temperature, therefore, leading to high amorphous phase forming ability in Fe–Cr–Zr–Ni alloys. (Fig. 1)

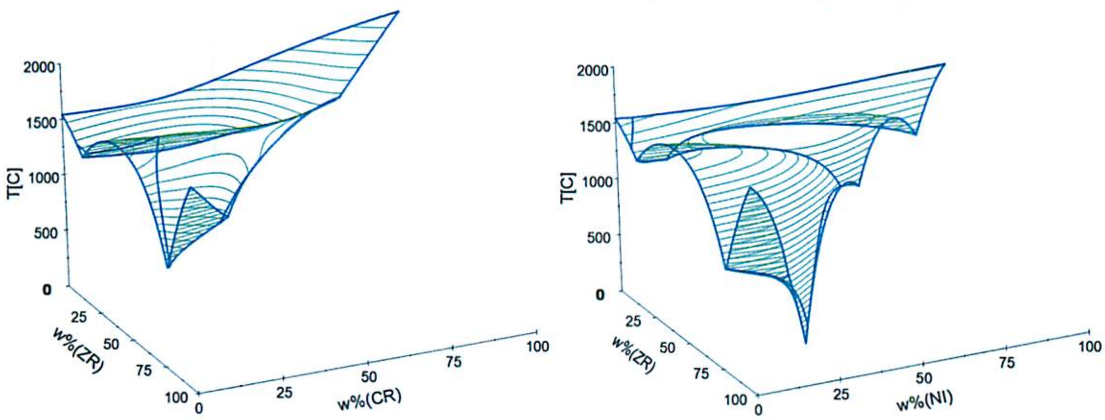

Figure 1. The Liquidus surface of Fe–Cr–Zr (left) and Fe–Ni–Zr (right), calculated from the currently developed thermodynamic database for Zr-containing ferritic stainless steels.

- Four Fe–Cr–Zr–Ni model alloys have been designed to understand the alloying effect of Ni on the phase stability. Ni and Zr were found to be greatly presented in a variety of Laves particles. (Fig.2)

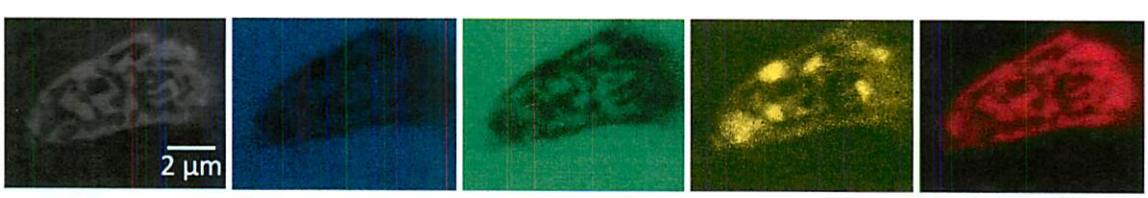

Figure 2. From left to right showing the BSE image of Laves particles in the BCC matrix of Fe–Cr–Ni–Zr model alloys and the EDS maps of Fe, Cr, Ni, and Zr, respectively