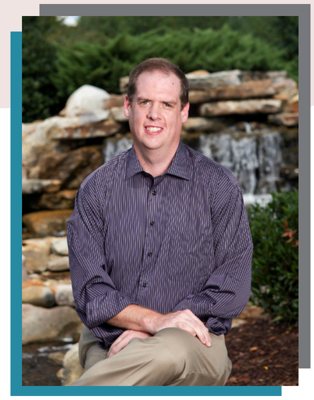
Nuclear Energy Enabling Technologies

NEET Reactor Materials

News for the Reactor Materials Crosscut *May 2015*

Materials science is vital to nuclear power research



Feature Updates: May 2015

Access the following NEET updates at http://energy.gov/ne/nuclear-energy-enabling-technologies/reactor-materials:

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

2. Nanocrystalline SiC and Ti_3SiC_2 Alloys for Reactor Materials

3. Nanoscale Stable Precipitation-Strengthened Steels for Nuclear Reactor Applications

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

5. Radiation-Induced Ductility Enhancement in Amorphous Iron-Based and Al_2O_3-TiO_2 Nanostructured Coatings in Fast Neutron and High Temperature Environments of Next Generation Reactors

Nuclear energy represents the single largest non-carbon emitting base load source of electricity in the United States, accounting for nearly 20% of the electricity generated and over 60% of low-carbon emitting energy production. Increasing demand for clean energy domestically and globally, combined with research to enhance nuclear power safety and cost-effectiveness, will keep nuclear in the energy mix for the near future.

Materials science plays a pivotal role in extending the life of existing nuclear reactors; in deploying new, modern light water reactors, advanced reactors with non-water coolants, and small modular reactors; and in storing, recycling, and disposing of used nuclear fuel. Understanding and overcoming material degradation in an extreme environment is essential for safe, efficient operation. New advanced materials may make plant construction more economical.

Materials research is featured in all of the major research thrusts within the US Department of Energy Office of Nuclear Energy (DOE-NE) research portfolio. The Nuclear Energy Enabling Technologies–Reactor Materials Crosscut (NEET-RM) provides support and coordination among these programs by enabling development of innovative, revolutionary materials, providing broad-based modern materials science support to research within DOE-NE programs, and coordinating with DOE-NE research and development (R&D) programs. These efforts afford an opportunity for response coordination and research sharing and collaboration nationally and internationally. There are ongoing needs for new research tools, improved infrastructure, and coordination to improve research efficiency.

Today, the NEET-RM Crosscut is pursuing all of these areas actively via a competitive open proposal process. Three rounds of open competition for 3-year awards have been completed.

- In 2012, nine awards were given for advanced materials development concepts, ranging from iron-based steels to radiation-tolerant cable insulation.
- The 2013 NEET-RM competition focused on the development of advanced characterization techniques, such as analytical measurement and model development to interpret results. Seven awards were granted, ranging from synchrotron diffraction techniques to spherical nano-indentation.
- The 2014 competition focused on the development of advanced joining methods for advanced materials.

We are working to increase awareness and communication in key nuclear materials programs, share results from the ongoing project, and improve coordination with other nuclear materials efforts through this document and others. In the box at left, we are sharing a list of several summary fact sheets of key activities within the major DOE-NE programs and open competition projects. As the projects develop and new milestones are met, we will share additional information on research centers, university awards, and key reports. We hope you'll visit http://energy.gov/ne/nuclear-energy-enabling-technologies/reactor-materials to learn more.

Thank you.

Jeremy Busby

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Highlights from key DOE-NE programs

Light-Water Reactor Sustainability

Materials degradation affects reactor reliability, availability, and, potentially, safety. Routine surveillance and component replacement can mitigate these factors, although failures still occur. With reactor life extensions to 60+ years, many components must tolerate the reactor environment for longer than intended, which may increase susceptibility and introduce degradation. While all components (except perhaps the reactor vessel) can be replaced, doing so may not be economically favorable. Therefore, understanding, controlling, and mitigating degradation processes are key priorities for reactor operation, power uprate considerations, and life extensions. The DOE Light Water Reactor Sustainability Program supports research in materials science and technology to understand and predict long-term degradation behaviors.

Key activities will support the second license renewal. Irradiation effects on concrete are one potential knowledge gap for extended service. Post-irradiation examination of aggregate specimens irradiated in the High Flux Isotope Reactor will be completed later in 2015 and will provide key insights into degradation potential at high fluences. High-priority cable insulation studies also will continue, and harvesting relevant materials from fleet reactors will be emphasized. Also, a key capsule irradiated in the Advanced Test Reactor at Idaho National Laboratory with 1,600 specimens of model and commercial reactor pressure steels will be opened and examined. Analysis from this major Nuclear Scientific User Facility experiment will be critical for understanding high fluence effects on steels. Harvesting of reactor pressure vessel steels from the Zion Nuclear Generating Station should be completed later in 2015.

High Temperature Materials

Nuclear component design rules are contained in Section III of the American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code. The new Division 5 of Section III covers design needs for high temperature structural components for both gas- and liquid-cooled (sodium, salt, and lead) systems. The Advanced Reactor Technologies (ART) high temperature materials program has had several notable accomplishments related to this section.

A new Appendix HBB-Y, "Guidelines for Design Data Needs for New Materials," is being incorporated in the 2015 edition. It provides guidelines for data needed for new materials in support of design rules for elevated temperature nuclear applications with significant creep effects. The appendix helps define the information needed to incorporate new materials for nuclear design and discusses time dependent properties, creep-fatigue, weldments, and aging. These guidelines are important because only five materials are allowed for high temperature nuclear design, and a new material has not been added for several decades.

One of the ART materials program's long-standing goals to support high temperature gas-cooled reactors is to qualify the Ni-Cr-Co-Mo material Alloy 617 according to ASME Code for use in a steam generator or heat exchanger up to 950°C. A draft Code Case to allow design using Alloy 617 in Section III Division 5 up to 423°C is under consideration by relevant ASME committees. The follow-on high temperature Code Case is underway, and a complete draft is expected by the end of this fiscal year.

Graphite R&D Program

The ART Graphite R&D program is addressing the scientific and engineering issue for using graphite or graphitic components within a nuclear application. Graphite comprises 90–95% of the core in a high-temperature reactor and is critical in protecting the fuel and providing the core structure. Expected operating conditions are very high temperatures, minimal coolant reactivity, high structural strength, and moderate irradiation stability (useful lifetime of 10–15 displacements per atom).

The graphite program is researching unirradiated and irradiation material property performance, with a focus on irradiation creep behavior as the life-limiting mechanism for graphitic components. Irradiation-induced changes in the thermal conductivity, dimensional changes, and mechanical strength must be understood fundamentally to predict component behavior under various conditions (temperature, dose, stress levels, corrosion, accident conditions, etc.). Irradiation damage mechanisms, underlying mechanisms inducing irradiation creep behavior, and thermal changes caused by irradiation damage are important to research. Degradation issues related to chronic oxidation and component reaction during accidents are important for answering safety issues required for reactor design licensing.

Advanced Reactor Technologies

The ART Fast Reactor Structural Program supports the design and licensing of sodium fast reactor (SFR), a leading candidate for possible missions, including used fuel recycling and power generation. Advanced structural material with improved performance is one of the key enabling technologies for capital cost reduction and economic return improvement important incentives for the private sector to invest in large-scale, industrial SFR. Recent advanced materials R&D s has led to down-selection of an advanced ferritic/martensitic alloy and an advanced austenitic stainless steel that showed enhanced performance over current generation materials.

Intermediate testing determined the performance enhancement observed from small lots, and accelerated test conditions during down-selection were sustainable for longer-term tests and larger lot sizes. Intermediate term testing for austenitic stainless steel led to a recommendation to qualify Alloy 709 for inclusion in the nuclear section of the ASME Boiler and Pressure Vessel Code as a less-expensive replacement material for 316 stainless steel. A detailed testing and execution plan for Alloy 709 is being developed to support design and licensing of the next generation SFR.

NEET Nuclear Energy Enabling Technologies
Reactor Materials
News for the Reactor Materials Crosscut
May 2015

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

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Contributors: Erika Benda, Yiren Chen, Peter Kenesei, Ali Mashayekhi, Jun-Sang Park, Hemant Sharma, Xuan Zhang (Argonne National Laboratory), Chi Xu (University of Florida)

The objective of this project is to demonstrate the applications of high-energy synchrotron x-ray measurements with in situ thermal-mechanical loading in understanding the microstructure–property relationship. 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.

Argonne National Laboratory researchers developed an in situ radiated materials straining/annealing apparatus (iRadMat) designed to interface with MTS servo-hydraulic materials testing system equipped at beam line 1-ID at the Advanced Photon Source (APS), as illustrated in Fig. 1. The iRadMat provides a protected environment for high temperature mechanical testing and, at the same time, serves as a shielded containment of an irradiated specimen that permits an activated specimen to be characterized at a public beam line. The high temperature vacuum furnace equipped with x-ray windows can reach the maximum temperature of 1,200°C under vacuum of 10^{-5} Torr.

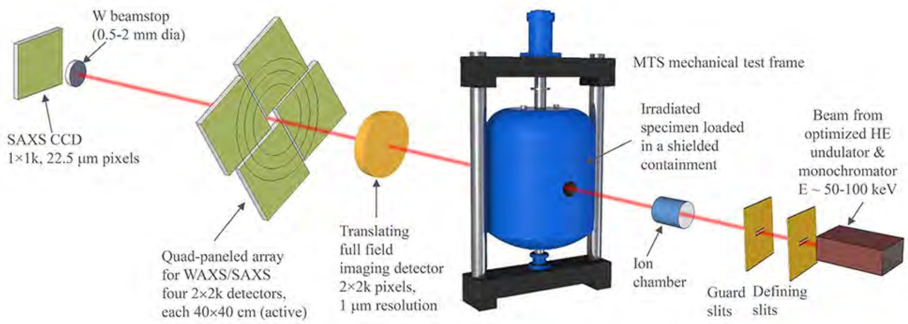

Fig. 1. Beam line layout for in situ characterization of an activated specimen using multiple probes at APS

High-energy x-rays have unique advantages for such a chamber environment not only because of their high penetrating power but also because the small Bragg angles at high energies so that several diffraction peaks can be collected. The specimen chamber is equipped with 5 mm thick tungsten plates attached to all six internal walls and tungsten shield shutters covering four windows 90° apart on the four sides of the chamber. This integrated interior radiation shielding provides comprehensive protection from radiation exposure and is suf ficient for handling low-activity specimens (with the dose rate < 100 mR/h at a 30 cm distance) with minimal radiological control at APS beam lines. To permit in situ three-dimensional (3D) characterization, an in-grip rotation mechanism was designed to allow the rotation of a specimen ±180° under a tensile load up to 2.5 kN. With the iRadMat apparatus, an activated specimen can be characterized in situ under thermal–mechanical loading, with combined high-energy x-ray techniques of wide-angle x-ray scattering (WAXS), small-angle x-ray scattering (SAXS), 3D far-field high energy diffraction microscopy (FF-HEDM), and tomography.

The ability to use multiple probes simultaneously is a unique advantage of synchrotron x-ray measurements. Structures ranging from the atomic scale to the macroscale can be interrogated concurrently, rather than sequentially as in a traditional experiment. WAXS is a powerful tool in characterization of phases, lattice strains, defect populations, and crystallographic texture, whereas SAXS is useful for characterization of the structural properties of a material on the nanometer scale. Tomography can reveal initiation of microcracks and voids as well as phase morphology in 3D, and HEDM is a new non-destructive technique developed in recent years for 3D structural characterization of individual grains in a polycrystalline bulk specimen. Far-field HEDM provides crystallographic orientation, center-of-mass (COM), grain size, stress-state, and defects of individual grains in the polycrystalline aggregate.

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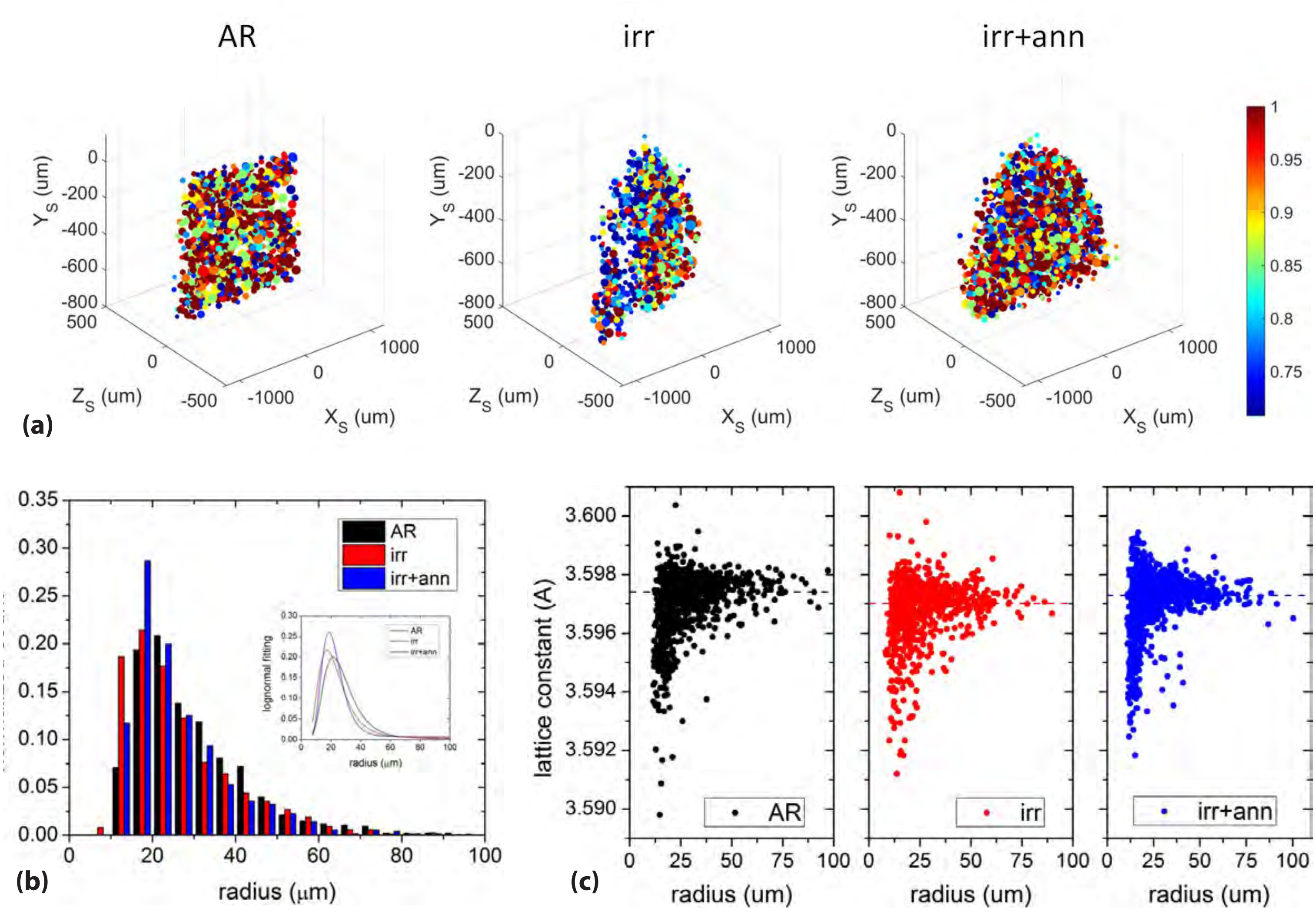

Fig. 2. (a) Center of mass map, (b) grain size distribution, (c) and lattice constant as a function of grain size for as-received, neutron irradiated (3 dpa/500°C), irradiated and annealed (3 dpa/500°C + 1 h/600°C) HT-UPS steel specimens.

Figure 2 shows the results from the first study using the FF-HEDM technique to characterize irradiated microstructure. Three specimens of a high-temperature ultrafine-precipitate-strengthened (HT-UPS) austenitic stainless steel were examined: neutron-irradiated (3 dpa/500°C), irradiated and post-irradiation annealed (3 dpa/500°C irradiated + 1 h/600°C annealed), and non-irradiated control specimens. About 1,000 grains in each specimen were measured by HEDM. The COM maps are shown in Fig. 2(a) and the grain size distributions are shown in Fig. 2(b) for these three HT-UPS specimens. The grain size distribution measured by FF-HEDM of the non-irradiated control specimen showed an excellent agreement with that measured by optical microscopy. The lattice constants also were measured for each individual grain and are plotted as a function of corresponding grain size in Fig. 2(c).

Ff-HEDM results reveal that irradiation changed the state of the material at the crystal length scale, and the irradiation-modified microstructure can be recovered to a certain extent by post-irradiation annealing. FF-HEDM proves to be an effective tool in understanding the behavior of heterogeneous materials with complex microstructures such as nuclear reactor materials.

Capability of in situ straining/annealing with multiple probes of high-energy x-rays for activated specimens being developed in this project is expected to facilitate the fundamental understanding of (1) irradiated microstructure and interaction with grown-in defects and second-phase precipitates, and (2) how nano- and microscale structures behave collectively to yield the observed macroscopic behavior.

NEET Nuclear Energy Enabling Technologies Reactor Materials

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May 2015

Nanocrystalline SiC and Ti_3SiC_2 Alloys for Reactor Materials

C. H. Henager, Jr., Pacific Northwest National Laboratory

This research project seeks to develop a high-temperature structural material based on nanocrystalline silicon carbide (SiC) alloys that can be used in advanced reactors as an accident tolerant replacement for zircaloy cladding, as an improved SiC cladding for tristructural-isotropic (TRISO) fuel, and for certain other in-core applications that require higher temperature properties. These materials are expected to have temperature utility up to 1,700 K (~1,425°C). The focus will be on the synthesis of a carbon nanotube (CNT)-reinforced SiC/Ti_3SiC_2 nanocomposite using textured CNT mats to achieve improved mechanical and thermal properties. Microscopy evaluation of the composite material will determine CNT/SiC adhesion, strength, and toughness; results will be used with mechanical models to help design a three-dimensional texture in the CNT mats for optimal mechanical properties. Thermal diffusivity measurements will be used to determine thermal conductivities, and ion implantation will be used to explore radiation damage and fission product (surrogate) transport in these new materials.

Thus far, researchers have accomplished the synthesis of SiC-based alloys with the general structure of SiC/Ti_3SiC_2 dual phase composites formed by simultaneous polycarbosilane pyrolysis and Si + TiC displacement reactions. An example is shown in Fig. 1. Liquid polycarbosilane polymers filled with a combination of SiC, silicon, and TiC-particles are pyrolyzed at 1,800°C at 20 MPa to produce a crystalline SiC + Ti_3SiC_2 dual phase ceramic. The ternary Ti_3SiC_2 phase is formed in conjunction with SiC from a displacement reaction between the silicon and TiC particles during the polymer pyrolysis. Variants of this process also can be used to produce materials containing graphene or graphitic nanodomains. Researchers believe these are the first such dual phase composites made by combining these two methods.

Fig. 2 shows scanning electron microscopy (SEM) images of one of the more dense composites processed at 1,800°C and 20 MPa with the two phases identified. This data suggests that processing conditions are not producing fully dense materials as evidenced by the pores in the SiC phase. Current plans include refurbishing a hot-press system to obtain temperatures up to 2,300°C and slightly higher pressures in a graphite die system.

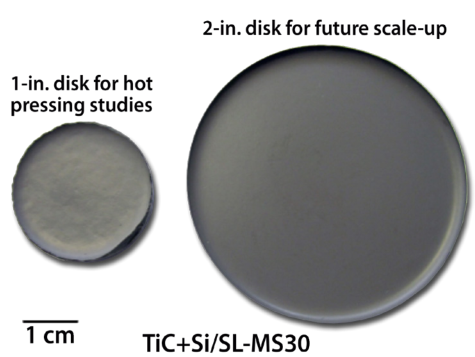

Fig. 1. A gel cast mixture of polycarbosilane polymer filled with 30 vol % SiC-particles and then filled with 40 vol % Si + TiC powders and cured at 60°C. This creates a smooth solid green disk that can be handled and machined. (Note: SL-MS30 is a Starfire Systems [Schenectady, N.Y.] designation for a 30 vol % SiC particulate-filled polycarbosilane.)

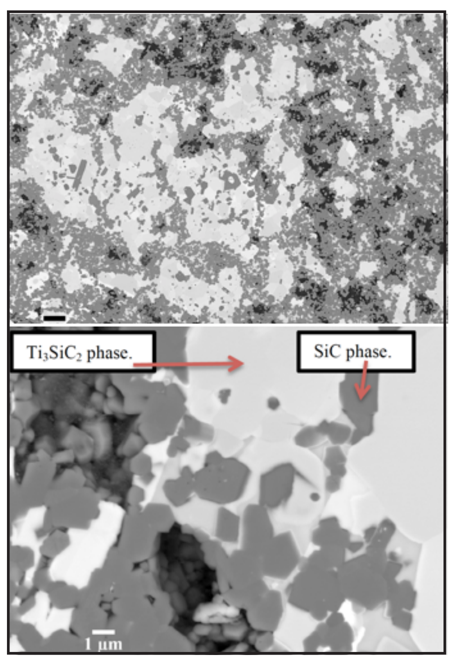

Fig. 2. SEM images of a SiC-alloy hot pressed at 1,800°C (2,073 K) and 20 MPa pressure from SiC-filled polymer plus Si + TiC powders at 63% powder loading. The white phase is the Ti_3SiC_2 phase, and the darker gray phase is SiC. The pores are contained entirely within the SiC phase.

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Data for thermal conductivity and fracture toughness indicate that properties of these composites agree with what others have measured for similar materials synthesized by different methods. This is a positive result indicating that the polymer pyrolysis methods likely can be used to combine in situ displacement reactions with CNT mats to make tough composites. Figure 3 shows thermal conductivity data from typical dual phase alloys such as those imaged in Fig. 2. Future goals are to densify the composite fully to achieve higher thermal conductivities and to add carbon nanotubes for both thermal conductivity and toughness improvements.

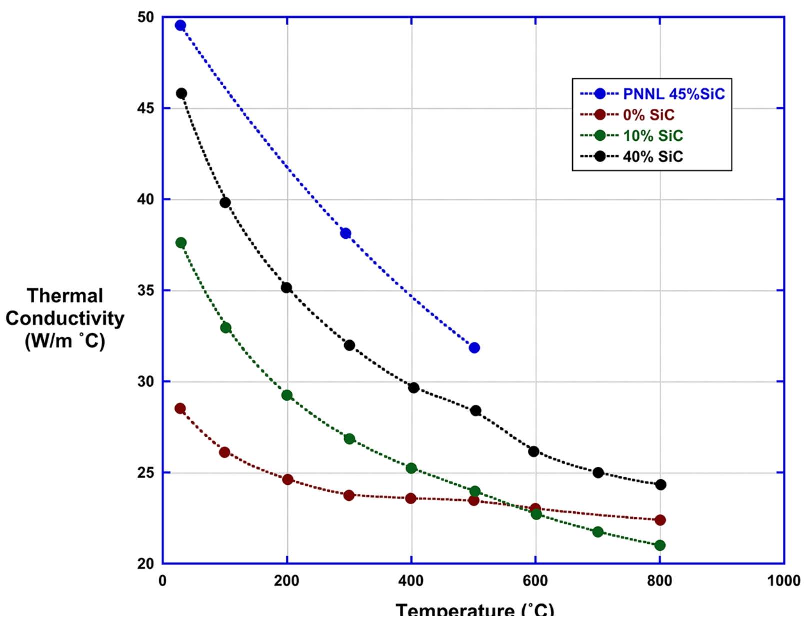

Fig. 3. PNNL laser flash thermal diffusivity is plotted as an average of thermal conductivity showing agreement with other measured data. (Source: Jianfeng, Z., W. Ting, W. Lianjun, J. Wan, and C. Lidong. 2008. Compos. Sci. Technol. 68[2], 499.)

Because SiC is used in TRISO fuels to clad the fuels and to block fission product transport, the new dual phase alloys were studied to determine their transport properties. Ion implantation with silver, gold, and cesium ions was performed on several materials of interest for this project, including a dense dual phase alloy made previously, a chemical vapor deposition (CVD)-SiC, and a nominally single-phase Ti_3SiC_2 material. The results from this study indicate that diffusion of silver, gold, and cesium in MAX phase Ti_3SiC_2 occurs at relatively low temperatures (773–973 K), compared with diffusion of species in SiC. Although the reasons for the fast diffusion of these species in this ternary phase have not been established, one hypothesis is that the layered crystal structure may play a significant role in determining impurity diffusion rates in MAX phases like Ti_3SiC_2. Because these temperatures are relatively low for advanced high-temperature reactor designs, this study suggests some caution in using Ti_3SiC_2 as a fuel cladding material for advanced nuclear reactors operating at very high temperatures. Further studies of impurity transport in the related materials may be warranted based on these results.

Finally, one of this project's primary tasks has been to study the additions of carbon nanotubes (CNTs) to these dual phase composites to increase both thermal conductivity and fracture toughness. Although it has not actually solved the problem, this work has shown conclusively that CNT additions as dispersed reinforcement will not be able to achieve the desired results. This has led to a focus on the addition of CNTs as a two-dimensional reinforcement "mat," which has yet to be demonstrated. However, work on dispersed CNTs has set the stage for going forward with the CNT mat idea.

Impact and value to reactor applications: The value of this material for reactor applications is that it potentially can be made dense with improved fracture toughness and much higher thermal conductivity than SiC/SiC composites. Polymer processing allows for use of near net shape processing. One caveat for the use of Ti3SiC2, and potentially for other such MAX phases, in reactor applications is that preliminary data taken for this project suggest that fission produce diffusion appears to be much higher than SiC. This may have serious implications for the use of this material for fuel cladding.

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Nanoscale Stable Precipitation-Strengthened Steels for Nuclear Reactor Applications

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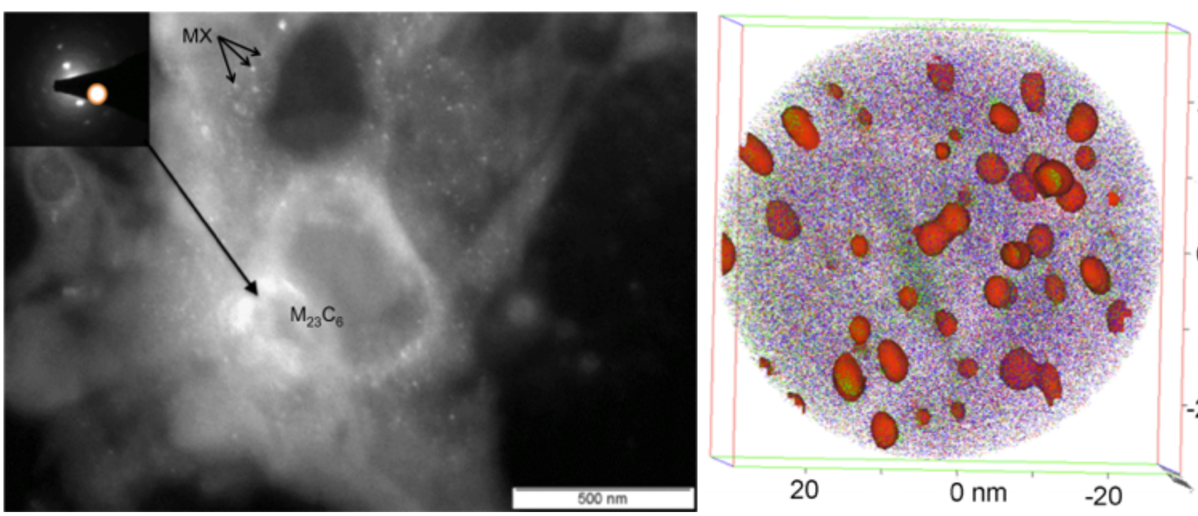
In this Nuclear Energy Enabling Technologies Reactor Materials project, we are generating a toolbox of modern alloying and processing strategies to enable the design and engineering of steel microstructures for Gen IV reactor concepts. These steels are designed to incorporate microstructural characteristics similar to mechanically alloyed oxide dispersion strengthened (ODS) steels. Two opportunities have been identified for performance improvements to conventionally manufacturable steels for reactor applications:

- Advanced high-chromium martensitic or ferritic-martensitic (AHCr-FM) steels, similar to those historically used in reactor service, that have been optimized in composition and processing for high temperature and creep resistance
- Novel tailored-precipitate ferritic (TPF) steels with specific nanoscale precipitates for strength, irradiation resistance, and thermal stability, with considerations for weldability

The steels we design are conventionally manufacturable. In particular, we focus on nickel aluminide (NiAl), niobium nitrogen/vanadium nitrogen (NbN/VN), and/or niobium carbide/vanadium carbide (NbC/VC) precipitation strengthened steels, processing-path development, thermal/mechanical and irradiation experiments to evaluate performance, and advanced characterization to assess microstructure-performance relationships for reactor steels.



(a) (b)

Fig. 1. (a) An AHCr-FM steel displaying both desirable nanoscale MX-type precipitates dispersed in the matrix and larger, undesirable M23C6-type precipitates, shown in a dark field Transmission Electron Microscope image. (b) A TPF alloy showing desirable nanoscale NiAl precipitate dispersion designed for irradiation resistance, displayed in an atom probe tomograph.

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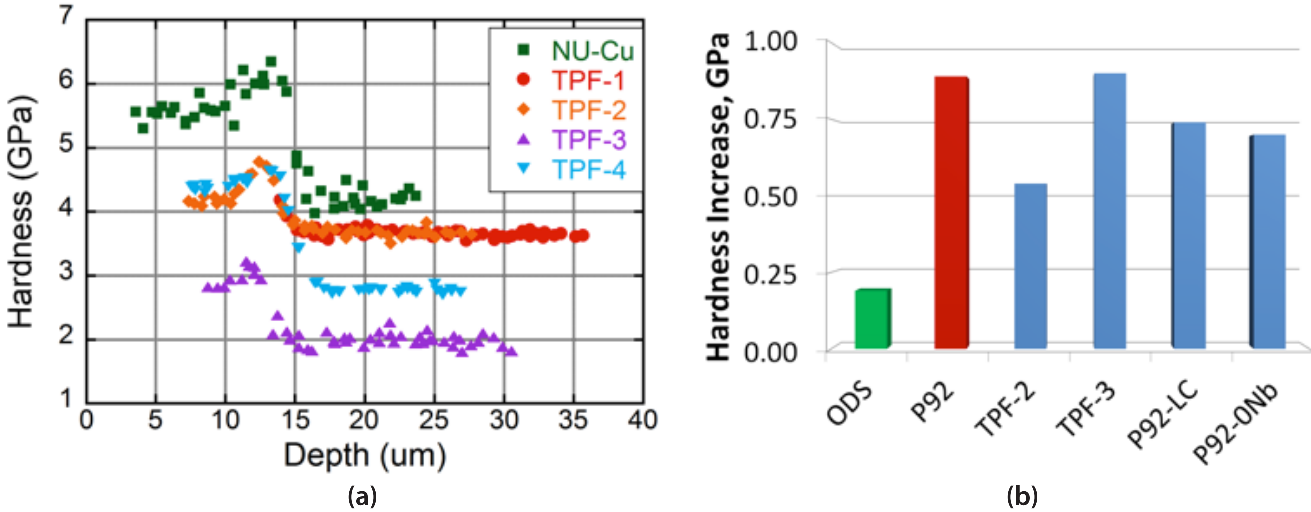

Fig. 2. (a) Hardness vs. depth for the NUCu baseline structural material and the developed TPF alloys after irradiation to 0.5 dpa. The hardness increase below approximately 15 μm is due to irradiation effects. (b) Hardness increase as a function of low-dose irradiation (0.5 dpa) for benchmark ODS 14YWT, baseline P92, and six of our experimental alloys using designed microstructures. At 0.5 dpa, our alloys outperform the baseline P92 alloy and start to bridge the performance gap to mechanically alloyed ODS material.

The alloying and processing concepts for AHCr-FM steels are designed to increase the volume fraction of nanoscale stable precipitates and dislocation sinks relative to current high-chromium FM steels. In particular, the goal is to maximize the volume fraction of fine, stable MX precipitates (Fig. 1a) in an alloy based on FM alloy P92. It is noted that carbon, manganese, vanadium, niobium, or nitrogen alloying may result in the presence of undesirable phases: $M_{23}C_6$ carbides, Z-phase (Cr[V,Nb]N), and Laves phase (Fe2W, Fe2[Mo,Nb]). Thermo-Calc™ models suggest that decreasing carbon increases MX and decreases $M_{23}C_6$ precipitation at the expense of slightly increasing Laves phase precipitation, whereas modifications to manganese, vanadium, and niobium at the levels under consideration have negligible effects on Laves phase precipitation while allowing increased austenite stability and MX precipitation, respectively. Nitrogen can be used to increase MX precipitation, which will improve the size distribution and stability of the precipitate dispersion.

In contrast to high-chromium steels, TPF alloying focuses on producing alternate precipitates (B2-type NiAl; Fig. 1b), in addition to MC type microalloy carbides, in a ferritic matrix. Body centered cubic copper precipitates with B2 NiAl shells are used extensively to strengthen ferritic structural steels. For reactor service, the copper precipitates used to strengthen these structural steels may not be ideal because they are metastable. However, the B2 structure of NiAl alloys is coherent with a ferritic matrix and is therefore significantly more stable. To this end, several steels have been designed based on thermodynamic phase stability. The ratios of aluminum-nickel produced in the heats varied relative to the designed alloys, and further work will be done to analyze the optimum aluminum-nickel ratio with respect to thermodynamic calculations, microstructural characterization, and irradiation performance.

For both the AHCr-FM and TPF alloys, irradiation results have shown improvements in low-dose irradiation hardening resistance versus a baseline P92 alloy that is in current reactor use. Figure 2a shows hardness vs. depth graphs for the TPF alloys compared with the base structural material NUCu. The hardness increase was quantified to compare the optimized alloys with the base alloys, and the low-dose irradiation performance (hardening resistance) of our steel microstructures compared with ODS (14YWT) and P92 steels is highlighted in Fig. 2b. State-of-the-art ODS steels represent the irradiation performance benchmark today, but remain challenging to implement because of manufacturing difficulties. P92 steel is in reactor use today and represents the irradiation, mechanical, and physical performance baseline for our work. Figure 2b shows our steels generally outperform the baseline P92 steel with respect to irradiation hardening and begin to bridge the performance gap to ODS steel.

These concepts show there is a significant window of microstructural refinements available to improve the performance of conventionally manufactured ferritic alloys and potentially reduce the performance gap with mechanically alloyed materials, while maintaining significantly lower cost.

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News for the Reactor Materials Crosscut May 2015

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

Jianmin Qu, Northwestern University, Rémi Dingreville and Khalid Hattar, Sandia National Laboratories

Understanding reactor material behavior in extreme environments is vital not only to the development of new materials for next generation nuclear reactors but also to the extension of the operating lifetimes of existing nuclear reactors. Researchers are conducting 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 currently are demonstrated on alpha-iron and later will be used to characterize two distinctive types of reactor materials—zirconium alloys and high-chromium (~9–12 wt %) ferritic/martensitic steels. Researchers chose these materials as the test bed because they are the archetypes of high-performance reactor materials (cladding, wrappers, ducts, pressure vessel, piping, etc.).

The research project has yielded the following recent results and highlights:

Radiation damage characterization: Nuclear reactor structural materials are subjected to extreme environments, including temperature and irradiation. In addition, transmutation reactions can result in the formation of significant amounts of helium. Helium is of particular concern to reactor materials because it precipitates into bubbles and causes swelling, which can deteriorate the mechanical properties substantially. The role of various radiation conditions on damage accumulation and material property changes is not understood fully. To elucidate fundamental defect accumulation and evolution mechanisms in irradiated body-centered cubic metals, researchers have performed a series of in situ Transmission Electron Microscopy (TEM) ion irradiation and implantation experiments at a variety of temperatures. In particular, researchers have investigated the effects of temperatures on the type of defects generated because of helium ion implantation and the associated defect evolution. Two types of in situ experiments have been performed:

The ability to use multiple probes simultaneously is a unique advantage of synchrotron x-ray measurements. Structures ranging from the atomic scale to the macroscale can be interrogated concurrently, rather than sequentially as in a traditional experiment. WAXS is a powerful tool in characterization of phases, lattice strains, defect populations, and crystallographic texture, whereas SAXS is useful for characterization of the structural properties of a material on the nanometer scale. Tomography can reveal initiation of microcracks and voids as well as phase morphology in 3D, and HEDM is a new non-destructive technique developed in recent years for 3D structural characterization of individual grains in a polycrystalline bulk specimen. Far-field HEDM provides crystallographic orientation, center-of-mass (COM), grain size, stress-state, and defects of individual grains in the polycrystalline aggregate.

- Helium implantation at room temperature followed by annealing
- Helium implantation at elevated temperatures

In both cases, dislocation loop sizes grew with temperature until they disappeared at approximately 500°C. At 600°C, Fresnel imaging revealed the presence of nanometer-sized voids in both cases. Figure 1(a) shows an under-focus image of the room temperature implanted sample after annealing to 600°C; the cavities appear to be evenly distributed through the grains. In the under-focus image of the sample implanted directly at 600°C, presented in Fig. 1(b), the observed behavior is drastically different because cavities are seen only along grain boundaries.

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Experimental observations suggest that different mechanisms are active during helium implantation under sequential versus simultaneous helium implantation and annealing. These results currently are being compared to the spatially resolved stochastic cluster dynamics code (SRSCD), which was developed as part of this project. In this model, the defect accumulation, including voids, bubbles, and dislocation loops, is simulated to provide insights into the various mechanisms contributing to the different behaviors. Particular focus is placed on the difference between high temperature implantation and room temperature implantation with postimplantation annealing.

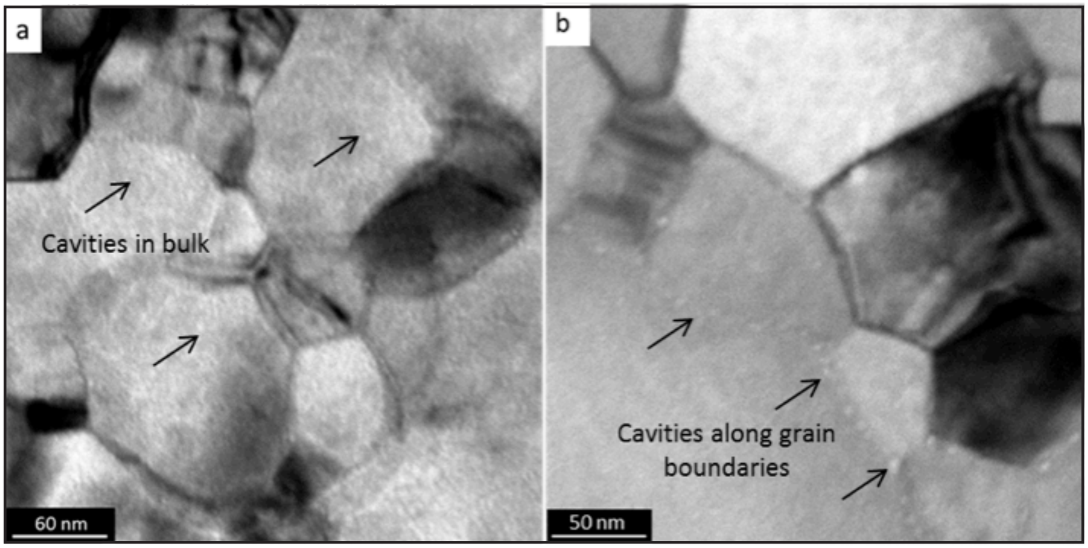

Fig. 1. Under-focus images of cavities at 600°C in samples implanted at (a) room-temperature and (b) 600°C.

Radiation effects characterization: In addition to irradiation damage studies, we also have investigated the resulting radiation effects on mechanical properties. To simulate stress–strain response in iron irradiated to different doses under different conditions, including temperature and dose rate, the predicted defect concentrations and average defect sizes from the SRSCD model are used as an input in the Lagrangian material point method (MPM) crystal plasticity code developed at Sandia National Laboratories. Results have been compared to experimental values for radiation hardening of neutron-irradiated iron, and a good match is achieved. The novelty of this scheme is the direct link between simulations of defect accumulation and hardening. Using this scheme, the combination of SRSCD and MPM has been used to predict the effect of changing irradiation parameters such as dose and dose rate on the expected hardening.

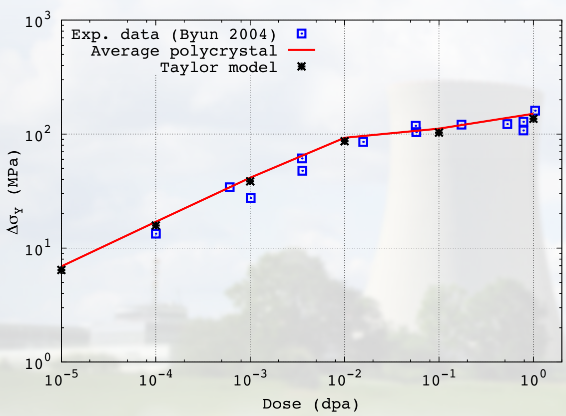

Fig. 2. Simulated hardening of neutron-irradiated alpha-iron. Explicit polycrystalline results using the MPM framework match both one-dimensional Taylor model and experimental results.

In the next phase, researchers will focus on characterizing the effects of different radiation conditions, including temperature, stress, and damage type, on defect accumulation and the resulting material property changes. To study the impact of radiation damage type on resulting defect accumulation, successive and concurrent heavy ion irradiation and helium implantation will be performed in situ in TEM on polycrystalline iron and steel samples. The impact of stress and temperature will be studied via in situ irradiation of samples under different temperature and load conditions. These studies will take place in concert with continued SRSCD and hardening simulations including explicit polycrystalline structures. Particular interest will be paid to the synergistic effects of various radiation parameters, such as the coupling between the diffusion of defects and mechanical fields and the subsequent effects on material aging.

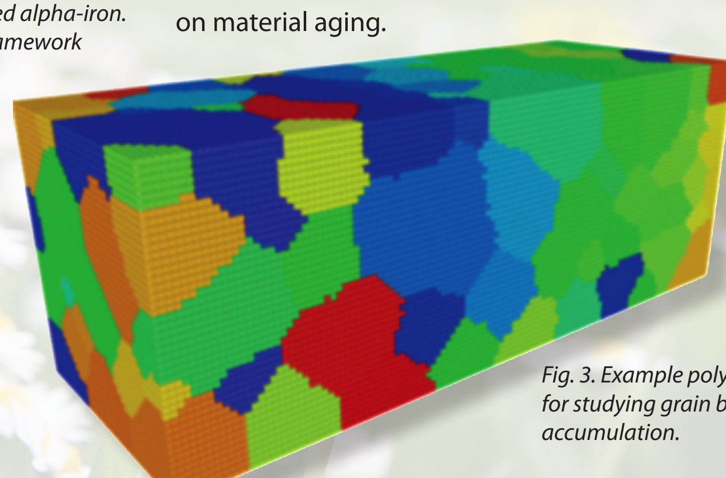

Fig. 3. Example polycrystalline microstructure for studying grain boundary effects on defect accumulation.

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Radiation-induced ductility enhancement in amorphous iron-based and Al2O3-TiO2 nanostructured coatings in fast neutron and high temperature environments of next generation reactors

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Amorphous iron-based and ceramic coatings Al2O3+TiO2 and Al2O3 are being explored for the enhancement and protection of next generation reactor materials expected to operate under severe neutron irradiation conditions and high temperatures. Aluminum, or the combined aluminum and titanium, oxides are characterized by their resilience at extreme temperatures. However, their behavior when in the form of 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. Amorphous iron-based nanostructures exhibit radiation damage resistance to loss of ductility and oxidation/corrosion stemming from the nature of their structure, but experimental assessment is needed to qualify whether such behavior is also observed when in the form of nanostructured coating on nuclear steel substrates. Therefore, the ductility enhancement of nuclear steels and other alloys as well as of mechanical and physical properties under a combination of extremes anticipated in next generation reactors for the combined structures of amorphous iron-based and ceramic nanostructured coatings on nuclear steels and other alloys represent a significant technological advancement in the nuclear reactor materials frontier.

A comprehensive experimental and theoretical study is underway at Brookhaven National Laboratory, augmented with expertise in the areas of nanostructures and coating. The study integrates irradiation damage experiments from fast neutrons generated by the spallation of energetic protons, macroscopic and microscopic studies of the effects on the composite structures induced by the various extremes of neutron irradiation, temperature and aggressively corrosive and oxidizing environments (individually or in combination), electron microscopy, and state-of-the-art x-ray diffraction. The studies to date have revealed that fast neutrons and high temperatures trigger competing mechanisms in amorphous iron-based nanostructured coatings, namely radiation-induced excess volume and temperature-induced amorphous-to-crystalline transformations, which influence the ductile behavior of the composite coating–steel substrate structure. Further, phase transformations and phase diffusion in the ceramic coatings Al2O3-TiO2 and Al2O3 coatings on Ti-6Al-4V and steels influence the behavior of the composite structure at high temperatures and especially in the case of steel substrates where a metastable, α-alumina layer tends to form regardless of the environment because of phase diffusion weakening the coating–substrate interface. The research focuses on identifying a bonding layer different from the Ni-Al that will be compatible and preventing interface degradation between these ceramic coatings and nuclear steels during high neutron fluence and elevated temperatures.

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Amorphous iron-based nanostructured coatings on steel substrates

The post-irradiation mechanical behavior of amorphous iron-based nanocoatings on steel (Fig. 1), observed during a scoping study, prompted further research on the potential of the composite structure in maintaining ductility under irradiation. Contrary to the typical loss of ductility following irradiation, radiation-induced enhancement of ductility was observed along with the KNE behavior over the elastic range attributed to the amorphous coating. To address the two competing mechanisms that affect ductility, thermal annealing to temperatures reaching 1,000°C, followed by stress-strain testing, was performed and compared with radiation-induced effects. Figure 2 depicts the complex stress-strain behavior of the composite structure (amorphous iron-based coating on steel) stemming from irradiation-induced ductility enhancement in the coating, radiation-induced embrittlement in the substrate, amorphous-to-crystalline transitions in the coating, and "softening" in the steel substrate.

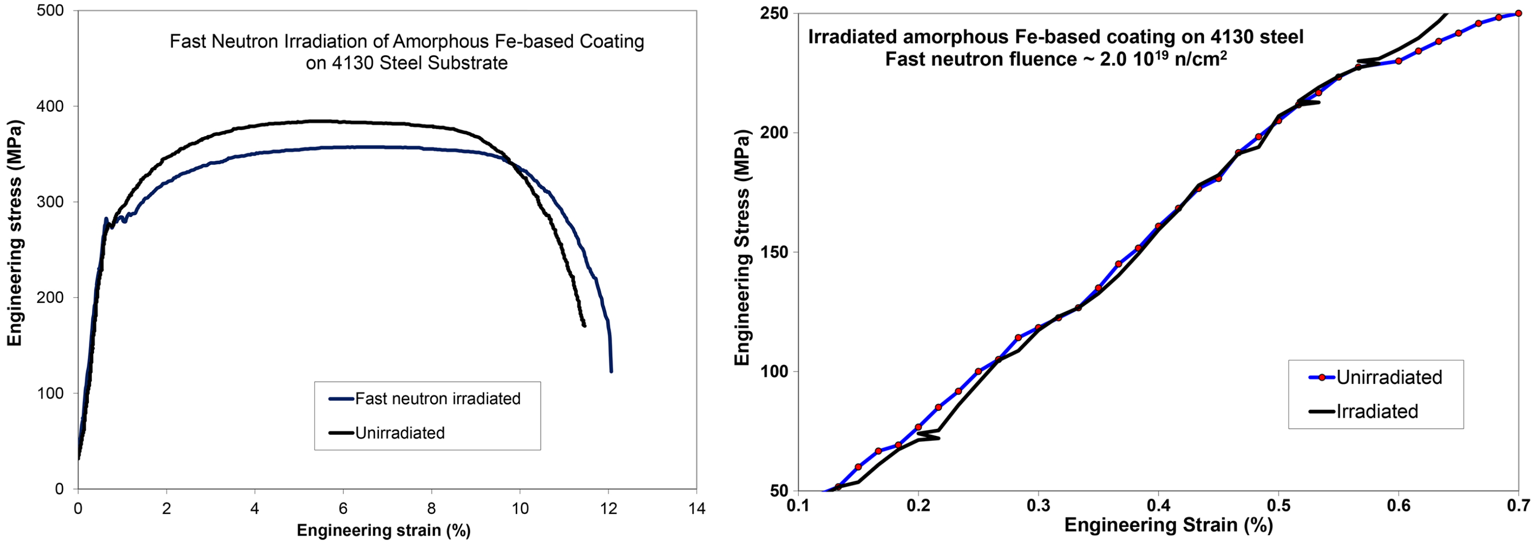
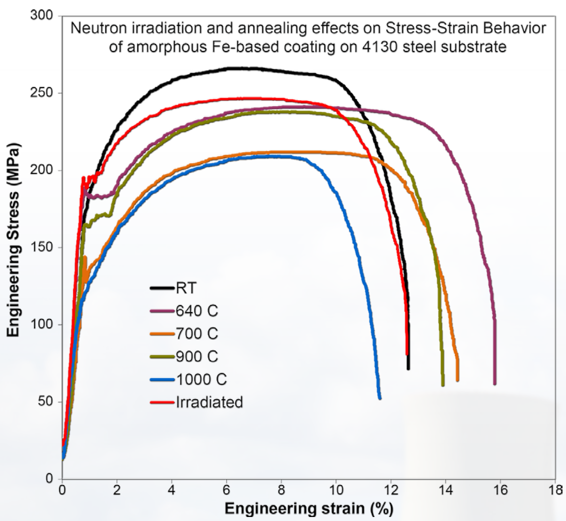


Fig. 1. Stress-strain behavior of amorphous iron-based nanostructured coating of steel substrate following fast neutron irradiation (left) and KNE behavior over the elastic regime (right).



A comprehensive x-ray analysis focused on the changes in the microstructure of both amorphous coating A comprehensive x-ray analysis focused on the changes in the microstructure of both amorphous coating and steel substrate to explain the post-yield behavior. This analysis was based on energy dispersive x-ray diffraction (EDXRD) of irradiated and/or annealed coating–substrate samples in combination with extreme state of stress induced in situ with the x-ray diffraction four-point-bending technique.

The ability of the amorphous coating to resist recrystallization from high temperature and/or irradiation, a key property sought for its application in the next generation reactor environment, was explored using x-ray diffraction, electron microscopy, and optical results of amorphous-to-crystalline transitions in the iron-based nanostructured coating.

Fig. 2. Stress-strain behavior of amorphous iron-based nanostructured coating on steel following irradiation and/or annealing that induces re-crystallization in the amorphous coating.

Al_2O_3 and Al_2O_3-TiO_2 nanostructured coatings on steel and Ti-6Al-4V substrates

The inherent potential of ceramic coatings operating at elevated temperatures has been explored to assess the performance of the composite structure (ceramic coating and metal substrate) to neutron irradiation, elevated temperatures, and high stresses. Findings to date of studies of alumina and alumina–titania (87%–13%) coatings on steel substrates and titanium alloys indicate that while these ceramic coatings are very compatible with titanium alloys as substrates under both neutron irradiation and elevated temperatures, the compatibility breaks down with steel as a substrate at elevated temperatures. The mismatch is the result of a nickel–aluminum interlayer introduced for ceramic–steel bonding.