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Addendum to "Capability Needs for Irradiated and Radioactive Materials Research: Ad Hoc Committee Summary Report"

July 2023

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EXECUTIVE SUMMARY

The inherent radioactivity and heterogeneous microstructures of nuclear materials and fuels make them challenging research and engineering targets. Advanced light sources have enabled significant underpinning scientific contributions to the current understanding of structural and fuel cladding material microstructure and properties, as well as the development of structure-property relationships. These facilities could potentially help advance Department of Energy Office of Nuclear Energy (DOE-NE) mission priorities by enabling an underpinning understanding of technically important challenges such as irradiation-induced embrittlement and swelling, stress corrosion cracking and corrosion in extreme environments, reactor pressure vessel (RPV) steel aging, nuclear fuel characterization, and nuclear waste form optimization, and they could play a major role in the development of a material performance matrix to aid in designing new materials to meet the needs of advanced reactor concepts. However, delivery of an underpinning understanding is the mission of the DOE Office of Science (DOE-SC). While filling such knowledge gaps is important, efforts in this direction should not be confused with DOE-NE's strategic, technology-focused mission goals of enabling the continued operation of existing U.S. nuclear reactors, supporting the deployment of advanced nuclear reactors, developing advanced nuclear fuel cycles, and maintaining U.S. leadership in nuclear energy technology.

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ACRONYMS

ANKA	Angströmquelle Karlsruhe
DOE-NE	Department of Energy Office of Nuclear Energy
DOE-SC	Department of Energy Office of Science
EPMA	electron probe microanalysis
LWR	light-water reactor
MARS	Multi-Analyses on Radioactive Samples
NSLS-II	National Synchrotron Light Source II
RPV	reactor pressure vessel
SAXS	small-angle x-ray scattering
SOLEIL	Source optimisée de lumière d'énergie intermédiaire du LURE
SS	stainless steel
TRISO	tri-structural isotropic
XAS	X-ray absorption spectroscopy
XES	X-ray emission spectroscopy
XPD	X-ray powder diffraction
XRD	X-ray diffraction
XRF	X-ray fluorescence
WAXS	wide-angle X-ray scattering

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1. INTRODUCTION

The mission outlined in the terms of reference for the Ad Hoc Committee on Capability Needs for Irradiated and Radioactive Materials Research highlighted four questions to be considered, the last one being: [1]

- Can structural and microstructural characterization utilizing synchrotron technologies be used to address the recognized knowledge gaps, and if yes, are these the optimal capabilities to be used, or are there more appropriate capabilities?

While this question was considered in-depth by the committee, the only time it is referenced in the summary report is in the **Critical Capability Gaps** section, in the context of discussing **Gap 1: Radiological Facilities for Irradiated Material and Fuel Studies**. The report states: [2]

> *The committee feels that it is essential to be able to handle materials with considerable activity as well as fuel samples with reasonable alpha radiation emitting content (i.e., not just "a few rem at 30 cm"). This emphasis highlights historical, continuing, and ongoing issues with access to cutting-edge science capabilities at DOE Office of Science (SC) user facilities, particularly synchrotrons and other light sources, as well as neutron scattering capabilities, that need to be addressed. The committee also noted that the neutron beam lines at NIST are considerably more user friendly than comparable facilities at the DOE-SC supported SNS and HFIR. Performing experimental studies on irradiated structural materials and nuclear fuels at DOE-SC capabilities is a formidable, in fact almost impossible, task. It is important to facilitate and increase access to DOE-SC User Facilities for research utilizing irradiated materials and nuclear fuels of significantly higher activity and with alpha radiation emitting content.*

This question is worthy of amplification. Many nuclear materials are heterogeneous at the sub-micrometer scale. These heterogeneous features determine the physical and chemical properties of the materials—and consequently material behavior and performance at the macroscale. This dependence necessitates spatially resolved characterization, which is most often performed using x-ray photon or electron probes featuring state-of-the-art resolution at the 10^{-9} to 10^{-10} m length scale.

2. X-RAY METHODS FOR CHARACTERIZING NUCLEAR MATERIALS

Interactions between x-rays and matter (e.g., absorption, diffraction/scattering, and fluorescence) allow for a broad range of non-destructive, sub-micrometer-resolution characterization methods that can provide both qualitive and quantitative information. Examples of these methods include: [3]

- Elemental composition distribution – x-ray fluorescence (XRF)
- Local oxidation state chemistry, local coordination and symmetries, and interatomic distances – x-ray absorption spectroscopy (XAS)
- Electronic structure – x-ray emission spectroscopy (XES)

- Structure in crystalline materials and on structural levels in disordered systems – x-ray diffraction (XRD) and scattering techniques such as wide-angle x-ray scattering (WAXS) or small-angle x-ray scattering (SAXS)
- Electron density distribution – coherent diffraction.

Furthermore, the high penetration depth of x-ray photons in both liquids and solids enables in situ and in operando studies, as well as 3-D tomographic analyses.

The signal-to-noise ratio and spatial resolution of an x-ray characterization experiment (i.e., the quality of the data obtained) are functions of the source brilliance and the acquisition time. Tabletop x-ray microscopes and laboratory x-ray microbeam systems featuring a beam diameter of about 10 micrometers enable 2-D and 3-D studies of the grains in a material, the embedded heterostructures, and the material microdomains. Synchrotron light sources produce x-ray beams with nanometer resolution, as well as photon fluxes that are many orders of magnitude greater than those in laboratory systems.

3. USES OF SYNCHROTRON METHODS IN NUCLEAR ENERGY SCIENCE

One of the biggest challenges in predicting the effects of irradiation and other types of extreme environments on material performance is determining the mechanistic relationship between the stressors and microstructural damage, then elucidating how microstructural changes affect macroscopic properties and material behavior. A material's mechanical response reflects competing dynamic processes that occur at both the nano- and microscale. In situ synchrotron characterization techniques have been used to investigate the combined effects of microstructural changes and stress states induced in nuclear materials as a result of radiation damage and elevated temperatures.

3.1 Structural Materials

Understanding a material's microstructure-property relations is central to evaluating its behavior and performance in a nuclear environment. High-energy synchrotron x-ray studies allow non-destructive determination of microstructure in bulk materials and this microstructure can be correlated with macroscopic property measurements. Appropriate techniques for this type of study include WAXS, SAXS, high-energy XRD microscopy (i.e., 3-D XRD microscopy), and x-ray computed microtomography. Specific examples of nuclear material studies include (but are not limited to) reactor pressure vessel (RPV) steels, [4] austenitic stainless steels (SSs) and advanced reactor materials, [5]-[8] zirconium-based cladding alloys, [9]-[12] silicon carbide, [13],[14] and nuclear graphite, [15],[16] as well as model alloys investigated for use in mechanistic model development. [17] Details on these materials are given in the paragraphs below.

Reactor pressure vessel steels. Extending the lifetime of the current nuclear reactor fleet to beyond 60 years is dependent on operational safety. RPV steels, the primary structural material used in light-water reactors (LWRs), undergo embrittlement that increases with increased neutron exposure. This phenomenon is evidenced by an increase in the ductile-to-brittle transition temperature and is caused by formation of nanometer-sized precipitates and solute-defect complexes. Current models of embrittlement do not include the effects of irradiation hardening and its mechanistic dependence on metallurgical—and irradiation—considerations. Radiation-induced precipitation in RPV steels was studied via small-angle neutron scattering and atom probe tomography; however, neither can provide crystallographic information on the precipitates. A combination of two advanced synchrotron x-ray techniques, SAXS and XRD, provided underpinning phase identification data on the nanoscale intermetallic precipitates that cause embrittlement. These studies were performed using the X-ray Powder Diffraction (XPD) beamline at the National Synchrotron Light Source II (NSLS-II) on samples taken from the Nuclear Science User Facilities University of California, Santa Barbara irradiation campaign conducted at Idaho National

Laboratory's Advanced Test Reactor. The data provide new insights that can be incorporated into predictive models for irradiated steel performance. [4]

Advanced reactor materials. Many current sodium fast reactor concepts utilize alloy 316, an austenitic SS, for structural applications. This is due to the strength and toughness of this alloy at high temperatures, as well as its sodium compatibility and corrosion resistance. One example of the utility of synchrotron techniques is that they can be utilized to investigate the relative performance of different SS grades. For instance, angle dispersive XRD has been used to compare the performance of two different SS variants—316 L(N) and a potential low-cost alternative, 304 L(N)—under sodium fast reactor operating conditions. Specifically, the effects of irradiation dose on XRD peak profiles were quantified and then related to irradiation-induced dislocation density. Next, the fundamental microstructural characterization was correlated with macroscopic changes in mechanical properties so as to gauge whether 304 L(N) may, for certain applications, be more cost-effective (i.e., cheaper) than 316 L(N) and other advanced materials. [6]

Synchrotron studies were also employed to obtain mechanistic information on phase transformations in irradiated 316 SS so as to provide an underpinning understanding of the effects of irradiation on mechanical properties, in addition to examining how microstructure and chemical heterogeneity affect the corrosion of 316 L(C) SS manufactured via laser powder bed fusion. [7]

Zirconium alloys. Zirconium alloys are widely used for fuel cladding and core structural materials in LWRs, due to their low neutron absorption cross-sections, favorable mechanical properties, and corrosion resistance. Despite these advantageous properties, zirconium alloys suffer from several drawbacks, including irradiation-induced growth (leading to dimensional instability), irradiation-enhanced creep, and hydrogen pickup. Examples of synchrotron x-ray studies of those zirconium alloys commonly deployed in nuclear environments include:

- Determination of the irradiation-induced microstructural evolution [9]
- Examination of microstructural changes in order to understand anisotropic irradiation growth [9]
- Characterization of the behavior of second-phase particles in neutron-irradiated Zr-4 alloys [10],[11]
- Provision of fundamental information on metal speciation at the oxide-metal interface and in the adjacent oxide and metal. [12]

Nuclear graphite. X-ray computed tomography is a powerful tool for observing deformation and fracture processes in materials such as nuclear graphite. For instance, recent 4-D synchrotron x-ray microtomography experiments documented the various stages of crack development in neutron-irradiated and radiolytically oxidized nuclear graphite. Such studies provide in-depth information on the microstructural factors that define strength and fracture and affect component reliability and reactor service lifetimes. [15]

3.2 Nuclear Fuels

Synchrotron characterization methods could potentially be employed to examine irradiation-induced microstructural changes in nuclear fuels so as to afford a mechanistic understanding of various aspects of in-reactor fuel performance. [18]

Uranium dioxide. The U.S. commercial nuclear fleet of LWRs is fueled with uranium dioxide, a fuel type that is also likely to be deployed in small modular reactors and certain advanced reactor types. Fracturing of UO_2 during reactor operation affects the fuel cladding integrity and can result in fission products being released into the coolant. This phenomenon has been studied extensively but remains only partially understood. The fuel form undergoes radial and hoop cracking, both of which are affected by temperature, stoichiometry, and microstructure. Synchrotron x-ray microdiffraction was employed to determine the development of local stress/strain within UO_2 fuel cracks so as to generate insights into the

development of strain and to support the development and validation of mechanistic models for incorporation into advanced fuel performance codes. [19]

Metallic fuels. Metallic fuels were deployed in fast breeder reactors up until 1960s and are currently the focus of renewed interest in regard to their potential application in advanced reactor concepts, thanks to their high thermal conductivity and potential recyclability. Various advanced characterization techniques have been used to examine the evolution of fuel behavior with neutron dose, including synchrotron x-ray microtomography, which is a non-destructive tool usable for characterizing the evolution of 3-D microstructures. The major advantages of this approach are (1) its ability to generate high-spatial-resolution information and (2) that it requires minimal sample preparation. For example, recent studies conducted utilizing this approach resulted in the identification of five stages of neutron-irradiation-induced pore evolution in U-10Zr metallic fuel: nucleation, growth, coalescence, interconnected porosity, and extended porosity. [20],[21] Though the data obtained from the synchrotron studies foster a valuable underpinning understanding of pore evolution, additional studies involving complementary techniques are needed to generate information on the metallic fuel's heterogeneous crystal structures and chemical compositions.

Uranium nitride. Uranium nitride (UN) is an advanced ceramic fuel currently being considered for use in Generation IV nuclear fission reactors. It is believed to have a number of desirable traits in comparison to traditional UO_2 fuel. Among these traits are a high uranium density, higher thermal conductivity at high temperatures, and a reduced swelling rate and lower fission gas release rate—in addition to the fact that many fission and actinide products form stable nitrides. However, due to the novel nature of this fuel, UN entails significant knowledge gaps. To address this, synchrotron extended x-ray absorption fine structure experiments were used to determine the local atomic structure and phase identities of unirradiated powder samples of both pristine and oxidized UN. These studies underpinned a larger investigation into the thermal behavior and thermal stability of UN. [22]

3.3 Nuclear Fuel Cycle

Synchrotron techniques are ideal for characterizing radioactive materials and elucidating chemical processes central to the nuclear fuel cycle. They provide high-spatial-resolution information on heterogeneous systems, along with energy resolution data on actinide speciation and oxidation states. The data obtained, including chemistry and bonding information, drive new theories, enable benchmarking, and support the development of modeling and simulation methods. [23],[24]

The extreme brilliance of synchrotron light sources and the deep penetration of x-rays (as compared to other probe beams) makes radiological containment of active samples possible. Valuable related techniques include x-ray absorption spectroscopy, 3-D imaging of elemental species, and the investigation of the f-element oxidation states and electronic structures. Most published examples of applying synchrotron techniques to fuel cycle research are related to actinide redox and coordination chemistry. Examples include determinations of:

- The local structure and oxidation state of U, Pu, and Am in irradiated mixed oxide (MOx) fuel, which influences the stability of the fuel form and consequently actinide release from the spent fuel.
- Radionuclide speciation—molecular form, chemical and physical environment—in nuclear waste forms so as to assess waste form stability and the potential radionuclide leaching rate.
- Actinide (and corrosion and fission product) structural information and speciation in molten salts, which are important for underpinning the data used in modeling the chemistry of pyro-processing and molten-salt reactors,
- Structure and speciation of actinides and lanthanides in aqueous and organic reprocessing solutions. [25]

It must be highlighted that the most important enabling requirement for studying actinides and nuclear fuels at synchrotron light sources is the availability of dedicated instrumentation that allows for broad characterization. Examples of synchrotron techniques applied to actinide and nuclear studies were comprehensively reviewed by both Shi et al. [23] and Denecke. [24]

4. ALTERNATIVE SPATIALLY RESOLVED MATERIAL CHARACTERIZATION METHODS

Laboratory-based tabletop alternatives to synchrotrons enable the study of radioactive materials—all without the cost and difficulty of transporting radioactive samples from a storage location to a synchrotron light source. [26] Furthermore, the ready accessibility of tabletop devices allows for experiments that reflect the behavior of radioactive materials over longer timescales than are feasible at multi-user synchrotron facilities. Examples of studies conducted using tabletop capabilities include:

- X-ray tomographic studies of the structure of tri-structural isotropic (TRISO) fuel particles, performed at a "spatial resolution of about 1.2 μm with good sensitivity to defects in the carbon buffer layer" [27]
- 3-D x-ray microscope characterization of the morphological evolution of TRISO-coated fuel kernels after high-temperature neutron irradiation [28]
- Trace element (impurity) characterization of nuclear materials, utilizing total reflection XRF. [29]

The fundamental scientific information available thanks to x-ray synchrotron light source capabilities is often accessible when using other complementary and synergistic characterization techniques (e.g., electron microscopy, positron annihilation spectroscopy, and atom probe tomography). [14] Each different characterization probe (i.e., photons, electrons, ions, and neutrons) offers both advantages and disadvantages. [3]

Lasers. Laser light-source techniques offer capabilities similar to those afforded by x-ray methods, the main difference being the wavelength of the photon. Photon beams from lasers have a greater coherence than those from x-ray synchrotrons, and these laboratory-scale instruments offer better power performance, efficiency, and optics. They do, however, present drawbacks: their lower energy (longer wavelength photon) limits spatial resolution and they offer limited energy (wavelength) tunability, elemental specificity, and penetration. The last of these drawbacks, limited penetration, essentially restricts studies to surfaces or optically transparent materials.

Electron beams. Most x-ray characterization methods have an electron microscopy equivalent: selected area or convergent beam electron diffraction – XRD; electron energy loss spectroscopy – XAS; energy dispersive x-ray spectroscopy – XRF; electron probe microanalysis (EPMA) – XES. The principal advantage of electron microscopy over corresponding synchrotron methods is cost. Dedicated scanning and transmission electron microscopes are included among the service capabilities of most university chemistry and materials science departments. Furthermore, a single laboratory-based microscope lends itself to multiple characterization techniques.

The spatial resolution of an electron microscope depends on the analytical technique employed; however, the electron probe beam can be focused to about 2 and <0.1 nm in most scanning and transmission electron microscopes, respectively. The measured signal may be transmitted electrons or emitted secondary electrons, visible photons, or x-rays. Two techniques, EPMA and Auger spectroscopy, are of particular importance in the study of nuclear fuels. High-energy electrons create core holes in the atoms of a material, and the decay of these holes can provide chemical composition and oxidation state information. Radiative decay of core holes is the basis of EPMA and the non-radiative route of Auger spectroscopy. EPMA enables qualitative chemical analyses of large-area samples but, due to its limited energy resolution, does not allow for discrimination between different oxidation states. There are two common observation methods: energy dispersive spectrometry and wavelength dispersive spectrometry.

Even the penetration of energetic electrons is limited, restricting electron microscopy to studies of the first few nanometers closest to the surface of a material or necessitating sample preparation activities that may alter the material's local properties. Another disadvantage stemming from the smaller penetration of electrons—as compared to photons—is the need to operate under a vacuum. This restriction limits in situ studies.

Ion beams. Ion beam techniques that utilize ion of energy from a few MeV to a few hundred MeV can be used to non-destructively investigate the elemental composition and the electronic properties of materials at sub-micrometer length scales. The spatial resolution of an ion beam analysis depends on the lateral straggling of the probe beam, and it is possible to obtain a level of precision on the order of 10 nm. As with x-ray methods, the various ion beam techniques are classed according to the ion-material interaction involved.

- Particle-induced x-ray emission (PIXE) is analogous to XES and EPMA, which utilize x-rays and electrons as the probe beam, respectively. PIXE involves detection of x-rays that are emitted as a result of electron transitions to fill core holes produced by ion bombardment.
- Secondary ion mass spectrometry and Rutherford backscattering spectrometry are used to measure the composition depth profile of a "bulk" material. The former determines the elemental profile by destructively sputtering the surface and then measuring the elemental, isotopic, and molecular composition of the ejected secondary ions through the use of a mass spectrometer. Rutherford backscattering spectrometry is generally used to measure heavy elements in a light matrix material. This non-destructive technique determines the elemental profile by measuring the elastic backscattering of a beam of He ions or protons.
- Elastic recoil detection analysis measures the elastic scattering of an ion beam in the forward direction (i.e., in transmission) in order to determine the elemental depth profile of a thin film of material. This technique is selective for light elements in a heavy matrix material.

Neutrons. Neutron beam techniques are generally considered complementary to x-ray and electron beam methods, though neutron characterization affords a lower resolution. Neutrons are exceptional bulk probes, having significant penetration in many materials. They are particularly valuable for measuring hydrogen and for their isotope specificity. The technical drawbacks to neutron methods are associated with resolution and include the focusing of the beam and the brightness of the source—a brightness many orders of magnitude lower than synchrotron x-ray sources. [30]-[32]

5. ADVANTAGES OF SYNCHROTRON CHARACTERIZATION METHODS

Compared to other advanced post-irradiation characterization methods, synchrotron light sources and tabletop x-ray sources have a unique property that facilitates non-destructive characterization of mesoscale samples. [33],[34] Namely, this property is the significant penetration of high-energy photons relative to, say, energetic electrons, and it allows for non-destructive 3-D structural studies of materials. Electron microscopy—as well as techniques such as photo-electron spectroscopy, atom probe tomography, and laser-induced breakdown spectroscopy—can only obtain "near surface" information. Obtaining a 3-D description necessitates the destructive milling of samples from different depths in a bulk sample. Furthermore, x-ray scattering studies provide more statistically reliable data than electron microscopy, which is limited to very small volumes.

In addition to affording the ability to interrogate the bulk of a material, synchrotron studies are feasible in a variety of extreme environments involving high and low temperatures, aggressive corrosive atmospheres, and highly ionizing and displacement-inducing radiations. It should be noted that not all advanced light sources are equal, and usability will vary from facility to facility.

Advanced light sources have made significant underpinning scientific contributions to the current understanding of structural and fuel cladding material microstructures/properties and the development of structure-property relationships. Such facilities could help advance Department of Energy Office of Nuclear Energy (DOE-NE) mission priorities by providing an underpinning understanding of numerous technically important challenge areas, including irradiation-induced embrittlement and swelling, stress corrosion cracking and corrosion in extreme environments, RPV steel aging, nuclear fuel characterization, and nuclear waste form optimization, and by playing a major role in the development of a material performance matrix to aid in designing new materials for meeting the needs of advanced reactor concepts.

Synchrotron x-ray sources offer tunable high-flux, high-energy beams. Highly focused beams and the broad energy range enable excitation of the K, L, M, N, and O edges of heavy elements. They offer a large variety of spectroscopic and microscopic characterization techniques. X-ray beams are highly penetrating, allowing for sample encapsulation.

The fundamental scientific information available thanks to synchrotron light source capabilities complements and synergizes with information obtained via other advanced characterization techniques such as electron microscopy, positron annihilation spectroscopy, and atom probe tomography. These various characterization methods should be considered partners—not competitors—in the enhancement of fundamental knowledge.

The mission of the Department of Energy Office of Science (DOE-SC) is to deliver an underpinning understanding of energy systems. While filling such knowledge gaps is important, efforts in this direction should not be confused with the following strategic, technology-focused mission goals of DOE-NE:

- Enable continued operation of existing U.S. nuclear reactors.
- Enable deployment of advanced nuclear reactors.
- Develop advanced nuclear fuel cycles.
- Maintain U.S. leadership in nuclear energy technology.

6. CHALLENGES OF RADIOACTIVE MATERIAL STUDIES AT SYNCHROTRONS

Exploiting the potential of x-ray synchrotron techniques for radioactive and contaminated materials is difficult and time consuming. Major experimental issues arise in regard to radioactive sample management in accordance with facility radiation protection regulations, shielding, and designing and performing in situ experiments under extreme conditions. Actinides pose health hazards to users as well as risks to facility operations, and thus require strict standards such as limiting the amount of material utilized in an experimental study, in addition to appropriate safety procedures for conducting examination, storage, and shipping activities. Despite efforts aimed at standardization, utilization of U.S. synchrotron capabilities to study radioactive materials is regulated by ad hoc safety review processes. For instance, the radioactive material limit at the Stanford Synchrotron Radiation Light-source is less than half the Department of Energy's limit for a Category 3 non-nuclear facility. In comparison, the limit at ANKA is 10^6 times the European Union exemption limit, allowing the study of 140 mg 243-Pu, 66 g 242-Pu, 200 mg 235-U, or 239-Pu.

Many international synchrotron facilities encourage the study of radioactive solution and solid samples. The samples are usually prepared in the user's (radiochemistry) laboratory, then shipped to the light source facility. Powdered samples are usually diluted with an inert solid. These limitations allow the synchrotron capability to fall under the classification of a low-hazard nuclear facility. Radioactive samples must be enclosed in two or more independent containments. Double containment usually avoids the need for a glovebox in the experimental hutch.

In situ experiments conducted under extreme conditions necessitate modular instrumentation that can temporarily operate on a beamline for the duration of a single experimental campaign. This approach is both time-consuming and inefficient.

Nuclear-friendly synchrotron capabilities are now available worldwide. Europe has several dedicated beamline capabilities for studying radionuclides: the Rossendorf Beamline at the European Synchrotron Radiation Facility in Grenoble, France; the Karlsruhe Institute of Technology's Institute for Nuclear Waste Disposal beamline at ANKA in Karlsruhe, Germany; the μXRF beamline at the Swiss Light Source in Villingen, Switzerland; and the Multi-Analyses on Radioactive Samples (MARS) beamline at the SOLEIL synchrotron in France. [35],[36]

Development of the "hot" MARS beamline was a result of collaboration between the French Alternative Energies and Atomic Energy Commission and SOLEIL. It has two end stations and is optimized for x-ray absorption spectroscopy (x-ray absorption near edge structure / synchrotron extended x-ray absorption fine structure) as well as powder diffraction (XRD), although XRF, high-energy resolution fluorescence detected XAS, XES, and μXAS/XRF/XRD are also available. The facility can characterize about 380 different radionuclides and is able to accept radioactive and radioactively contaminated samples by utilizing a "mobile biological protection module" to transport samples and dock with the analysis end station, thus preventing user exposure to radiation. The facility is comprised of a lead-shielded optics hutch, a control room, and an experiment area with radiation protection controls for handling and storing samples. The experiment area is air-tight and has a dedicated ventilation system. All samples must be prepared outside SOLEIL. They can be liquid or solid but must be double encapsulated and shipped in an air-tight transport package. Up to 185 Gbq of total storage is allowed at the beamline, with an individual sample limits of 2 GBq activity for γ emitters and 18.5 Gbq for α emitters. The maximum dose rate permitted in the experimental hutch area is 200 mrem/hr.

The first synchrotron experiments to characterize a sample of irradiated uranium oxide fuel via XRD were performed in November 2018 at the MARS beamline at the SOLEIL synchrotron in France. The high-resolution XRD study provided detailed information on the material structure/microstructure and the local changes across the diameter of a 4.5% enriched fuel pellet—changes that had occurred over the course of 5 years in a pressurized-water reactor. The aim of this experiment was to provide information for incorporation into mechanistic models.

In the U.S., the XPD beamline at NSLS-II is a multipurpose capability that can collect data at monochromatic x-ray energies of 40–70 keV. It offers researchers advanced synchrotron techniques such as XRD, pair distribution function analysis, and SAXS for the structural characterization of "materials for advanced nuclear energy systems," using an automated radioactive-sample handling capability. [37] The equipment was provided through DOE-NE funding and includes a robotic sample manipulator as well as sample magazines able to handle large numbers of samples in a short period of time. The XPD beamline can operate as a Radioactive Area (following IAW 10CF835 [38]) in which the maximum total allowable dose rate is 100 mrem/hr at 30 cm (i.e., each of the 100 samples may have an individual dose rate of 1 mrem/hr at 30 cm). All samples must be doubly encapsulated at the user's home institution prior to beginning an experimental campaign.

In addition, the in situ Radiated Materials end station module at the Advanced Photon Source (APS) beamline 1-ID was developed for WAXS and SAXS studies of (lightly) radioactive samples subjected to thermo-mechanical loading. [39] This capability consists of a radiation-shielded vacuum furnace in which the sample rotates under load. Initial studies examined the effect of neutron irradiation (0.01 dpa) on the tensile properties of a model Fe-9Cr ferritic steel. More recent studies have looked at austenitic 316 SS and high-temperature ultra-fine precipitation-strengthened SS irradiated to significantly higher doses. These studies and other applications of high-energy synchrotron x-ray techniques for examining structure-property relationships were reviewed by Zhang (2022).

Studies of nuclear materials have also been carried out at synchrotron facilities in the United Kingdom, [40] India, [18] China, and Brazil.

7. CONCLUSIONS

Current DOE-SC-operated synchrotron facilities have the demonstrated ability to facilitate underpinning fundamental studies on nuclear materials and fuels that are of interest to the academic and national laboratory nuclear science and engineering communities. In contrast, there are fundamental challenges to industrially impactful utilization of U.S. synchrotron light sources for conducting irradiated and radioactive materials research, the most important being that the Nuclear Regulatory Commission will not accept, for licensing purposes, any data derived from neutron-irradiated micro-samples or ion beam irradiations.

Studies at most synchrotron light-source end stations are, and must be, performed on samples whose sizes are constrained by their dose rate. The usual limit is a maximum dose rate of 5 mrem/hr at 30 cm, and approval of experiments is granted on a case-by-case basis rather than in accordance with a prescribed prescription. Currently, only one beamline in the U.S. (i.e., 28-ID-2 XPD at NSLS-II) has a special accommodation for higher-dose-rate samples, allowing for limited studies of samples with dose rates of up to 100 mrem/hr at 30 cm. Dose rate restrictions limit meaningful, industrially impactful studies on material properties/structures and component size samples. Use of a small number of samples for macroscale property investigation studies could potentially introduce anomalous stochastic effects if the sample set considered is inappropriate. In addition, nano- and microscale sample preparation (e.g., focused ion beam milling) may influence material composition and properties through deposition and secondary focused ion beam irradiation-induced damage.

In addition to dose rate issues, current synchrotron light source operating procedures in the United States are unamenable to any possibility of dispersion and contamination. This precludes experiments on:

- Structural materials in which samples are driven to break under compression or load.
- Powders and dispersions.

Anecdotally speaking, this makes all authorized experiments prohibitively time-consuming and complicated.

Studies to address industrially significant challenges such as creep and corrosion require experiments performed over an extended timeframe of days to weeks. Such studies are incompatible with the current model for allocating synchrotron time, as well as with the current operational practices at DOE-SC synchrotron light sources. Facilitating in situ studies on this type of challenge would require facilities in which multiple studies could be performed in parallel.

In conclusion, a paradigm shift in the approach to performing experiments on irradiated samples is required in order to enable facile utilization of DOE-SC synchrotron light-source capabilities that would help meet the industrially oriented mission goals of DOE-NE. For synchrotron light sources to be valuable to operators of the current fleet and vendors of advanced reactors, suitable logistics are required for experiments conducted on large irradiated/radioactive material samples, and for long-term in situ experiments carried out under different types of extreme environments.

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