Final Report on Developing Microstructure-Property Correlation in Reactor Materials using *in situ* High-Energy X-rays

Nuclear Engineering Division

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Final Report on Developing Microstructure-Property Correlation in Reactor Materials using *in situ* High-Energy X-rays

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ABSTRACT

This report provides a summary of research activities on understanding microstructure – property correlation in reactor materials using *in situ* high-energy X-rays. The report is a Level 2 deliverable in FY16 (M2CA-13-IL-AN_-0403-0111), under the Work Package CA-13-IL-AN_-0403-01, “Microstructure-Property Correlation in Reactor Materials using *in situ* High Energy X-rays”, as part of the DOE-NE NEET Program.

The objective of this project is to demonstrate the application of *in situ* high energy X-ray measurements of nuclear reactor materials under thermal-mechanical loading, to understand their microstructure-property relationships. The gained knowledge is expected to enable accurate predictions of mechanical performance of these materials subjected to extreme environments, and to further facilitate development of advanced reactor materials. The report provides detailed description of the *in situ* X-ray Radiated Materials (*iRadMat*) apparatus designed to interface with a servo-hydraulic load frame at beamline 1-ID at the Advanced Photon Source. This new capability allows *in situ* studies of radioactive specimens subject to thermal-mechanical loading using a suite of high-energy X-ray scattering and imaging techniques. We conducted several case studies using the *iRadMat* to obtain a better understanding of deformation and fracture mechanisms of irradiated materials. *In situ* X-ray measurements on neutron-irradiated pure metal and model alloy and several representative reactor materials, e.g. pure Fe, Fe-9Cr model alloy, 316 SS, HT-UPS, and duplex cast austenitic stainless steels (CASS) CF-8 were performed under tensile loading at temperatures of 20-400°C in vacuum. A combination of wide-angle X-ray scattering (WAXS), small-angle X-ray scattering (SAXS), and imaging techniques were utilized to interrogate microstructure at different length scales in real time while the specimen was subject to thermal-mechanical loading. In addition, *in situ* X-ray studies were complemented and benchmarked by *ex situ* characterization using advanced electron microscopy, atom probe tomography (APT) and micro/nano-indentation. The report presented *in situ* tensile test results on neutron-irradiated pure Fe, Fe-9Cr model alloy, 316 SS and CASS CF-8. These *in situ* experiments demonstrate the broad applications of the new capability in understanding several outstanding issues related to irradiated materials.

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1 Introduction

The development and deployment of advanced nuclear energy systems is critically dependent on the development of advanced high performance materials that can tolerate extreme environments of intensive radiation fields, high temperature, stress, and corrosive environments. Life extension of existing nuclear power plants heavily relies on understanding of materials degradation due to irradiation after a long-term service. Designing radiation, heat and corrosion-resistant materials and predicting the response of materials in extreme nuclear environments are two grand challenges for future growth of nuclear energy.

Irradiation generates a large population of point defects and defect clusters in a crystalline solid. Further evolution of these defects leads to formation of extended defect structures, such as dislocation loops, dislocation network, stacking fault tetrahedra, voids, and helium bubbles, as well as precipitates, and phase transformations. The formation of a given type of defect structure is dependent on a particular material, irradiation temperature, dose and dose rate. Uniformly-distributed dislocation loops are typically formed at low doses and low temperatures ($T < 0.3T_m$, T_m is the melting point) [1]. Heterogeneous dislocation loop structures and dislocation network are observed at intermediate and high doses [2]. Significant void and helium bubble formation occurs at higher irradiation temperatures ($T \sim 0.3-0.5T_m$) [3]. Phase transformations have been observed in various alloys under irradiation due to ballistic transport and radiation-enhanced diffusion [4]. Each type of these defects and new phases can affect the physical and mechanical properties of a material and severely degrade the reliability and lifetime of nuclear reactor components. Common degradation phenomena in nuclear reactor materials include low-temperature embrittlement, void swelling and dimensional instability, irradiation creep, high-temperature grain boundary helium embrittlement, irradiation-induced stress corrosion cracking. [5]. Characterization and quantification (e.g. number density, mean size, size distribution) of these irradiation-induced defects as well as understanding their effects on mechanical properties are of fundamental importance to the applications of materials in nuclear energy systems.

Prediction of mechanical properties of a material under irradiation, at the current stage, is still limited to a few simple cases and model configurations. Modeling the relationship between the state of microstructure and the deformation and damage mechanisms has remained largely phenomenological. Many existing deformation mechanism models and radiation effect models do not truly capture the fundamental physical processes of the interactions of dislocations with barriers (e.g. second-phase particles, radiation defects, grain boundaries, etc.), and not necessarily provide a good description of material behavior beyond the testing conditions. The mechanical response of a material is a result of many competing dynamic processes. These processes originate at atomic and microscopic scales with macroscopic consequences. Quantitative modeling is needed to incorporate individual deformation and damage micro-processes into meso-scale material models to accurately describe the evolution and interactions of microstructural elements, so that the required macroscopic mechanical response can be produced under any given external conditions (temperature, strain rate, stress and stress state) in complex material systems. Evidently, multiscale computational modeling must be closely coupled with *in situ* experiments of nano- and micro-scale structural evolution and mechanical behavior of materials to truly capture the fundamental link between microstructure and properties.

Recent progress has shown great promise of synchrotron X-ray techniques for material characterization for nuclear energy applications. Third-generation synchrotron sources produce high-brilliance, hard X-rays, which enable *in situ* measurements with high spatial and temporal resolution. For example, diffraction measurements of internal stresses for different crystalline phases during tensile, compressive, and/or cyclic loading revealed deformation mechanisms in polycrystalline materials [6]. Strain and texture mapping around a crack tip by *in situ* synchrotron XRD provided a better understanding of deformation and crack propagation mechanisms under static and fatigue loading [7].

Combined imaging studies, such as grain structure mapping by diffraction contrast tomography and crack mapping by synchrotron X-ray micro-tomography in corrosive environments, have provided insight into the dynamic interaction of a growing crack with crystal structure and the contribution of different boundary types to crack resistance [8]. These studies show great potential of high-energy (50 – 130 keV) X-rays in understanding the mechanical performance of a material in nuclear environments.

High energy X-rays offer unique opportunities for *in situ* and time-resolved investigations of dynamic processes in bulk materials. They are ideal for probing bulk, millimeter sized samples of engineering materials, and require minimal specimen preparation when applied to activated specimens. Due to the deep penetration of high-energy X-rays, regular lab-size specimens can be examined without compromise of bulk property measurements. This is in contrast to several other *in situ* straining techniques that are often constrained by the small volume of a specimen using either micro-size or nano-size samples. For instance, specimens examined by TEM with *in situ* straining are thin foils with ~100-nm thick. Dimensional constraints make it difficult, sometimes impossible to correlate the observed microscopic deformation mechanisms with the macroscopic mechanical behavior in a quantitative way. The use of small-scale specimens also suffers a statistical sampling problem due to inhomogeneous distributions of lattice defects, chemical segregation, etc. While both high energy X-ray and neutron scattering are capable of probing bulk specimens, the high brilliance of high-energy X-rays provided by the third-generation synchrotron sources make the data acquisition much faster than what can be achieved by neutron sources, enabling the investigation of dynamic processes such as phase transformation, evolution of internal stresses and dislocation activities.

Another unique advantage for *in situ* synchrotron X-ray measurement is the ability of using multiple probes simultaneously. When several probes are combined, material evolution can be monitored across different length scales. Wide-angle X-ray scattering (WAXS) provides several types of microstructural information about a material, e.g. phases, lattice defects, lattice strain, texture, etc. The X-ray diffraction condition can be described by the Bragg equation, $n\lambda = 2d\sin\theta$, where λ is the wavelength, d is the lattice spacing, and θ is the Bragg angle, and n is the integer. Diffraction data contain the reflection position 2θ (hkl), the integral intensity, and the full width of the half maximum (FWHM) of the reflection. Information about the material state can be derived from these data. For example, the hkl values are the fingerprint of the phases in the material. The shift of the reflection position 2θ (hkl) indicates the change of lattice spacing due to the presence of internal strains and stresses or chemical segregation. The relative intensities of the reflection hkl of a given phase reveal the orientation distribution of the reflecting phase, and the preferred orientation is related to the texture present in the material. The relative intensities of the reflections of two or more phases can be used to calculate the volume fractions of phases in a material. The FWHM of the reflection is associated with grain sizes and microstrains. Evaluation of the line profiles gives the information about microstructural defects such as dislocation densities and dislocation cell sizes. X-ray diffraction is a valuable tool for determining grain orientation- and phase-specific stresses because of its sensitivity to phases and crystallographic orientation. Diffraction peaks can be used as an internal strain gauge to characterize the mechanical response of each phase. In materials composed of phases with different mechanical properties, or single phase alloys with texture, the collection of x-ray diffraction profiles *in situ* during mechanical testing provides an effective way of tracking the evolution of the internal stresses due to the incapability between the constituents.

Small-angle X-ray scattering (SAXS) is useful for characterization of the structural properties of a material on the nanometer scale. It uses the contrast between the electron density in the scattering body and the surrounding matrix to derive the information about structures in a material. SAXS is ideal for probing voids, and/or bubbles relative to the matrix. The SAXS technique was applied to investigate the formation of helium bubbles in borosilicate glass [9]. The results showed that the radii of helium bubbles increased from 0.5 to 1.5 nm and the volume fraction from 0.001 to 0.1, respectively as the dose increased

from 9×10^{13} to 2.8×10^{16} ions/cm². The high photo flux of synchrotron radiation sources makes possible monitoring changes of nanostructures in alloys by *in situ* time-resolved scattering experiments.

High-energy diffraction microscopy (HEDM) is a three-dimensional X-ray diffraction technique that measures grain- and sub-grain-scale structural and micro-mechanical behavior of bulk materials. HEDM is classified as near-field (nf), far-field (ff), and very far-field (vff) in terms of the sample-to-detector distance. For all techniques, the sample is rotated about a single axis (typically 180°) and 2D diffraction images are captured with an area detector at defined intervals, and then reconstructed to produce 3D maps of diffraction information. In the nf geometry, the detector is placed only a few millimeters behind the sample. Spatial and orientational resolutions are gained at the expense of the strain resolution in this set-up. Three-dimensional grain maps, similar to ones obtained by 3D-EBSD, can be obtained with the nf-HEDM technique. However, *in situ* experiments are severely limited with the nf-HEDM technique due to limited space for sample environment. In the ff-HEDM geometry, a large area detector is placed at an intermediate distance from the specimen, typically ~1 m away. The ff-HEDM provides crystallographic orientation, center-of-mass (COM), grain size, and stress-state of individual grains in the polycrystalline aggregate. In the vff-HEDM configuration, a detector is placed even further away from the sample, typically ~5 m away, and the angular resolution is significantly improved such that dislocation structures within a grain can be analyzed from broadening of the diffraction spots. For ff-HEDM and vff-HEDM, the detector is placed sufficiently far away from the sample such that the effect of thermal-mechanical load on the micromechanical behavior of the sample can be investigated *in situ*.

X-ray tomography is a non-destructive method for 3D structural measurements. A series of high-resolution radiographs are taken while the sample is rotated by a small increment between images. A 3D image is then reconstructed to obtain a 3D representation of the object, revealing voids, pores, and cracks in the material. It has become an important characterization tool for study of degradation and damage mechanisms, and has been used to study cavitation, fracture, micro-cracking, fatigue cracking, and stress corrosion cracking [10,11].

While the combination of the above X-ray diffraction/scattering/imaging techniques and thermal-mechanical loading is powerful in studying macroscopic mechanical behavior at a microstructural level, so far these techniques are limited to unirradiated specimens only, and irradiated specimens are largely *ex situ* tested. The objective of this project is to demonstrate the capability of high-energy X-rays in characterization of irradiated materials subjected to *in situ* thermal-mechanical loading to advance the understanding of microstructure-property relationship in nuclear reactor materials. Three tasks were proposed:

- Task 1: Effect of Irradiation on Deformation Behavior of Ferritic Alloys
- Task 2: Effect of Irradiation on Deformation Behavior of Austenitic Alloys
- Task 3: Effect of Irradiation on Deformation Behavior of Duplex Microstructure

In the following sections, we first present an *in situ* X-ray Radiated Materials (*iRadMat*) apparatus designed to interface with a servo-hydraulic load frame at beamline 1-ID at the Advanced Photon Source. This new capability allows *in situ* studies of a radioactive specimen subject to thermal-mechanical loading using a suite of high-energy X-ray diffraction, scattering and imaging techniques. We then present the results of a series of *in situ* tensile tests conducted on neutron-irradiated ferritic and austenitic alloys, and cast austenitic stainless steels (CASS) using the *iRadMat* to understand the effects of neutron irradiation on deformation behavior of common reactor materials.

2 *In situ* X-ray Radiated Materials (*iRadMat*) Apparatus

2.1 Design of the *iRadMat*

An *in situ* straining/annealing apparatus for irradiated materials (*iRadMat*) was developed at the Argonne National Laboratory. The apparatus was designed to interface with the MTS servo-hydraulic load frame equipped at beamline 1-ID at the Advanced Photon Source (APS) for conducting *in situ* straining/annealing tests of neutron-irradiated specimens with high-energy X-rays. The design objectives and functions of the *iRadMat* apparatus are as follows:

- **Radiation protection:** The apparatus must shield ionizing radiation from, and provide containment of, an activated specimen. A flexible yet robust design of radiation control was particularly important for this project, as the *in situ* thermal-mechanical tests were carried out at a public beamline of a national user facility (in this case the APS 1-ID beamline). The apparatus is expected to function as the primary radiation shield between the radioactive specimen and the environment, and the secondary containment in normal situations. It also needs to be able to function as the primary containment (and the only containment) should the original primary containment fail.
- **Mechanical loading capability:** The apparatus was designed as an insert to an existing servo-hydraulic load frame (MTS load frame model 858 equipped with hydraulic pump model 505.07) at the APS 1-ID beamline. The insert concept leverages the robust and versatile mechanical loading capability that a standard load frame delivers.
- **Heating capability:** The apparatus was designed to function from room temperature (RT) to 1200°C at a vacuum level of 1×10^{-5} Torr with a cylindrical zone of uniform temperature of about 25 mm in diameter and 50 mm in height in the center of the furnace. This temperature range covers the interest for a majority of materials for nuclear applications. The large uniform temperature zone ensures that the sample has a relatively uniform temperature during the experiment.
- **Compatibility with X-ray measurements:** Both imaging and scattering X-ray techniques should be possible during thermal-mechanical loading. This requirement, together with the definition of maximum scattering angle, means that the furnace must have large windows (on both the incident side and the exit side) which are capable of withstanding radiation, heat and vacuum pressure, while producing minimal X-ray scattering.
- **Rotational capability:** The sample can be rotated $\pm 180^\circ$ with respect to the beam while applying uniaxial tension. The ability to rotate the sample with respect to the incident X-rays, combined with the use of area detectors, enables 3D rather than 2D studies. To realize this functionality, an in-grip rotation mechanism was developed.
- **Mobility:** The ability to perform tests with the apparatus *ex situ* is needed to commission and improve its capability. Furthermore, should the failure of the original primary containment occur and the device in turn takes on the role of the primary (and the only) containment, the entire apparatus needs to be able to be easily transported offsite.

Figure 1 shows the *iRadMat* apparatus inserted into the load frame. The apparatus was custom manufactured by OXY-GON INDUSTRIES, INC. The detailed design considerations, features and constraints of the *iRadMat* apparatus are described below.

Figure 1. A 3D view of the *iRadMat* apparatus inserted into the load frame. The power and the control systems are not shown here. The direction of the in-coming beam is along $+Z_L$.

2.1.1 Radiation safety

The design considerations for radiation safety encompass three aspects: 1) integrated radiation shielding within the furnace chamber; 2) radioactive sample containment; 3) negative pressure relative to the surrounding environment and particle filtering at the exhaust line.

The vacuum furnace has a double-walled, water-cooled 304L stainless steel chamber with the approximate interior dimensions of 241 mm $\times$ 241 mm $\times$ 385 mm. Integrated interior radiation shielding was designed to provide an effective protection from ionizing radiation emitted by a radioactive specimen. Locating this shielding close to the sample inside the chamber limits the cost, weight and complexity relative to putting it outside the chamber. As shown in Fig. 2, a set of 5 mm thick tungsten plates was attached to all six internal walls of the chamber with sufficient overlap at all seaming lines, leaving only openings at the two X-ray windows and the two viewing ports orthogonal to the X-ray windows. Motorized tungsten shutters were installed directly in front of those windows. These shutters can be remotely actuated, both for viewing the sample with cameras and for X-ray experiments. Radiation shielding and stainless steel walls reduce the dose rate from a Co-60 source (mostly the high-energy gamma) by $\sim 50\%$, and provide sufficient protection for handling low-activity specimens with minimal radiological control at a public beamline.

Vacuum provides a negative pressure and prevents removable radioactive contaminants from spreading outside the furnace chamber. A high efficiency particulate air (HEPA) filter was installed in the vacuum exhaust line to further prevent any contamination release. After an *in situ* thermal-mechanical loading experiment on an activated sample, the interior of the device is surveyed and approved by the Health Physics on-site, to confirm that the equipment is clean and can be released from the experimental hutch.

Figure 2. A 3D view of the *iRadMat* furnace interior.

2.1.2 High temperature and vacuum

The vacuum environment is important for high-temperature operations to minimize sample oxidation, protect the radiation shielding materials as well as provide a negative pressure for radiation protection. The vacuum system consists of a dry scroll pump and a turbo molecular pump. Steel bellows are used to connect the chamber to the pumping system to minimize sample vibration. An ion gauge that directly sees the chamber pressure serves as the vacuum sensor.

A set of tungsten meshes surrounding the sample provides heating (shown in Fig. 2) when energized. Five layers of Mo sheets with the total thickness of 1.25 mm are installed on all sides for heat insulation. Both W heating meshes and Mo insulation sheets have sufficient openings for X-rays to pass and for viewing. The exterior walls of the furnace are water cooled with the pipes buried inside the double walls.

The temperature is monitored by thermocouples at multiple locations inside the furnace. Two thermocouples are located near the specimen as the primary thermocouples for temperature control and monitoring. Having a thermocouple directly in contact with the sample is not easily achievable; instead, two thermocouples reach close to the top and bottom of the sample through the thermocouple wells within the gripping device. The tips of the thermocouples are buried right underneath the surface, close to where the sample is clamped. The actual temperature on the sample is taken as the average of the two independent readings. An over-temperature control thermocouple is placed near the center of the furnace. The temperature of the X-ray window is monitored by a thermocouple in contact with the window.

2.1.3 X-ray and optical windows

Two X-ray windows provide the X-ray paths for the incident and exit X-rays. Two additional windows provide viewing ports in the directions perpendicular to the X-ray path. The design is a balance between multiple, often conflicting, requirements. All windows must be mechanically stable with respect to heat and radiation, and withstand thermal loads arising from heating and cooling. Additionally, the X-

ray window material must have high X-ray transmission and minimal scattering signals. The incident X-ray window must accommodate the beam size, while the exit window must be large enough to ensure sufficient diffraction angle coverage for WAXS and HEDM experiments. Given these constraints, single-crystal sapphire windows of 45 mm (diameter) $\times$ 2 mm (thickness) were selected as the incident and exit X-ray windows, allowing a 2θ scattering angle coverage of 8° . The two orthogonal view ports use 45 mm (diameter) $\times$ 6.35 mm (thickness) quartz windows.

2.1.4 Sample-grip assembly

The sample-grip assembly provides the primary radiation containment and the gripping mechanism that can transfer the load exerted by the load frame to the sample and prevent sample twisting if rotated. The grip assembly is designed such that it can be removed from the chamber without disturbing the alignment of the chamber. This modular approach is adopted so that multiple radioactive samples can be transferred in one shipment from the radiological facility to the APS beamline, and the samples can be installed and tested while the *iRadMat* apparatus remains aligned in the beamline.

Figure 3 (a) shows two tensile specimen geometries that fit the current design of the gripping device: one has a gauge length of 5 mm and the other has a gauge length of 7.6 mm. The gauge width varies between 1.0 and 1.5 mm, and the gauge thickness between 0.5 and 1 mm. Figure 3 (b) shows the grip assembly with details of the gripping mechanism. A more general view of the grip assembly interfaced with the load frame is shown in Fig. 2. The gripping device consists of a pair of 25.4 mm-diameter metallic bars that are connected to the chamber pull rods through loading pins. The material of the bars is Inconel X750 for room temperature to moderately high temperature applications, or TZM refractory alloy for very high temperature applications. The bars are connected through two guide rods embedded in two deep channels inside the grips. The guide rods can slide freely in the channels, allowing only one dimensional motion along the loading axis, and minimizing torsional stresses on the sample when rotating under tension.

The gripping mechanism includes two alignment pins (on the lower and upper grips, respectively). These pins are used to position the sample in the loading axis but not for applying uniaxial tension on the sample. The gripping sections of the sample are placed on the roughened surfaces with pins going through the pin holes on the sample. Then, a set of clamping pieces is tightly bolted onto the grips. The mechanical loading is achieved by the friction between the gripping sections of the sample and the roughened surfaces of the grips and clamps. The grips are connected with the top and bottom pull rods through loading pins. The pull rods are water-cooled. The top pull rod is connected to the chamber using steel bellows and moved vertically. The bottom pull rod is sealed to the chamber using a compression seal and nominally fixed against the vertical motion.

Figure 3. (a) Tensile sample geometries and nominal dimensions, and (b) a detailed view of the grip-sample assembly.

To avoid contamination of the furnace by an activated sample, encapsulation is needed between the sample and the chamber. This encapsulation serves as the primary containment (Fig. 4 (a)). For room temperature tests of activated samples, two Kapton tubes and 3D-printed plastic adapters are used. The thin Kapton tubes give minimal X-ray scattering signal and absorption [12]. For experiments at elevated temperatures and under vacuum, amorphous fused quartz tubes and stainless steel adapters are used for the double-walled containment. Because quartz is largely opaque to infrared wave radiation from the heaters, sample heating is primarily carried out by conduction.

An activated tensile sample is pre-assembled into the gripping device and surveyed by the Health Physics at a radiological facility prior to transfer to the beamline. Multiple sets of grips are manufactured so that several samples can be transported to the beamline in one shipment.

External radiation shielding is used to minimize exposure to radiation during the transport of the grip assembly and its installation inside the furnace, as shown in Fig. 4 (b). It is made from a tungsten tube with a wall thickness of 10 mm secured inside a stainless steel shell structure. The shell structure is designed for quick assembly and removal of the radiation shield. The external radiation shield is removed after specimen installation prior to closing the chamber door, and re-assembled inside the chamber before removing the sample-grip assembly from the chamber.

Figure 4. (a) The double-encapsulated gripping assembly for an activated specimen, and (b) the double-encapsulated gripping assembly with external radiation shielding.

2.1.5 In-grip rotation – tension mechanism

The servo-hydraulic load frame is capable of exerting up to 15 kN uniaxial force in tension and compression, and with a 100 mm stroke that allows for a variety of sample geometries and environments to be used [13-16]. It is capable of monotonic loading as well as cyclic loading in both displacement and load control.

An integrated in-grip rotation mechanism (shown in Fig. 5) was designed to permit the sample to be rotated around the loading axis by $\pm 180^\circ$ with minimal torsional stresses applied on the sample. This rotational capability (designated as ω) can be used for increasing the number of measurement directions as well as improving the crystallite sampling statistics for scattering measurements. Furthermore, it enables three-dimensional techniques, e.g. HEDM and μ -CT that require full sample rotation. The furnace and the load frame are aligned with respect to the incoming X-rays using a set of translational and rotational stages below the load frame, and remain stationary during sample rotation.

The rotation about the loading axis is realized by attaching a Huber 410 stage at the bottom of the loading train, as shown in Fig. 5. The Huber 410 stage can withstand 2.5 kN axial load in tension, which defines the maximum tensile load that can be exerted on a specimen. The rotational motion of the Huber 410 is transferred to the bottom pull rod through a housing structure, which then drives the bottom grip of the grip assembly. The two guide rods translate the rotation to the top grip of the grip assembly. The blind holes where the guide rods are inserted to are machined with tight tolerances to minimize rotational slop. This allows the rotation to be rigidly translated upward to the top thrust bearing assembly. The thrust bearing contains Diconite™ coated steel balls, which provide a very low friction coefficient (< 0.04) and have high temperature capability. The top pull rod is separated from the bearing so that the rotational motion is confined within the chamber.

Figure 5 – The rotation stage of the *iRadMat* apparatus.

2.1.6 Integration of the *iRadMat* apparatus with the beamline infrastructure

The vacuum furnace, when not engaged with the load frame, is fully supported on a mobile cart for transportation and storage. When engaged with the load frame, the chamber is detached from the cart while the power, pumping and cooling systems remain on the cart. The manifolds controlling the water circulation are fixed on the cart with only two end connections for water supply and return, which are supplied at the beamline. A quick connect fitting is attached to the backfill manifold to connect to the inert gas supply, if needed.

To prepare for an *in situ* thermal-mechanical loading experiment, the chamber is detached from the mobile cart and securely placed on a set of manual translational plates mounted to the base of the load frame (Fig. 5). A set of finely adjustable jacks allow small rotations of the *iRadMat* apparatus with respect to the load frame and its loading axis. When the rotation axis of the *iRadMat* apparatus and the loading axis of the load frame are nominally aligned, the top and bottom pull rods are secured to the respective grips of the load frame with pins. Finally, the load frame, while the *iRadMat* apparatus inserted, is aligned with respect to the X-ray beams with a set of rotational and translational stages placed below the load frame.

The layout of the major hardware in the experiment hutch is illustrated in Fig. 6. The WAXS detector array, composed of four GE-RT41 detectors [17] (namely, GE1, GE2, GE3 and GE4, and referred as the Hydra array), can be placed in between 1.0 - 4.5 m downstream from the sample, and the SAXS detector is placed nominally 6.5 m away from the sample. The downstream X-ray window of the furnace allows the scattering angle coverage of 8° such that 7 to 10 diffraction rings from iron-based ferritic and austenitic alloys can be recorded by the WAXS detectors at the X-ray energy of 125 keV with a sample to detector distance of 2 to 3 m. For SAXS, the q range is 10^{-2} - $10^0 \text{ \AA}^{-1}$ with X-ray energy of 125 keV and the detector – sample distance of ~ 6.5 m, where q is the magnitude of the reciprocal-space scattering vector defined as $q = 4\pi \sin \theta / \lambda$ (2θ is the Bragg scattering angle and λ is the X-ray wavelength).

Figure 6. A schematic of the *iRadMat* apparatus in the APS beamline 1-ID.

2.1.7 Evaluation of radiation effect on X-ray detector performance

To assess the response of the GE area detectors to an elevated radiation background due to the presence of an activated sample, the detector was tested in a radiological facility using a neutron-irradiated austenitic stainless steel specimen and a nickel alloy specimen with two different dose rates (35 mR/h and 150 mR/h at 30 cm, mostly gamma exposure). These specimens were placed at a distance of approximately 1 m from the detector. The detector was also tested with a Cs-137 sealed source (3 mR/h on contact, beta and gamma exposure) in direct contact with the detector to simulate an elevated radiation background. The recorded signal levels over typical exposure time scales (1 - 10s) were only slightly over the background, even for the highest activity. This is largely attributed to the energy sensitivity of the detector, which is the highest in the 60 - 120 keV range (typical X-ray energies used for these synchrotron experiments) while typical gamma emissions from the samples are at higher energies (>200 keV) where the detector sensitivity is lower.

2.2 Performance of the iRadMat

Both *ex situ* and *in situ* tensile tests were conducted at temperatures of 20-400°C in vacuum to evaluate the performance of the iRadMat. Tensile tests were carried out in displacement control at a constant crosshead speed. The nominal strain rate was in the range of 10^{-5} - 10^{-3} /s. While inserting the iRadMat apparatus into the load frame is a challenging task due to its weight and complexity, the instrument stiffness remains relatively constant with approximately 10% scatter. The *in situ* stress-strain curves obtained using the iRadMat apparatus were regularly compared to those obtained from other well-maintained, calibrated load frames to confirm that the iRadMat setup was stable and reliable. Figure 7 shows representative stress-strain curves from *ex situ* tensile tests with the iRadMat. In all these tests, the nominal strain rate was on the order of 10^{-4} /s. Figure 7 (a) compares the stress-strain curves for 316 stainless steel specimens tested at room temperature with >100% total elongation, and at 300-320°C where both strength and ductility were reduced. Figure 7 (b) is for a cold-worked G92 ferritic-martensitic steel with high strength. Figure 7 (c) shows the stress-strain curves of a dual-phase cast stainless steel, where the tensile test with the iRadMat was performed with continuous $\pm 30^\circ$ ω rotation, causing the step-like response. In all cases the data from the iRadMat tests agrees well with the reference lab tests.

Figure 7. Stress-strain curves for (a) 316 SS tested at 20-320°C, (b) G92 ferritic-martensitic steel tested at 20°C, (c) cast stainless steel tested at 20°C using the iRadMat and lab tensile machines.

The diffraction signal from the single crystal sapphire window is strongly dependent on the angle between the incoming X-ray beam and the window. Figure 8 shows two cases when the angles were slightly different. The diffraction condition was better met in (a) than in (b). Before an *in situ* experiment, the chamber was rotated about $\pm 5^\circ$ and a position was chosen where the intensities of these diffraction spots were minimized. This background can be removed by subtracting the bright image (the image taken without the sample in the beam) from the sample image, after normalizing by the measured sample transmission.

Figure 8. Bright field images from GE1 of the Hydra array showing (a) strong single crystal diffraction from the sapphire window and (b) much weaker diffraction after rotating the window about 1 degree. The color bar denotes intensity in natural log scale.

As mentioned earlier, double-layered quartz containment was applied around the activated sample for elevated temperature tensile tests. The total thickness of the amorphous quartz in the X-ray path was 7 mm, giving a transmission factor of 0.79 when the energy of the incoming X-rays was 123 keV. Three set of SAXS signals were compared (with two X-ray windows in the beam path):

- No sample and no quartz in the beam path (“bright condition”)
- Quartz containment only in the beam path (“quartz condition”)
- Quartz containment and a piece of 10 mm-thick glassy carbon in the beam path (“quartz + glassy carbon condition”)

Figure 9 shows the SAXS profiles of these three cases. Each curve is the integration of the detector image with 1 s exposure. The intensity is normalized by the transmission factor (0.79 for quartz and 0.77 for quartz+glassy carbon) measured by two ion chambers located before and after the *iRadMat* apparatus, respectively shown in Fig. 6. It was found that the quartz does not affect the small-angle diffraction in the q range of interest.

Figure 9. SAXS profiles for the bright condition, the quartz condition and the quartz + glassy carbon condition.

A cold-worked, fine-grained pure bcc Fe sample neutron-irradiated to 0.01 dpa at 300°C at the Advanced Test Reactor, Idaho National Laboratory was tested at 300°C in vacuum to demonstrate the high-temperature capability of the *iRadMat* at the APS beamline 1-ID. The tensile test was performed at

300°C at a strain rate of 1×10^{-5} /s. Figures 10 (a) and (b) show the temperature profile and the engineering stress-strain curve, respectively, obtained from the *in situ* tensile test. The temperature variation during the tensile test was within 20°C. The measured 0.2%-offset yield strength of the material was 300 MPa, the ultimate tensile strength of 316 MPa, the uniform elongation of 1.7%, and the total elongation of 5.5%. The X-ray energy was 123 keV and the beam size was $0.3 \text{ mm} \times 0.3 \text{ mm}$. The WAXS detector calibration was performed using a NIST standard CeO_2 powder sample (SRM 674b), yielding lattice strain uncertainty of 1×10^{-4} . The corresponding instrumental broadening was characterized using a NIST standard LaB_6 sample (SRM 660a). With $8^\circ 2\theta$ angle coverage from the *iRadMat*, the minimum d-spacing that can be recorded on the WAXS detectors was $0.72 \text{ \AA}$, cutting off at the $\{321\}$ reflection for the bcc Fe. Figure 11 shows the transformed diffraction patterns recorded on all four GE detectors at the onset of the tensile test. It is observed that the intensity was rather constant around the bcc-Fe diffraction rings, confirming that the sample was fine-grained with a few larger grains in the aggregate, and the diffraction signal from the sapphire window was unnoticeable relative to the scattering signal.

Figure 10. (a) Temperature profile during the heating and the tensile experiment and (b) the stress-strain curve of neutron-irradiated pure Fe.

Figure 11. The transformed detector images taken at the onset of the tensile test showing multiple $\{hkl\}$ reflections from the bcc phase. The colorbar denotes intensity in natural log scale.

The lattice strains measured during the uniform deformation ($<1.7\%$ strain) is shown in Figure (a) where the lattice strains for the loading direction (LD) and transverse direction (TD) are shown as a

function of applied macroscopic stress in closed and open symbols, respectively. The lattice strain for a particular family of crystallographic planes, $\epsilon(s)$, is computed as:

$$\epsilon(s) = \frac{d(s) - d_0(s)}{d_0(s)} \quad (1)$$

where $d(s)$ and $d_0(s)$ are the lattice plane spacings measured along a direction, s , at a macroscopic applied load and zero load, respectively. Figure 12 (b) shows the peak broadening, B in terms of the full-width at half-maximum (FWHM) as a function of macroscopic strain, corrected for instrument broadening using $B_{\text{sample}}^2 = B_{\text{total}}^2 - B_{\text{instrument}}^2$.

Figure 12. (a) Lattice strain and (b) fwhm measured during the uniform deformation of an *in situ* tensile test at 300°C for neutron-irradiated pure Fe.

In general, the lattice strains along the LD are increasing and the lattice strains along the TD are decreasing with increasing macroscopic load. The magnitudes of the TD lattice strains are approximately 1/3 of the LD lattice strains, demonstrating the Poisson's effect. At the macroscopic yield, the lattice strains start to deviate significantly from linearity and different crystallographic planes carry different magnitudes of lattice strains indicating yielding and change in mechanical state at the grain scale. The FWHM plot shows that the peaks broaden with increased deformation.

Utilizing the in-grip rotation capability of the *iRadMat*, the necking region was imaged using X-ray radiography in the specimen width and thickness directions, as seen in Figs. 13(a) and (b), respectively. The images were taken at the end of the test at which the applied macroscopic load was close to zero. The final rupture induced by shearing is clearly revealed in Fig. 13(b). SAXS measurement did not show any signal during the entire deformation process, which indicates that for this thermal-mechanical condition the pure Fe sample had no measurable nano-void formation.

Figure 13. X-ray radiography showing the necking region in (a) the width direction and (b) the thickness direction.

3 Effect of Irradiation on Deformation Behavior of Ferritic Alloys

The goal of Task 1 is to understand the effect of irradiated microstructure on the deformation behavior of ferritic alloys. Below we present a set of *in situ* tensile experiments conducted on neutron-irradiated Fe-9wt.%Cr model alloy using the *iRadMat* apparatus.

3.1 Experimental procedure

The material investigated was a ferritic Fe-9wt.%Cr model alloy with the chemical composition of Fe-9.04wt.%Cr-0.005wt.%C. It has an average grain size of 180 μm (determined from electron backscatter diffraction (EBSD) measurement) and no significant preferred orientation [18]. The nominal gauge dimensions of the tensile specimen were $5 \times 1.2 \times 0.5$ mm and the total length of 16 mm.

Neutron irradiation was performed at the Advanced Test Reactor (ATR) at the Idaho National Laboratory. Two irradiated specimens were examined, one irradiated to 0.01 dpa at 300°C and the other irradiated to 0.01 dpa at 450°C, both under a dose rate of 1×10^{-7} dpa/s. The activity of each sample was less than 5 mR/h at a distance of 30 cm. Before being transported to the APS, each irradiated specimen was individually mounted into a gripping device and enclosed in a double-layered Kapton containment at an ANL radiological facility. Irradiated sample assemblies were transferred in lead-shielded containments to the APS 1-ID beamline. The unirradiated Fe-9wt.%Cr specimen was mounted in the same type of gripping device without the Kapton enclosure. At the beamline, the grip assembly was installed into the *iRadMat* apparatus. Tensile tests were performed at room temperature at a nominal strain rate of $2 \times 10^{-5} \text{ s}^{-1}$. The WAXS and SAXS signals were recorded simultaneously and continuously during the tensile test.

The X-ray energy was 123 keV (wavelength 0.01011 nm). The WAXS detector calibration was performed with a CeO_2 NIST standard (SRM 674b). The sample to detector distance was nominally 2,635 mm, and 9 Debye-Scherrer rings were recorded. The size of the incident beam was 0.1 mm $\times$ 0.1 mm. Because of the coarse-grained structure and small size of the irradiated tensile specimen, the illuminated volume contained only a few grains, which made the diffraction data analysis difficult with traditional powder-averaged techniques. To obtain higher grain statistics in the diffraction data, the specimen was translated along X_L with a step size of 0.2 mm and Y_L with a step size of 0.4 mm with respect to the beam to interrogate 30 volumes of material along the gauge length. The resulting 30 diffraction images per GE detector were summed to produce a more powder-like data from approximately 100 grains. Due to the slow strain rate of the tensile test, the accumulated strain during these 30 exposures was about 1% strain, yielding a sufficient data acquisition rate. During tensile deformation, the constituent grains developed intragranular misorientation and form subgrains, thereby improving the diffraction statistics with increasing deformation.

Figure 14 shows a series of 2D diffraction patterns acquired by one of the WAXS detectors (GE1) at different deformation levels of the unirradiated specimen, each of which is a sum of the diffraction images from 30 measurements. As can be seen, prior to *in situ* deformation even 30 volumes cannot produce a powder-like pattern because of the small number of interrogated grains. With increasing strain, more powder-like data can be obtained due to broadening of diffraction spots, particularly in the azimuthal direction. The 1D profiles were obtained by integrating over $\pm 15^\circ$ azimuthal angle of the WAXS patterns in GE1 (marked by the solid lines in Fig. 14) in the direction nominally parallel to the tensile loading axis (marked by the dotted lines in Fig. 14). The same was done for the patterns recorded by GE4 for the direction nominally perpendicular to the loading axis (i.e. the transverse direction). The 1D diffraction profiles were then fitted using pseudo-Voigt functions yielding peak positions in the two directions. For peak broadening analysis, it was assumed that the broadening was homogeneous in the entire azimuthal range (i.e. no directional preference) and the entire azimuths covered by GE1 and GE4 were used. Since the diffraction rings deviate from circles due to deviatoric strains, the patterns were not integrated over the entire azimuth initially; instead, they were first caked into 20° azimuthal slices, and

then integrated into individual 1D profiles. Those profiles were fitted with pseudo-Voigt functions to obtain the integrated intensity, position and widths (represented by full-width at half maximum or FWHM) of each peak of interest. The FWHM results were summed over all measured azimuthal slices, using integrated intensity weighting. This maximized the number of grains in the FWHM analysis resulting in improved goodness of fits using the modified Williamson-Hall method, as discussed later. The instrument contribution to the broadening ($B_{\text{instrument}}$) was quantified using a LaB6 NIST standard (SRM 660a). It was subtracted from the measured FWHM (B_{total}) to get the actual sample broadening using $B_{\text{sample}}^2 = B_{\text{total}}^2 - B_{\text{instrument}}^2$, as a result of deconvolution, because the line profile is close to Gaussian (fraction of Gaussian in the pseudo-Voigt fitting is above 0.8 for most peaks).

Figure 14. 2D diffraction patterns from the unirradiated Fe-9Cr specimen on the GE1 detector at different deformation levels.

3.2 Results

The data from *in situ* WAXS measurements of the uniform tensile deformation up to the ultimate tensile strength (UTS) are presented below. These coarsen-grained single-phase specimens did not scatter strongly in SAXS during uniform deformation.

Macroscopic stress-strain curves

Figure 15 compares the stress-strain curves for the unirradiated and irradiated Fe-9Cr samples, with the solid symbols representing engineering stress-strain data and the open ones for the corresponding true stress-strain data (until the maximum load). Irradiation-induced hardening and reduction in ductility are significant even at this low dose. Hardening is more pronounced in the 450°C/0.01 dpa sample than in the 300°C/0.01 dpa sample. The 0.2%-offset yield stress, the UTS, the work hardening exponent, and the uniform and total elongations for each sample are listed in

Table 1.

Figure 15. Stress-strain curves obtained from *in situ* room-temperature tensile tests of the unirradiated and two irradiated Fe-9Cr specimens.

Table 1. Tensile properties obtained from the macroscopic stress-strain curves of the unirradiated and two irradiated Fe-9Cr specimens.

Sample	US (MPa)	UTS (MPa)	Work-hardening exponent	Uniform elongation (%)	Total elongation (%)
Unirradiated	115	222	0.380	20.8	28.7
300°C/0.01dpa	200	282	0.254	16.6	18.4
450°C/0.01dpa	280	338	0.261	16.1	22.7

In situ WAXS during tensile tests

WAXS measurements of the undeformed samples gave a lattice parameter of 2.8695 ± 0.0006 Å for the unirradiated specimen, 2.8720 ± 0.0009 Å for the 300°C irradiated, and 2.8722 ± 0.0008 Å for the 450°C irradiated specimens, respectively. These numbers were obtained from averaging the lattice parameters given by all the {hkl} families in each sample. The relatively large uncertainty, compared to the instrumental resolution of 1×10^{-4} strain from fitting the CeO_2 standard, was due to the large scatter in {hkl}-specific lattice parameters primarily from the residual strains.

Figure 16(a) shows the lattice strains ϵ_l as a function of the true stress up to the UTS for six {hkl} reflections in the loading and transverse directions in the three samples, calculated using $\epsilon_l = (d - d_0)/d_0$, where d_0 is the {hkl}-specific d-spacing prior to deformation and d is the instantaneous d-

spacing during deformation. Figure 16(b) shows the lattice strains in the loading direction as a function of true strain. In both figures the open symbols are for the loading direction and the solid symbols for the transverse direction. As expected, the lattice strains measured along the loading direction are tensile and the lattice strains measured along the transverse direction are compressive for all three samples as a result of the Poisson's effect. The ratio between the lattice strains along the loading and transverse directions is approximately 0.3 in the elastic regime.

Although the macroscopic stress-strain curves for the three samples show similar deformation behavior, the X-ray measurements of the lattice strain show different characteristics, either when

comparing different reflections within one sample or comparing between samples for a given reflection. For a given sample, the load redistribution between grains is manifested by the elastic anisotropy, i.e. the difference in stiffness of the reflection [19]. The {200} lattice strains are the largest while the {222} lattice strains are the smallest in the loading direction before the macroscopic yield stress. The lattice strains for other crystallographic planes mostly lie between the {200} and {222} lattice strains. These trends are consistent with commonly recognized the stiffest and most compliant directions in bcc metals. In the transverse direction, this relationship largely holds true in the 450°C irradiated sample, but is not so clear in the unirradiated and 300°C irradiated samples, which could be attributed to the uncertainties in determining d_0^{hkl} of the coarse-grained samples. It is also noted that lattice strains in the loading and transverse directions increase monotonically up to the macroscopic yield stress where linearity breaks down and inflections of the lattice strain vs. the macroscopic stress are evident. This is an indication that the crystal stresses are redistributing or reorienting [20].

When comparing between the samples, it is noted that the lattice strain evolution in the plastic deformation regime shows very different characteristics in the 450°C irradiated sample from the unirradiated sample, as shown in Fig. 16(b), where the deformation state is presented in terms of macro strain rather than stress for the loading direction. In the 450°C irradiated sample, after ~9% macroscopic strain the lattice strains all decrease, which is also observed in the 300°C irradiated sample after ~12.4% strain, but not in the unirradiated sample.

Figure 16. (a) Lattice strain vs. true stress for six {hkl} reflections in the three Fe-9Cr samples, showing for both the loading direction (open symbols) and the transverse direction (solid symbols). (b) Lattice strain vs. true strain for the loading direction.

Figure 17 shows the FWHM values of the diffraction peaks as a function of true strain (starting from ~4%) for the same six reflections shown in Fig. 16. The FWHM values increase with macroscopic

strain, indicating a high level of nano-scale activities due to plastic deformation. The modified Williamson-Hall (W-H) analysis was performed to understand the FWHM evolution. According to the diffraction theory, the size broadening is independent of the diffraction vector length g_{hkl} ($g_{hkl} = \frac{1}{d_{hkl}}$, d_{hkl} is the d-spacing of the $\{hkl\}$ plane) while strain broadening increases with g_{hkl} [21]. The modified W-H theory takes into account the anisotropy in strain broadening due to geometric factors controlling dislocation movement on different $\{hkl\}$ planes and relates the strain broadening *linearly* with g_{hkl} [22] (under the condition that the diffraction angle θ is small so that $\cos \theta \approx 1$, which is the case here with high-energy X-rays). The slope of this linear relationship is proportional to the square root of the mobile dislocation density ρ_l (in particular, the dislocations that do not belong to sharp boundaries [23]), and the intercept is inversely proportional to the coherent scattering domain size D by the Scherrer equation $\beta = 0.9\lambda/D$, where β is the intercept and λ is the wavelength. The anisotropy due to geometry factors is accounted for by including $\{hkl\}$ -dependent dislocation contrast factors C_{hkl} using $g_{hkl} \cdot C_{hkl}^{0.5}$. Values for C_{hkl} for edge type $\{110\}\langle 111 \rangle$ and screw type $\langle 111 \rangle$ dislocations in BCC-Fe can be calculated using the software ANIZC [24] with input parameters specified in [25]. The effective C_{hkl} used in the analysis is a linear superposition of values from these two types of dislocations weighted by their density fraction. To find the optimum C_{hkl} , the modified W-H fitting using the C_{hkl} with the screw/edge ratio ranging from 0 to 1 was performed with 0.01 step size, and the C_{hkl} that gave the highest R-square value was selected, giving the optimum edge/screw dislocation fraction. In this way, the evolution of the edge/screw dislocation fraction can be tracked with deformation.

The slopes, the coherent scattering domain sizes, and the screw dislocation fractions obtained from the modified W-H analysis are plotted as a function of true strain for the three Fe-9Cr samples in Fig. 18. The R-square values from all the fittings are above 0.92 and most of them are above 0.95. Domain size values at low deformation levels and greater than $1.5 \mu\text{m}$ are removed from the plot, because they may not be statistically accurate.

It was found that the deformation in each sample progresses in distinct stages. For example, in the unirradiated sample, between 7.5% and 11.5% strain, the dislocation density (reflected by the slope values in Fig. 18(a)) increases slowly, the screw dislocation fraction is almost unchanged, but the domain size decreases quickly. Beyond 11.5%, the dislocation density increases quickly as well as the screw dislocation fraction, but the domain size reaches a plateau. In the 450°C irradiated sample, before 9% strain the dislocation density increases quickly as well as the screw dislocation fraction, while the domain size remains relatively stable. After 9% strain, the dislocation density decreases slowly as well as the screw fraction, while the domain size decreases quickly.

Figure 17. FWHM vs. true strain for six $\{hkl\}$ reflections in the three Fe-9Cr samples.

Figure 18. Results from modified Williamson-Hall analysis of the three samples as a function of true strain: (a) the slope; (b) the coherent scattering domain size; (c) the screw dislocation fraction.

The coherent scattering domain size at the end of uniform deformation in the unirradiated sample given by the modified W-H analysis was verified by post-mortem TEM examination of the unirradiated specimen. A TEM sample was prepared using the Focused Ion Beam lift-out method. It was extracted from the uniformly deformed gauge section of the fractured unirradiated sample in cross section, with the long edge parallel to the gauge width. Figure 19 shows a bright-field image of cells/subgrains formed during tensile deformation. The mean size of dislocation cells measured by TEM is about $0.5 \mu\text{m}$, which is consistent with the coherent scattering domain size, $0.5 \mu\text{m}$ given in Fig. 18(b).

Figure 19. Bright field TEM image showing the dislocation cells formed during tensile deformation of the unirradiated Fe-9Cr specimen.

3.3 Discussion

1). Barrier strength of irradiation-induced defects

Neutron irradiation significantly increases the yield stress and the ultimate tensile strength of the Fe-9wt.%Cr alloy as shown in Fig. 15, which can be understood by the pinning of the dislocation motion by irradiation-induced defects. TEM observations of microstructures of as-irradiated samples [18,26] showed no visible irradiation-induced defects after neutron irradiation to 0.01 dpa at 300°C, while small dislocation loops with the average size of 3.8 nm and a density of $2.9 \times 10^{21}/\text{m}^3$ in the 450°C/0.01 dpa specimen. The majority of the dislocation loops have the Burgers vector of $a\langle 100 \rangle$. Atom probe

tomography showed no phase separation in either of the two irradiated specimens. WAXS measurements of the samples before tensile deformation showed that irradiation resulted in an increase in the lattice constant of 0.0025 Å in the 300°C irradiated sample, and 0.0027 Å in the 450°C irradiated sample, implying that the dislocation loops are predominantly interstitial-type and the irradiation-induced vacancies may form sub-microscopic voids which do not affect the lattice constant [27].

It is widely recognized that dislocation loops can pin the dislocation motion by either contact or elastic interaction, which leads to an increase in the yield stress and a reduction in ductility [28]. The barrier strength of dislocation loops, α can be estimated using the dispersed barrier model [29]:

$$\Delta\sigma = M\alpha\mu b\sqrt{Nd} \quad (2)$$

where $\Delta\sigma$ is the yield stress increase after irradiation, M is the Taylor factor (3.06 for BCC), μ is the shear modulus (8.3×10^4 MPa at room temperature), b is the magnitude of the loop Burgers vector, and N and d are irradiation-induced loop density and diameter, respectively. For the 450°C irradiated sample, the barrier strength α of the TEM-visible dislocation loops was estimated to be ~0.68 for the 165 MPa increase in the 0.2%-offset yield stress determined from the macroscopic stress-strain curves. In this study, the application of *in situ* WAXS technique provides a method in estimating the micro-yield stress using the lattice strain – macroscopic strain curve. More specifically, for both the unirradiated sample and the 450°C irradiated sample, the micro-yield stress was determined from the first deviation from linearity in the lattice strain-true strain curves in Fig. 16(b). The micro-yield stress was estimated to be 101 MPa for the unirradiated sample and 197 MPa for the 450°C irradiated sample. Therefore, the irradiation-induced micro-yield stress increase was 96 MPa, giving an estimated α of ~0.40, which is significantly lower than the value calculated from the 0.2%-offset yield stress. Zinkle and Matsukawa [30] attributed the unusually high barrier strengths obtained from macroscopic stress-strain curves for visible defect clusters to submicroscopic nanovoids in bcc metals. Our work shows that the α value is also sensitive to the measurement of the yield stress. In the 300°C/0.01dpa specimen, almost no loops were visible under TEM, and therefore a lower yield stress increase compared with the 450°C irradiated sample. Its increase in yield strength may be explained by the source hardening from the locking of grown-in dislocations by the submicroscopic defects [31].

2) Effect of irradiation on lattice strain evolution during plastic deformation

The lattice strain evolution in the plastic deformation regime shows significant differences between the unirradiated and the irradiated samples, as shown in Fig. 16. To better reveal the lattice strain evolution in the plastic regime, Fig. 16(b) was re-plotted in Fig. 20 by taking the d-spacings at ~4% strain in each reflection as the reference d-spacings to calculate the {hkl}-specific lattice strains. This eliminates the elastic anisotropic effect and reduces the data scatter from the diffraction patterns not being “powder-like” at lower deformation levels. We denote these lattice strains as *pseudo* lattice strains and they are plotted as a function of plastic strain in Fig. 20 in solid symbols.

The equivalent elastic strains were calculated using the method A reported by Daymond [32], which weighs the strain contribution from individual reflections by α_{hkl} , defined as:

$$\alpha_{hkl} = T_{hkl}m_{hkl}E_{hkl}/\bar{E}, \quad (3)$$

where T_{hkl} is the texture index, which is unity for the untextured alloy in this study, m_{hkl} is the multiplicity of the {hkl} reflection, E_{hkl} is the diffraction elastic constant for the {hkl} reflection, which is measured as the slope in the elastic deformation regime in the lattice strain- stress curve in Fig. 16(a), and $\bar{E}$ is the bulk modulus. The equivalent elastic strain $\bar{\epsilon}$ is then calculated as:

$$\bar{\varepsilon} = \frac{\sum_{hkl} \alpha_{hkl} \varepsilon_{hkl}}{\sum_{hkl} \alpha_{hkl}}, \quad (4)$$

where ε_{hkl} is the instantaneous lattice strain of the $\{hkl\}$ reflection. The equivalent elastic strain is plotted as a function of plastic strain in Fig. 20 in open symbols. For the unirradiated sample it shows a steady increase with plastic strain, despite the fact that the $\{hkl\}$ -specific lattice strains are largely anisotropic. In contrast, for the 300°C irradiated sample, both the $\{hkl\}$ -specific lattice strains and the equivalent elastic strain show a decrease starting at 12.3%. For the 450°C sample this synchronized decrease start at 8.5%. It is also noted that the equivalent elastic strain and $\{321\}$ lattice strain show a similar trend with respect to the plastic strain in all three samples, implying that the 321 reflection is suitable for characterizing the bulk behavior of this alloy.

Figure 20. *Pseudo* lattice strains and the equivalent strains in the loading direction as a function of true strain for the three samples, taking the d-spacing of each $\{hkl\}$ reflection at ~4% strain to be the reference point.

3) Work hardening mechanisms

In situ WAXS data provides new insights into the work hardening mechanisms in the Fe-9wt.%Cr alloy, as revealed by the lattice strain data and the results from the modified W-H analysis. To better understand the WAXS data, Figure 21 is plotted by reorganizing the content in Figs. 18 and 20, with an emphasis on changes with plastic strain in individual samples. Note that the slope from the W-H fitting in Fig. 18 is replaced by $A\rho^{0.5}$ in Fig. 21, where A is a constant and ρ the dislocation density.

Based on the modified W-H theory [22], $A = \lambda \left(\frac{\pi m^2 b^2}{2} \right)^{\frac{1}{2}}$, where λ is the X-ray wavelength, m is a constant depending on the effective outer cutoff radius of dislocations, and b is the magnitude of the Burgers vector. The value of m is not being pursued here and A is assumed to be the same for the three samples. Dotted vertical lines in Fig. 21 define deformation regimes: for the unirradiated sample, two lines are proposed at 7.3% and 11.2% plastic strain; for the 300°C irradiated sample at 12.3%, and for the 450°C irradiated sample at 8.5%, respectively.

The first deformation regime in all three samples has strikingly similar characteristics: 1) the $\{hkl\}$ -specific lattice strains first show heterogeneous behaviors followed by a synchronized increase with the transition at 6.1% for unirradiated, 10% for 300°C irradiated and 7.2% for 450°C irradiated samples, respectively; 2) dislocation density shows a rapid increase with plastic strain at the same rate, i.e. $\frac{dA\rho^{0.5}}{d\varepsilon_p} = 2.0 \times 10^{-5}$ in all three samples, as marked by the green dotted lines; 3) coherent

scattering domain size is relatively unchanged in the two irradiated samples with some data scatter. The lattice strain increase can arise from the dislocation pile-up at grain boundaries, and the decrease may be

attributed to the relaxation of intergranular stresses from slip transfer across grain boundaries [33]. The constant slope in the dislocation density – plastic strain curve indicates a constant work hardening rate ($\frac{d\sigma}{d\varepsilon} \propto \frac{d\sqrt{\rho}}{d\varepsilon}$), i.e. “stage II” athermal work hardening [34,35], and the work hardening rate was unaffected by irradiation. The minimal change in the coherent scattering domain size implies insignificant dislocation patterning and dislocation substructure formation.

Figure 21. Illustration of deformation stages in the three samples. Dotted vertical lines mark the transition points between stages. Dotted inclined lines mark the slope of the dislocation evolution curves in the first stage of each sample.

The effect of irradiation is most exemplified by a significantly higher density of dislocations and fraction of screw dislocations in the 450°C irradiated sample relative to the unirradiated sample in stage II. Before transitioning to next deformation stage, the dislocation density is the highest in the 450°C irradiated sample ($A\rho^{0.5} = 3.81 \times 10^{-4}$), lower in the 300°C irradiated sample ($A\rho^{0.5} = 3.57 \times 10^{-4}$), and the lowest in the unirradiated sample ($A\rho^{0.5} = 2.92 \times 10^{-4}$). The higher density of mobile dislocations in the irradiated specimens may be understood by considering the interactions of dislocations with irradiation-induced defects. As revealed by TEM during *in situ* ion irradiation and straining of an stainless steel specimen [36], dislocations moved relatively slowly and in a wave-like motion, in contrast to fast and smooth dislocation motion observed in a comparable unirradiated specimen. This difference was attributed to pinning effects from the irradiation induced defects. The jerky flow in the irradiated specimen increased the overall length of dislocation segments. The higher fraction of screw dislocations observed in the 450°C irradiated specimen in Fig. 18(c) can also be explained by dislocation – irradiation-induced loop interactions. When the loop pin a mobile dislocation, the line bows out forming screw segments, resulting an increase in length (i.e. an increase in line density) and the screw dislocation fraction. This dislocation behavior has been demonstrated in simulations of various situations of dislocation-loop interactions [29,37-39]. The submicroscopic radiation defects in the 300°C specimen seem to have minimal effect on the screw dislocation fraction, probably due to the defect annihilation by dislocation lines [29].

While the first regimes in all samples indicate active dislocation multiplication with little effect on the coherent scattering domain size, the second regimes is manifested by significant reductions in coherent scattering domain size in all three samples. The lattice strains in the two irradiated samples show synchronized decrease but are heterogeneous in the unirradiated sample. The dislocation density increases

slightly in the unirradiated, while decreases rapidly in the 450°C irradiated sample and relatively unchanged in the 300°C irradiated samples. Those phenomena suggest active dislocation cell formation and dynamic recovery, characteristic of working hardening “stage III”

To better understand the effect of neutron irradiation on dislocation density evolution, dislocation cell formation and their consequent work hardening behavior, the following empirical relationship is considered [40]:

$$\sigma = \sigma_0 + \alpha\mu b\rho^{0.5} + K\mu b/D \quad (5a)$$

where σ_0 is the friction stress, α and K are the constants, μ is the shear modulus, b is the magnitude of the Burgers vector, and D is the cell size. The work hardening rate is deduced as:

$$d\sigma/d\varepsilon = \alpha\mu b(d\rho^{0.5}/d\varepsilon) - K\mu b/D^2(dD/d\varepsilon) \quad (5b)$$

Stage II is dominated by the first term in Eq. (5b), which is the constant in all three specimens. In stage III, while cell formation occurred in both the unirradiated and the 450°C irradiated specimens at a similar rate, the dislocation density continues to increase (though at a much lower rate than in stage II) in the unirradiated specimen, as oppose to a decrease in the 450°C irradiated specimen, which leads to the overall reduced work hardening rate and an early onset of necking in the 450°C irradiated specimen. This greater dynamic recovery in the 450°C irradiated specimen may be attributed to its higher screw dislocation fraction and the resultant extensive cross-slip of screw dislocations. In the 300°C irradiated specimen, the “stage II” linear work hardening was extended to a larger strain, ~12%. Due to the lack of data points in stage III, the change in work hardening rate in the 300°C irradiated sample is speculated to be similar to the 450°C irradiated specimen, as they both have the synchronized reduction in lattice strains and the cell size.

After the second deformation regime (i.e. stage III), the two irradiated samples entered the necking stage. In contrast, the unirradiated sample exhibited further work hardening, showing a third regime, characterized by the rise in dislocation density and the plateau in domain size. The rate of dislocation density increase is smaller compared to that of the first regime. This may indicate the sample entered the work hardening stage IV at large strains. In the study of fcc metals, Rollett et.al. [34] explained the mechanism for stage IV being the dislocation debris accumulating inside the dislocation tangles or loose cell walls resulted from stage III. Their model predicted a constant strain hardening rate (i.e. a constant $\frac{d\sqrt{\rho}}{d\varepsilon}$), and its magnitude is smaller compared to that of the stage II. The constant strain hardening rate was also reported in X-ray line profile analysis by Ungar and co-workers [41,42]. Estrin et.al. [43] explained stage IV as a net result from the decreasing hardening from the cell walls as they became thinner and the increasing hardening from the accumulation of dislocations from the cell interiors, and predicted a decreasing hardening rate in stage IV. Our results show that the microstructural domain size remains constant in this stage while the hardening rate is also a constant, the value of which is about half of the rate in stage II, significantly higher than the prediction by Rollett et.al. [34]. The fact that the two irradiated samples exhibited localized deformation without the stage IV implies that the interaction between the dislocations and the irradiation-induced defects exhausts work hardening capability, resulting in the onset of instability at large strains.

While the macroscopic stress-strain curves are important for identifying the overall effect of the complex deformation processes, *in situ* X-ray diffraction is a sensitive technique which enables us to probe the mechanical deformation processes within a polycrystalline aggregate, revealing the microscopic mechanisms that are masked in the bulk response. In the effort to understand the effect of neutron irradiation to mechanical properties, this technique is demonstrated to be particularly valuable.

4 Effect of Irradiation on Deformation Behavior of Austenitic Alloys

Task 2 was to understand the deformation mechanisms in irradiated austenitic stainless steels, and the effect of alloying on radiation resistance of austenitic stainless steels. Two austenitic stainless steels were studied, namely, 316 stainless steel and a high-temperature ultrafine-precipitate-strengthened (HT-UPS) steel. 316 SS specimens neutron irradiated to 5.5 and 12 dpa at 300-315°C were tensile tested *in situ* at room temperature, and HT-UPS specimens neutron irradiated to 3 dpa at 400°C were tensile tested *in situ* at room temperature and 400°C, respectively. Below we will briefly describe the *in situ* tensile test results of neutron-irradiated 316 SS.

The 316 SS specimens were in the solution annealed condition with the grain size of 10 – 100 microns prior to irradiation. Neutron irradiation was conducted in the BOR-60 reactor at 300 - 315°C. Two neutron-irradiated 316 SS specimens were examined: (1) 5.5 dpa, and (2) 12 dpa.

The set-up for *in situ* tensile test with X-ray measurements is shown in Fig. 6. Tensile tests were carried out at room temperature with a nominal strain rate of 3×10^{-5} /s. The X-ray energy was 123 keV. Simultaneous WAXS and SAXS measurements were conducted during the tensile test with the sample to detector distance of 2,638 mm for the WAXS hydra array and 6,600 mm for the SAXS detector.

Figure 22 shows the engineering stress-strain curves recorded during the *in situ* tensile tests for unirradiated and two irradiated 316 SS specimens. Neutron irradiation significantly increased the yield stress of 316 SS and reduced its ductility. The 5.5 dpa specimen showed an upper/lower yield and an extended yield region followed by slight strain hardening before necking, while the 12 dpa specimen showed an extended elongation after the upper yield without strain hardening before necking. This is in contrast to the stress – strain behavior of the unirradiated 316 SS that shows a much lower yield stress, significant strain hardening, and a uniform elongation of ~100%.

Figure 22. Engineering stress-strain curves obtained from *in situ* tensile tests of unirradiated and irradiated 316 SS.

An interesting finding from the X-ray measurements during the *in situ* tensile test was deformation-induced martensitic transformation in both unirradiated and irradiated 316 SS specimens. As shown in Fig. 23(a), the bcc ferrite peaks were detected during tensile deformation in the unirradiated 316 SS specimen. The quantitative analysis of the XRD data showed that the ferrite fraction increased from 0

to 13% when the engineering strain increased from 0 to 100% (at the ultimate tensile strength), as shown in Fig. 23 (b). The ferrite fraction increased slowly with increasing strain initially, and then grew rapidly at a nearly constant rate when strain > ~75%.

Figure 23. (a) XRD lineouts of the unirradiated 316 SS specimen showing ferrite peaks with increasing strain, and (b) the ferrite fraction vs. engineering strain for the unirradiated 316 SS specimen.

Deformation-induced martensitic transformation was also observed in the irradiated 316 SS specimens. In contrast to the extensive uniform deformation and significant strain hardening observed in the unirradiated 316 SS specimen, plastic deformation occurred in a localized mode after the yield, and martensitic transformation occurred only in the deformed region behind the propagating deformation band, as shown in Fig. 24, where the gauge section of the 12dpa specimen was scanned with X-ray beams. The XRD scan along the gauge section showed that the deformation bands were initiated at one end of the gauge section, and propagated towards the other end along the gauge, and transformation of the austenite to the ferrite occurred in the deformed region only, but not in the undeformed area. The deformation-induced martensitic transformation provided a hardening mechanism that can compensate the deformation-induced geometrical softening, moving the deformation band forward to the undeformed region. In the necking region, severe localized deformation at the center of the neck induced the formation of a high fraction of ferrite, as shown in Fig. 25. In the 5 dpa specimen, the deformation bands spread across the entire gauge length before uniform deformation occurred. It is evident that deformation-induced phase transformation played an important role in extending the uniform elongation and ductility in the neutron-irradiated 316 SS specimens.

Figure 24. Diffraction patterns revealing propagation of localized deformation bands along the gauge of the irradiated (12 dpa) 316 SS specimen.

Figure 25. The distribution of deformation-induced ferrite along the gauge section of the irradiated (12 dpa) 316 SS specimen: (a) at 20% strain (before necking), and (b) at 34% strain (after necking).

5 Effect of Irradiation on Deformation Behavior of CASS (Duplex Microstructure)

The goal of Task 3 is to understand the deformation mechanism in a duplex cast stainless steel. Cast austenitic stainless steels (CASS) and stainless steel (SS) welds are structural materials used for components at the primary pressure boundaries and core internals of light water reactors (LWRs). Consisting of a dual-phase microstructure of delta ferrite and austenite, CASS alloys and SS welds are vulnerable to thermal aging embrittlement and neutron irradiation damage. Both TEM and APT analysis [44,45] showed significant microstructural changes with the precipitation of a high density, nanometer-sized G-phase and phase separation of α/α' in the ferrite phase, while minimal changes in the austenite phase after thermal aging for 10,000 h at 400°C in a CF-8 alloy.

The effects of aging-induced microstructural changes on the mechanical performance of CASS specimens were evaluated with tensile tests. Miniature flat tensile specimens with a gauge section of 0.8×1.5×7.6 mm were used. The tests were conducted with a strain rate of $1 \times 10^{-3} \text{ s}^{-1}$ at 20 and 315°C, respectively. Figure 26 shows the tensile results from the CF-8 alloy with 23% ferrite. While both yield stress (YS) and ultimate tensile strength (UTS) were increased after thermal aging, uniform elongations were reduced considerably at both temperatures. This large reduction in ductility indicates a significant effect of the deteriorated microstructures of the aged specimens, consistent with the observation in the fracture toughness tests of the CF-8 alloy. Compared with the room-temperature tests, both strengths and elongations are lower at 315°C, and the effect of test temperature is less evident for the aged sample.

Figure 26. Effects of thermal aging on tensile properties of CF-8 at 25 and 315°C.

To understand the embrittlement mechanism, the relationship between the deteriorated microstructures and plastic deformation of the CF-8 was investigated with high-energy X-rays. Wide-angle X-ray scattering (WAXS) measurements were performed *in situ* during the tensile tests on the unaged and aged specimens at room temperature. The tests were performed with a nominal strain rate of $1 \times 10^{-4} \text{ /s}$, and the WAXS data were taken with high-energy monochromatic beams of 123 keV. During the tensile test, the entire gauge length of the sample was scanned while rotating the specimen about its loading axis to illuminate as many grains as possible with maximum orientations. Figure 27 shows the evolution of the lattice strains of {111} and {110} in the austenite and ferrite, respectively during tensile deformation. The lattice strain of the aged ferrite was significantly higher than that of unaged ferrite, indicating a much higher local stress developed in the deteriorated ferrite phase. The differences in lattice strains between the austenite and ferrite increased with engineering strain, and were much larger in the aged than in the unaged specimens during the entire tensile deformation process. Incompatible lattice strains developed at the ferrite/austenite interface were much higher in the aged than the unaged specimens, suggesting a critical role of phase boundaries in the fracture of aged samples.

Figure 27. *In situ* wide-angle XRD measurements showing a significantly higher incompatible strain between the ferrite and austenite in the aged than in the unaged CF-8 alloy.

6 Summary

The objective of this project is to demonstrate the application of *in situ* high energy X-ray measurements of nuclear reactor materials under thermal-mechanical loading, to understand their microstructure-property relationships. The gained knowledge is expected to enable accurate predictions of mechanical performance of these materials subjected to extreme environments, and to further facilitate development of advanced reactor materials. The report provides detailed description of the *in situ* X-ray Radiated Materials (*iRadMat*) apparatus designed to interface with a servo-hydraulic load frame at beamline 1-ID at the Advanced Photon Source. This new capability allows *in situ* studies of radioactive specimens subject to thermal-mechanical loading using a suite of high-energy X-ray scattering and imaging techniques. We conducted several case studies using the *iRadMat* to obtain a better understanding of deformation and fracture mechanisms of irradiated materials. *In situ* X-ray measurements on neutron-irradiated pure metal and model alloy and several representative reactor materials, e.g. pure Fe, Fe-9Cr model alloy, 316 SS, HT-UPS, and duplex cast austenitic stainless steels (CASS) CF-8 were performed under tensile loading at temperatures of 20-400°C in vacuum. A combination of wide-angle X-ray scattering (WAXS), small-angle X-ray scattering (SAXS), and imaging techniques were utilized to interrogate microstructure at different length scales in real time while the specimen was subject to thermal-mechanical loading. In addition, *in situ* X-ray studies were complemented and benchmarked by *ex situ* characterization using advanced electron microscopy, atom probe tomography (APT) and micro/nano-indentation. The report presented *in situ* tensile test results on neutron-irradiated pure Fe, Fe-9Cr model alloy, 316 SS and CASS CF-8. These *in situ* experiments demonstrate the broad applications of the new capability in understanding several outstanding issues related to irradiated materials.

7 Publications and Presentations

Refereed publications:

Xuan Zhang, Chi Xu, Leyun Wang, Yiren Chen, Meimei Li, Jonathan Almer, Erika Benda, Peter Kenesei, Ali Mashayekhi, Jun-Sang Park, and Frank Westferro, “iRadMat: A Thermo-Mechanical Testing System for in situ High-energy X-ray Characterization of Radioactive Specimens,” (submitted).

Xuan Zhang, Meimei Li, Jun-Sang Park, Peter Kenesei, Jonathan Almer, Chi Xu, J. F. Stubbins, “In situ High-Energy X-ray Diffraction Study of Tensile Deformation of Neutron-irradiated Polycrystalline Fe-9%Cr Alloy,” (submitted).

X. Zhang, J. Park, J. Almer, M. Li “Characterization of Neutron-Irradiated HT-UPS Steel by High-Energy X-ray Diffraction Microscopy” J. Nucl. Mater. 471 (2016) 280.

B.K. Kim, L Tan, C Xu, Y Yang, X Zhang, M Li, “Characterization of Neutron-irradiated HT UPS Steel by High-energy X-ray Diffraction Microscopy”, Journal of Nuclear Materials, 471(2016).

J. Park, X. Zhang, H. Sharma, P. Kenesei, D. Hoelzer, M. Li, J. Almer, “High-Energy Synchrotron X-ray Techniques for Studying Irradiated Materials,” J. Mater. Res. 30 (2015) 1.

M. Li, L. Wang, J. Almer, “Dislocation evolution during tensile deformation in ferritic-martensitic steels revealed by high energy X-rays,” Acta Mat. 76 (2014) 381.

Presentations:

Xuan Zhang, et. al., “In-situ X-ray Study of Tensile Deformation in Neutron-Irradiated 316 Stainless Steel,” 2016 Materials Research Society Fall Meeting and Exhibit, Boston, MA, US, 11/27/16-12/02/16.

Xuan Zhang, et al., “3D Study of Deformation Behavior in Neutron Irradiated Fe-9Cr Alloy,” 65th Annual Conference on Applications of X-ray Analysis, Chicago, IL, US, 08/01/16-08/05/16.

Xuan Zhang, et. al, “In-situ and 3D X-ray Diffraction Study of Deformation Behavior in Neutron-Irradiated Fe-9%Cr Polycrystal, ” 2016 Nuclear Fuels Structural Materials Topical Meeting held at 2016 American Nuclear Society Annual Meeting, New Orleans, LA, US, 06/12/16-06/16/16.

Chi Xu, et. al., “In-situ High Energy X-ray Characterization of Tensile Deformation of Advanced Austenitic Stainless Steels,” 2016 Nuclear Fuels & Structural Materials Topical Meeting held at 2016 American Nuclear Society Annual Meeting, New Orleans, LA, US, 06/12/16-06/16/16.

Meimei Li, et al. “Understanding Radiation Damage and Deformation Dynamics in Neutron Irradiated Steels with High-Energy X-rays,” 2016 MRS Spring Meeting & Exhibition, Phoenix, AZ, March 28 – April 1, 2016 (invited).

Xuan Zhang, et al., “In-situ High-Energy X-ray Study of Neutron Irradiation Effect on Tensile Deformation Behavior of an Fe-Cr Model Alloy,” 145th TMS Annual Meeting and Exhibition, Symposium: Accelerated Materials Evaluation for Nuclear Application Utilizing Test Reactors, Ion Beam Facilities and Integrated Modeling, Nashville, TN, US, 02/14/16-02/18/16.

Xuan Zhang, et. al., "In-situ Synchrotron X-ray Study of Neutron-Irradiated Steels during Tensile Tests," MRS 2015 Fall Meeting, Boston, MA, Nov 29-Dec 4, 2015.

Xuan Zhang, et al., "Characterization of Neutron-Irradiated HT-UPS Steel by High-Energy X-ray Diffraction Microscopy," 144th TMS Annual Meeting and Exhibition, Symposium: Characterization of Nuclear Reactor Materials and Fuels with Neutron and Synchrotron Radiation, Orlando, FL, US, 03/15/15-03/19/15.

Meimei Li, "Characterization of nuclear reactor materials using synchrotron radiation," NuMat 2014, The Nuclear Materials Conference, Clearwater Beach, FL, Oct. 27-30, 2014.

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