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

May 2016

Highlights from key DOE-NE programs

In This Issue

Fuel Cycle R&D Program - Advanced Reactor Cladding

The Fuel Cycle Research and Development Program is investigating methods of burning minor actinides in a transmutation fuel. To achieve this goal, the fast reactor core materials (cladding and duct) must be able to withstand very high doses (>200 dpa design goal) while in contact with the coolant and the fuel. Thus, these materials must withstand radiation effects that promote low temperature embrittlement, high temperature helium embrittlement, swelling, accelerated creep, corrosion with the coolant, and chemical interaction with the fuel (FCCI). Research is underway at Los Alamos National Laboratory (LANL), Oak Ridge National Laboratory (ORNL) and Pacific Northwest National Laboratory (PNNL) to develop and test improved radiation resistant materials. Studies are centered on improved tempered martensitic steels and advanced nanostructured ferritic alloys. Recent research on the development of advanced tempered martensitic steels has revealed that new alloys of HT-9 with reduced interstitial content show an improved retention of ductility after irradiation at low temperatures (below 350C). Results were recently published in a Journal of Nuclear Materials article (Maloy, S. A., T. A. Saleh, O. Anderoglu, T. J. Romero, G. R. Odette, T. Yamamoto, S. Li, J. I. Cole and R. Fielding (2016). "Characterization and comparative analysis of the tensile properties of five tempered martensitic steels and an oxide dispersion strengthened ferritic alloy irradiated at approximate to 295 degrees C to approximate to 6.5 dpa." Journal of Nuclear Materials 468: 232-239.)

Excellent progress is also being made at developing improved nanostructured ferritic alloys. These materials are ferritic alloys with high chromium (9-14Cr), high density (>10^{23}/m^3), fine distribution of nanosized (~2 nm) oxide particles and a fine grain size (<0.5 micron).

This fine microstructure provides an alloy with high strength at high temperatures and excellent radiation tolerance (e.g. reduced void swelling and ductility retention at low temperatures) but also increases the difficulty of producing engineering parts (e.g. thin walled tubes) from these advanced materials. Through a collaborative effort between LANL, University of California at Santa Barbara (UCSB) and ORNL a large heat (50 kg) of a nanostructured ferritic alloy (14YWT) was produced and named FCRD-NFA1 (Cunningham, N. J., Y. Wu, A. Etienne, E. M. Haney, G. R. Odette, E. Stergar, D. T. Hoelzer, Y. D. Kim, B. D. Wirth and S. A. Maloy (2014). "Effect of bulk oxygen on 14YWT nanostructured ferritic alloys." Journal of Nuclear Materials 444(1-3): 35-38.). Research is underway to produce tubes from this alloy using techniques such as including high temperature hydrostatic extrusion at Case Western Reserve University, intermediate temperature plug drawing at Rhenium alloys and low temperature pilger processing at Pacific Northwest National Laboratory/Sandvik Special Metals.

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Fuel Cycle R&D Program- Enhanced Accident Tolerant Cladding

Zirconium-based alloys are used across the globe for uranium oxide fuel cladding material and certain fuel bundle structural components in light water reactors (LWRs). After the 1980s Zr alloys became the sole cladding material in LWRs owing to their combination of small neutron capture cross section, reasonable corrosion resistance, and structural integrity under normal operating conditions. Modern Zr alloys enjoy decades of active research in tailored alloy chemistry and production techniques for optimized performance under pressurized or boiling water reactor conditions leading to an impressive record of fuel reliability, on the order of one failure per every million fuel rods deployed. On the other hand, limitations exist with regards to the behavior of Zr alloys under severe accidents. Specifically, these alloys exhibit rapid oxidation kinetics in high temperature steam environments characteristic of LWR severe accident scenarios. The rapid oxidation of the cladding results in swift deposition of a large amount of heat and concomitant hydrogen gas generation, both of which significantly exacerbate the course of accident progression in LWRs.

One effective method to enhance safety margins in LWRs (by delaying core damage and radionuclide release) is to utilize cladding materials with reduced oxidation kinetics in steam. Although this is not the only or an ultimate solution for mitigation of severe accidents in LWRs, this approach reduces the burden on emergency core cooling systems by limiting the deposition rate of heat and hydrogen into the core and has the potential to mitigate the accident under certain scenarios. Accordingly, a significant effort led by the Advanced Fuels Campaign has been dedicated to development and demonstration of Accident Tolerant Fuel (ATF) cladding materials. The key performance criteria for the new ATF claddings is to limit the oxidation rate to 100X slower than the rate for Zr-based alloys. Two primary cladding ATF candidates are FeCrAl alloys and SiC ceramic matrix composites that exhibit >1000X slower oxidation rates owing to a protective alumina and silica layer that forms on their surface in high-temperature steam environments, respectively. Also, protective coating layers on the surface of Zr alloys are under consideration. They have the potential to at least marginally enhance the oxidation resistance. The ATF program is developing these materials in a comprehensive and multi-faceted manner with the goal of an optimized cladding for both normal and accident conditions. This is a significant effort involving both out-of-pile material development and testing as well as long term irradiation testing in relevant environments followed by post irradiation examination. The program goal is to demonstrate ATF concepts by 2022 as a lead test rod (LTR) in a commercial power plant.

Advanced Reactor Technologies - Fast Reactor Structural Program

The Advanced Reactor Technologies (ART) Fast Reactor Structural Program supports the design and licensing of a Sodium Fast Reactor (SFR), a leading candidate for several possible missions, including recycling of used fuel for closing the fuel cycle and power generation.

Advanced structural material with improved performance is one of the key technologies for capital cost reduction and improvement in economic return. Research and development efforts in the past several years have led to the down-selection of Alloy 709 as a replacement material for 316H stainless steel, and a recommendation to qualify Alloy 709 for inclusion in the nuclear section of the American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code. A comprehensive plan for the code qualification and resolution of licensing issues of Alloy 709 for SFR applications was developed in FY15. A multi-laboratory (Argonne National Laboratory, Oak Ridge National Laboratory and Idaho National Laboratory) Phase I implementation plan that includes a 100,000-hour, 650°C ASME code case and the initiation of very long term creep tests, and thermal aging and sodium exposure of Alloy 709 is being established. Gaps in creep and sodium testing infrastructure have been identified. They are being addressed by refurbishing and upgrading existing equipment and the procurement of new creep frames. A forced circulation sodium loop with large exposure vessels to accommodate standard size test specimens is being designed and constructed to address the testing needs. Collaboration with steel makers for fabrication scale-up is being pursued.

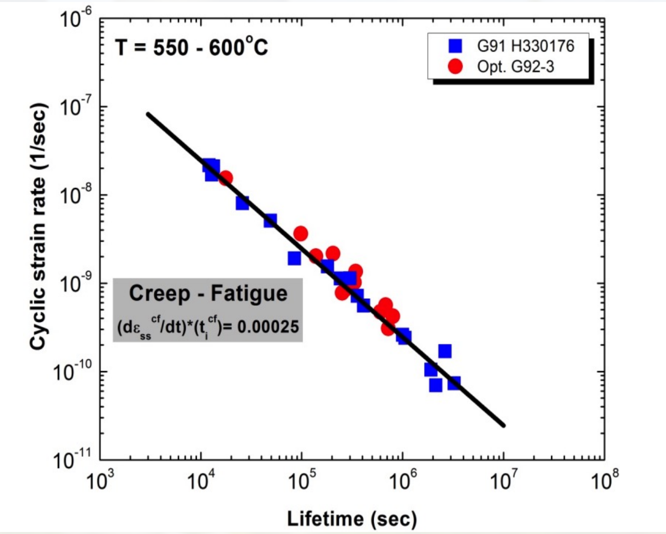

Figure 1: A new creep-fatigue correlation for Grade 91 and optimized Grade 92 steels

Advanced Reactor Technologies - Material Design Technology Program

Materials Design Technology area addresses key long-term design needs for the use of advanced materials for SFR applications. Research and development efforts have been conducted in the past several years to support SFR designs with a 60-year design lifetime. Specific topics include design allowables and creep-fatigue evaluation methods for 60-year design life, as well as creep-fatigue design rules for weldments with a focus on Type IV cracking. The focus of the development is on Grade 91 steel. These activities also support the U.S.-Japan Civil Nuclear Energy R&D Working Group collaboration. An alternate creep-damage evaluation method for creep-fatigue assessment has been developed and ASME code implementation strategy is being developed. A new creep-fatigue correlation for Grade 91 and optimized Grade 92 steels has also been developed. (see Figure 1)

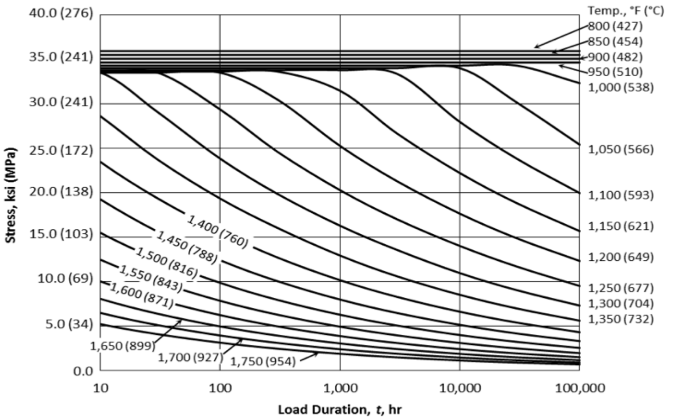

Figure 2 : Time dependent allowable design stresses for Alloy 617 from the draft code case.

Advanced Reactor Technologies - High Temperature Materials

The US Very High Temperature gas cooled Reactor (VHTR) program has been characterizing elevated temperature behavior of Alloy 617 as the leading candidate material for the intermediate heat exchanger. This effort is coordinated by Idaho National Laboratory and includes collaboration with Oak Ridge and Argonne National Laboratories. After analysis of these results, along with historical data and additional results available through the Generation IV International Forum VHTR Materials Program Management Board Materials Handbook, a draft ASME Code Case to allow nuclear construction with this material has been developed. The Code Case is in two parts. The first allows operation up to 800°F (427°C) where material properties are not time dependent and the second will qualify Alloy 617 for construction of nuclear components to be used in temperatures up to 1750°F (950°C) and for service life up to 100,000 hours. The lower temperature Code Case is in the approval process and balloting of the high temperature section will begin in May. Adoption of these Code Cases will significantly extend the maximum design temperature for nuclear construction and is an enabling technology for advanced reactor concepts. An example of time dependent allowable design stress for Alloy 617 from the draft Code Case is shown in the figure 2.

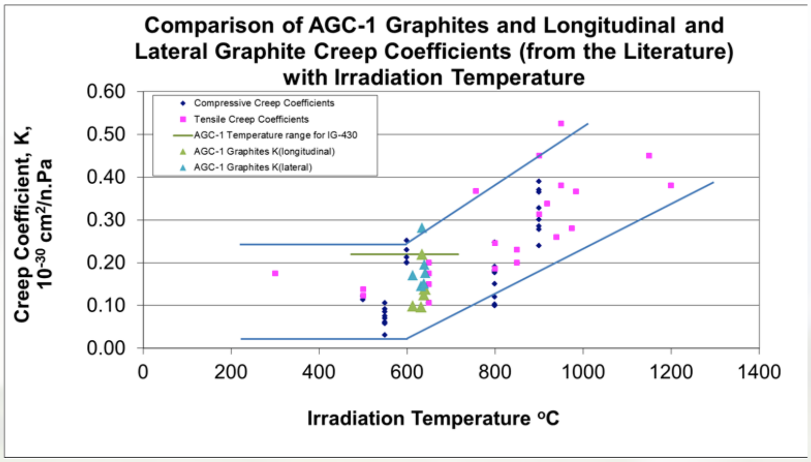

Figure 3: Irradiation Creep measured in AGC-1 Graphites compared to creep measurements in the literature.

Advanced Reactor Technologies - Graphite R&D Program

A wide range of nuclear graphite grades for core component applications are also being characterized. Idaho National Laboratory leads this effort in collaboration with Oak Ridge and Argonne National Laboratories. The Advanced Graphite Capsule (AGC) irradiation experiments are the centerpiece of the US Graphite Program and consists of three pairs of irradiation test trains to be irradiated over a dose range of 0.5 to 7 dpa. The first pair of capsules will be irradiated at 600°C, the second at 800°C, and final irradiations at 1100°C. Analysis of the first capsule, AGC-1, is shown in figure 3 and illustrates that the irradiation creep rates achieved (the life limiting material property for nuclear graphite core components) compare favorably with past graphite irradiation programs. Currently, irradiation results from the second irradiation capsule, AGC-2, is being analyzed, AGC-3 specimens are undergoing PIE, and AGC-4 is being irradiated in the Advanced Test Reactor (ATR) at INL.

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


Dr. Siddhartha Pathak (on left) and Dr. Nathan Mara (on right)

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In this Nuclear Energy Enabling Technologies (NEET) project we are developing a benchmarked measurement tool, which uses spherical nanoindentation stress-strain analysis, for the quantitative assessment of mechanical behavior of ion irradiated materials. In addition to irradiated materials, this technique can also be applied to any material with a modified surface layer – where the surface properties are altered intentionally such as in a graded microstructure, or unintentionally as a consequence of the service life of the material, such as in wear applications, such that its physical, chemical or biological characteristics are different than the bulk of the material – and are of increasing interest for a variety of applications ranging from enhanced wear and corrosion resistance, superior thermal and biomedical properties, higher fracture toughness, and reduced stress intensity factors etc. Quantifying the resulting property gradations poses a significant challenge, especially when the changes occur over small (sub-micrometer) depths. The issue is further complicated for irradiated materials due to the heterogeneous nature of the resulting damage (with strong gradients both on the surface as well as in the depth direction) depending on component location as well as the nature of the irradiation source itself.

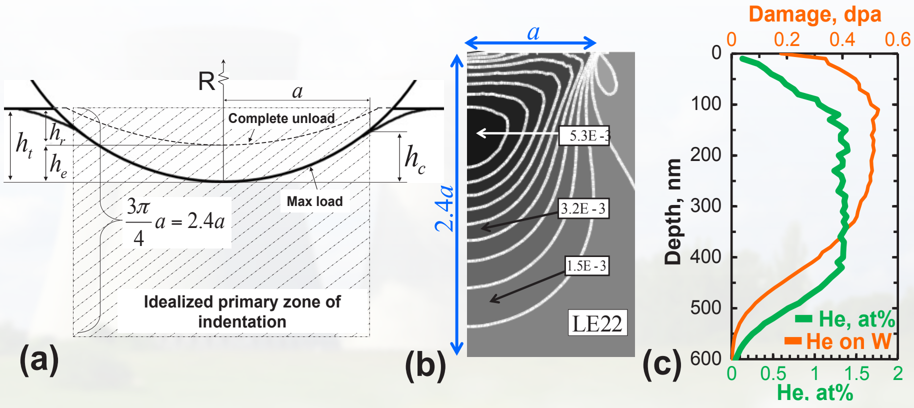

Figure 4: (a) Schematic of spherical indentation showing the idealized primary zone of indentation. (b) Logarithmic strain field (along the indentation direction) for a spherical indenter in the indentation zone (~2.4a, where a is the contact radius) close to the indentation yield. Both the contact radius a, and hence the volume probed by indentation, can be controlled with a proper choice of indenter radii. This approach is thus ideally suited for measuring any mechanical changes in the modified material surface layers, such as probing the (c) damage caused by He irradiation on a tungsten sample.

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Here, 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 [1]. We focus on ion beam irradiation to impart several dpa or more of ion-beam induced radiation damage. The volume of ion-irradiated material is limited by the beam energy to depths of fractions of a micron to several microns, making the investigation of bulk mechanical properties very difficult. On the other hand, nanoindentation testing is ideally suited for this application due to the small sample volumes (~0.5 μm^3) required (see Figure 4).

Among the experimental techniques available at these length scales, nanoindentation, with its high resolution load and depth sensing capabilities, shows the greatest promise due to its non-destructive nature, ease of experimentation and versatility [2,3]. In particular, using spherical indenters, our recent work [1,4] has demonstrated the feasibility of transforming the raw load-displacement data into meaningful indentation stress-strain curves. These indentation data analysis methods have captured successfully the local loading and unloading elastic moduli, the local indentation yield strengths, and certain aspects of post-yield strain hardening behavior in various polycrystalline metal samples, as described in our recent review paper on the topic[1]. As compared to ball indentation [5] efforts in the past that use indentation data after significant plastic deformation has already occurred under the indenter, our approach is advantageous in i) the ability to capture the entire indentation stress-strain response of the material, including the elastic-plastic transition (Yind), prior to the changes induced by the indentation itself, and ii) the ability to probe length scales of interest (by simply varying the indenter radii) from grain-level to bulk-level in a high throughput manner.

The effectiveness of using this technique on irradiated materials has been demonstrated in our recent work on ion-irradiated W (Fig. 5)[6], Zr[7] and 316 stainless steel (ongoing) over a range of ion-irradiation conditions. By coupling the local structure information measured via electron backscatter diffraction (EBSD, Fig. 5a) in the Scanning Electron Microscope (SEM, Fig. 5b) with the local indentation stress-strain response (Fig. 5f)[1], the local mechanical response of ion-irradiated materials from small volumes within individual grains could be determined as a function of lattice orientation (Fig. 5c) and irradiation dose (Figs. 5d and 5e). The described approach combined the high-throughput advantages of spherical nanoindentation, which requires only a metallographically prepared surface, with the ability to quantify the elastic, elasto-plastic transition, and work hardening response in volumes of material relevant to ion irradiation (~1 um^3). This is in sharp contrast to the challenges faced in most small scale testing techniques, such as sample preparation and low sample throughput for focused-ion beam (FIB) based approaches[8], and interpretation of results for hardness testing strategies[3,5]. Our techniques proved powerful in that by sampling a statistically significant number of grains (Fig. 5 a,b), the grain-scale responses could be compiled into maps depicting the elastic and yield behaviors (Fig. 5c), which in turn capture the orientation-dependent local (within grains) mechanical response of the material.

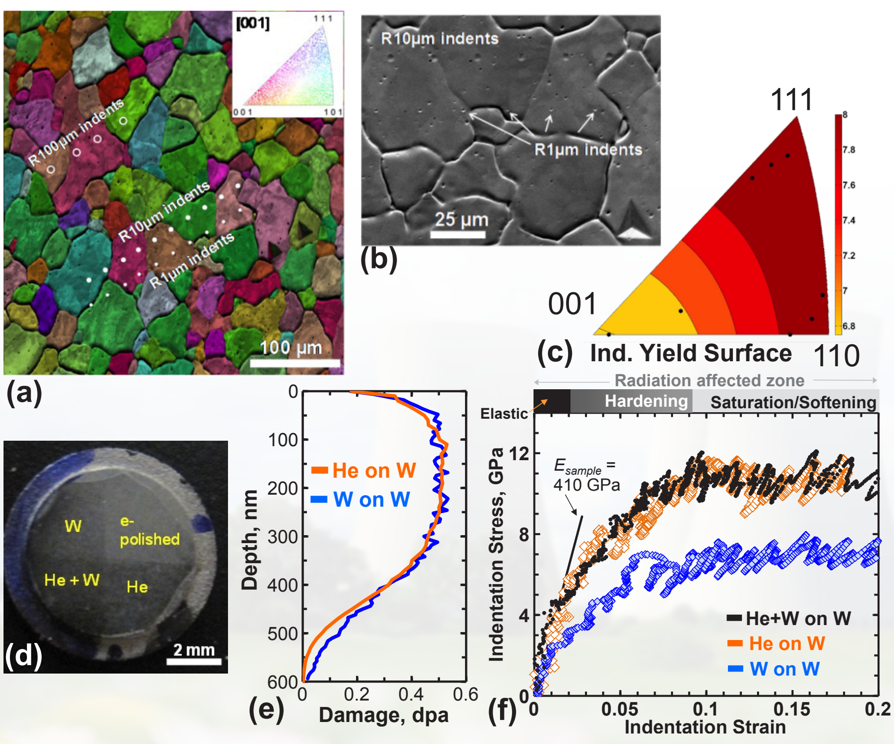

Figure 5: (a) EBSD (b) SEM and (c) yield surface maps of He irradiation on a W sample. (d) and (e) Different ion-irradiation doses on W and (f) their corresponding indentation stress-strain responses.

The lack of validated methods to characterize the changes in the local anisotropic elastic-plastic properties of the microscale constituents and interfaces in ion-irradiated material volume is a major impediment to attaining a deeper physics-based understanding of the effects of irradiation on the material microstructure and its associated mechanical properties – our NEET project aims at addressing this critical need. Our ability to determine the elastic response, elasto-plastic transition, and onset of plasticity in ion-irradiated metallic materials using spherical nanoindentation stress-strain analysis, and compare their relative mechanical behavior to the unirradiated state, is expected to be a breakthrough in mechanical testing of nuclear materials. It provides: 1.) Benchmarked measurement of the stress-strain response from small volumes of material for direct mechanical characterization of ion-irradiated materials 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, future applications of this technique can be easily adapted to indentation of radioactive materials in a glove box/hot cell environment.

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Theoretical Approaches to Understanding Long-Term Materials Behavior in Light Water Reactors

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Prediction of long-term performance of metallic alloys in light water reactor (LWR) environments requires an understanding of how microstructures develop and its impact on materials properties. Environmental conditions producing changes in materials can include temperature, water corrosion, high stress loading, fatigue and radiation. Dominant forms of degradation can vary greatly between different systems, structures and components in an LWR. Knowledge of the developing modes of degradation provides information that establishes materials limits, supports focused planning of materials inspections, and delivers economic and safety based decisions on component replacement. All of which are essential to plant operators. The goal of the Materials Aging and Degradation (MAaD) pathway in the Department of Energy Light Water Reactor Sustainability (LWRS) program is to conduct materials research that establishes a scientific foundation for licensing and managing the long-term, safe and economical operation of plants. Individual research tasks within the pathway focus on current knowledge gaps in long-term materials performance under LWR conditions. This is accomplished through a combination of experimental research, modeling, and information obtained from the examination of materials used in LWR service.

Accelerated irradiation of metallic alloys to high fluences through the use of high flux experimental reactors is useful for providing insight into general materials performance, but specific issues of phase stability, precipitation and swelling may differ from in-service exposed materials. Void swelling phenomenon in metallic materials has been known since the 1960's, but much of the scientific data originates from fast breeder reactor (FBR) programs and may not be fully applicable to LWR conditions over long periods of time. While swelling in austenitic steels under FBR conditions is minimal for temperatures below 350°C, due to differences in the neutron energy spectra and flux rate there is a potential for void swelling in high fluence LWR components. A current effort within the LWRS materials pathway is to develop a comprehensive microstructural-based model for swelling under LWR conditions, through the leveraging and modification of previous fast reactor based models for fuel cladding (see Figure 6). This requires adjustments to the primary radiation damage source terms to account for defect survival, clustering and mobility. The model also requires revisions to the primary source terms to account for different displacement and helium production rates, the accounting for different diffusion mechanisms and sink strengths for extended defects, and the incorporation of

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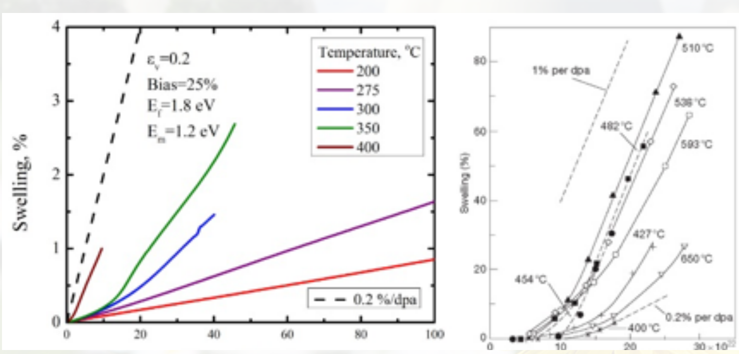

Figure 6: Modeled swelling (left) in 20% cold worked 316 stainless steel as a function of dose taking into account concurrent nucleation of vacancy and interstitial type loops, and experimental data (right) from the EBR-II fast reactor [data from Garner and Gelles].

a subcritical cavity nucleation component based on a cluster dynamics description of helium-vacancy clustering. The current model being developed shows good correlation with the known data from FBR experiments (see figure 6), but will require benchmarking against materials exposed to LWR conditions.

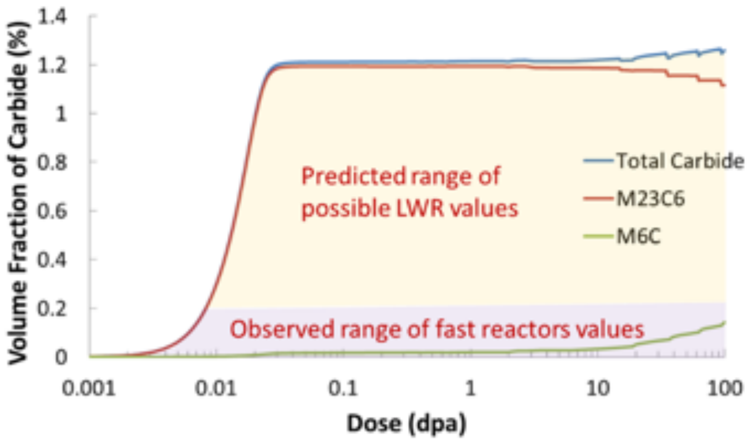

Figure 7: The evolution of total carbide ($M_{23}C_6 + M_6C$) volume fraction under LWR conditions (275 °C and $7{\times}10^{-8}$ dpa/s).

The LWRS materials pathway is also developing models for predicting phase development in high-fluence LWR core internal components. This effort involves combining cluster dynamics (CD) simulation with thermodynamic models (MatCalc and OCTANT) to predict precipitation of second phases in austenitic 316-grade stainless steel under LWR conditions. The CD model predicts the vacancy concentration and dislocation density evolution as input data for precipitation model. The CD-informed MatCalc model with OCTANT thermodynamic database predicts the formation of 1.2% volume fraction of carbide precipitates for typical stainless steel alloy with 0.054 wt.% carbon in its base composition (shown in figure 7). It is suggested that carbide volume fraction is governed by the competition between radiation enhanced precipitation and radiation enhanced dissolution. Experimental data at fast reactors show 0.1-0.2% volume fractions for carbide, but lower dose rates in LWR's may allow carbides to reach their saturation level.

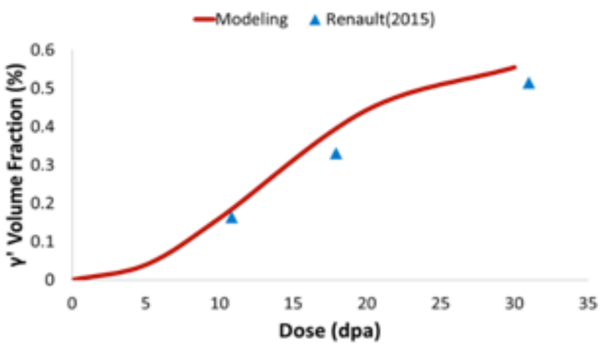

Figure 8: Comparison of modeled γ' development to experimental data in cold worked 316 stainless (390°C at $9.4{\times}10^{-7}$ dpa/s).

The development of computational tools to model radiation induced segregation (RIS) through experimental data, computational thermodynamics and CD modeling of defect concentrations, have resulted in further understanding of material stability under long-term LWR conditions. The development of the G and γ' phases, which are not thermally stable phases in 316 at LWR temperatures, are produced through RIS mechanisms. The development of γ' (Ni_3X, where X is typically Si, Nb or Al) at the expense of the G phase (silicide) in cold worked 316 is due to RIS of solute to dislocations favoring the formation of γ' in microstructures with large initial dislocation densities. The developed model for predicting radiation-induced precipitate development of γ' correlates to earlier fast reactor data as shown in figure 8.

To benchmark the modeling work of radiation-induced swelling and phase development in LWR alloys, it is essential to perform post-irradiation characterization of components harvested or removed from service from commercial LWR plants. To this end, the LWRS/MAaD program has been working with utilities in the procurement of LWR aged materials over the past several years. This includes high fluence baffle former bolts that will provide information on microstructural development comparable to both high flux experimental reactor data and the developed computational models. Baffle bolt harvesting is expected to take place this summer, with information on post-irradiation microstructures available to the modeling effort starting in the fall. The data obtained through experimental, modeling and post-exposure analysis can be further used by industry and regulators to identify operational limits of materials, optimize inspection and maintenance schedules to the most susceptible materials/locations, and, if necessary, qualify swelling-resistant materials for LWR service.

Reference:

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