Argonne NATIONAL LABORATORY

Report of the

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems

Advanced Photon Source
Argonne National Laboratory

April 2010

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EXECUTIVE SUMMARY

A workshop on the nuclear-energy community's synchrotron facility needs was held January 27-28, 2010, at Argonne National Laboratory. Convened at the request of Argonne's Advanced Photon Source (APS) and the Energy Sciences and Engineering (ESE) Directorates, the workshop was charged with assessing the potential contribution of synchrotron experiments to the scientific needs of the nuclear energy science and engineering community within the combined contexts of Argonne's Nuclear Energy Initiative and its Advanced Photon Source upgrade.

Over 100 people attended the workshop, including registrants from universities, government laboratories, industry, and funding agencies, that consisted of invited talks and a poster session. Attendees were asked to contribute to one of four breakout sessions; there they discussed their specific interests in nuclear energy systems and provided suggestions on how the APS might assist in their experimental needs. Under the direction of breakout chairs, each session provided summaries of its discussions as a scientific justification of their experimental needs, the contents of which are included in this workshop report. Also included are items of specific interest raised by the workshop participants concerning the sample quantities currently permitted at the APS. These items are summarized as Considerations in the report.

There is unanimous agreement that synchrotron studies hold enormous potential to answer questions central to all aspects of nuclear energy production, from simulation validation, to quantifying materials problems, to environmental remediation. Despite the hospitable environment provided by the APS to the nuclear energy community, major experimental challenges remain, some of which are specific to this community. These include managing radionuclides under U.S. Department of Energy (DOE) regulatory requirements, design and execution of *in situ* experiments (particularly those under extreme environments), and detecting synchrotron radiation under high background conditions.

The Workshop participants acknowledge that their recommendations will require an influx of resources and consequentially, additional financial support in order for the APS to address the community's needs. With this in mind, the workshop has generated a list of recommendations that support advanced nuclear energy systems research at the APS. These include:

Continued access to APS beamlines
The user community expressed their satisfaction with the APS' policy that allows radiological samples to be studied at any of the facility's beamlines, subject to the standard criteria for peer-reviewed proposals. Any new facilities or beamlines should not interfere with this current unfettered access policy.

Assistance with safety approvals for radioactive samples
Users expressed concern that the safety review process necessary for gaining radioactive sample access to the experimental floor is overly complex, often deterring potential users. The community recommends that increased information on safety-related requirements be made available through a more extensive webpage and more ready access to staff knowledgeable in this area.

Sample containment and hardware development
The users request APS technical and engineering assistance in the development of encapsulation and shielding hardware that is approved for work on the experimental floor. The designs should be pooled and available to any APS user for their use or modification on any of the beamlines.

Dedicated facility
The community strongly recommends that a dedicated facility be built for handling radioactive samples. A stand-alone building located adjacent to the APS would permit open sample manipulation and serve as a home base for the nuclear energy systems community, both of which were identified as needs by the user community.

Specialized beamlines
The construction of dedicated high-energy (> 60 keV) X-ray beamlines for experiments on radioactive samples would accommodate the relevant requirements associated with handling nuclear materials (radiation levels, safety, security) and provide capabilities unavailable elsewhere in the world. This recommendation is considered consistent with APS capabilities and its plans for an overall upgrade.

INTRODUCTION

WORKSHOP BACKGROUND

A workshop on the synchrotron facility needs of the nuclear-energy community was held January 27-28, 2010, at Argonne National Laboratory. Convened at the request of Argonne's Advanced Photon Source (APS) and the Energy Sciences and Engineering (ESE) Directorates, the workshop assessed the scientific needs of nuclear-energy science and engineering and addressed these needs in planning for the projected APS upgrade and Nuclear Energy strategies at the Laboratory.

This workshop built upon two earlier efforts to identify interests and concerns of parties interested in working with radioactive materials at the APS:

1. In 2008, a survey was taken to identify experimental needs for using synchrotron X-ray techniques to examine radioactive materials, and to encourage the community to think about the opportunities afforded by the APS to answer important questions within their fields. Based on 31 responses, it was determined that the community has a very broad interest in synchrotron-based studies, which are distributed over X-ray scattering, spectroscopy and imaging techniques in a manner similar to that seen in the broader synchrotron community.

2. This survey was followed by a small meeting held at Idaho National Laboratory the following summer that focused largely on issues associated with spent fuel and radiation damage. Based on this meeting, a letter of intent was submitted to the APS, requesting that a new CAT be formed that focuses primarily on materials aspects of nuclear energy systems and includes advanced capabilities for handling radioactive samples.

As of the end of the 2009 calendar year, there were 997 experimental safety approval forms (ESAFs) approved that identified radioactive samples in the list of hazards associated with those experiments. In the last three years (2007-2009), there were 241 experimenters associated with approved ESAFs that involved radiological samples on the APS floor.

With this background in mind, it was decided to invite a broad range of scientists and engineers to the APS to obtain a consensus of their current and projected needs and to present this consensus in the form of a report. The workshop was advertized on the APS website. Invitations were emailed to the 2008 survey respondents, the INL meeting participants, and all APS Users who had conducted experiments on radioactive materials over the last three years, as identified from the APS' experimental safety approval system. The workshop invitation and the brochure accompanying the email are included as Appendix I of this report. Online registration was handled through the APS.

REPRESENTATION FROM INTEREST GROUPS

Based on previous responses to the survey of the user community, the Idaho National Laboratory meeting attendance, and the relative number of "radioactive" experiments undertaken annually at the APS, it was estimated that there would be 50-60 workshop attendees. This proved to be a significant underestimation of the interest in this workshop. There were 115 registrants, forcing a last-minute change of venue to the APS auditorium in Building 401. One hundred-five registrants attended the meeting, with an approximate headcount of 97 attendees in the auditorium for one presentation.

The registration form included an inquiry about the interest/expertise of the respondent. Based on the answers received, the attendees were broken into four independent breakout groups to compile a consensus on their specific areas of technical interest. The breakout groups, listed with their session chairs, were:

1. Solutions and Amorphous Systems — Chairs: Kathryn Nagy (University of Illinois — Chicago) and Richard Wilson (Chemical Sciences and Engineering, Argonne). Seventeen people attended breakout sessions with this group.

2. Interfacial and Corrosion Studies — Chairs: Arthur Motta (Materials Science and Engineering, Nuclear Engineering, Pennsylvania State University) and Hoydoo You (Materials Science Division, Argonne National Laboratory). Twelve people attended breakout sessions with this group.

3. Radiation Damage Studies (including spent fuels) — Chairs: Todd Allen (University of Wisconsin-Madison) and Mark Kirk (Materials Science Division, Argonne). Thirty-seven people attended breakout sessions with this group.

4. Bulk Properties — Chairs: James Tobin (Lawrence Livermore National Laboratory) and Mark Bourke (Los Alamos National Laboratory). Twenty-four people attended breakout sessions with this group.

Numerous attendees expressed interest in more than one breakout session. Approximately 4 attendees identified themselves as having an interest in theory, simulation, and/or modeling. These attendees dispersed amongst the four established breakout groups.

Each of the groups produced a summary on their scientific challenges, opportunities, and synchrotron needs. These summaries are included in this report. Due to significant overlap with other sessions, the write-up for the Bulk Properties discussions is dispersed into other sections.

BREAKOUT SESSION SUMMARIES

1. SOLUTIONS, AMORPHOUS SOLIDS, AND ENVIRONMENT

Scientific Challenges and Opportunities

Development of advanced nuclear energy systems requires substantial fundamental research on solutions, amorphous solids, and environmental and biological systems. Solutions range from molten salts and metals used in pyrometallurgy and materials synthesis, to ionic liquids used in radionuclide separations, and to aqueous and organic solutions used in separations and essential to environmental processes.

Amorphous solids include not only true amorphous materials — glasses and gels — but also nanoparticles and colloidal aggregates previously deemed amorphous using traditional X-ray diffraction methods. These solids are important as 1) components of materials designed to contain nuclear wastes; 2) products of separations technologies; 3) as precipitates in liquid wastes; and 4) alteration products from degradation of wastes, waste containers, and repository infrastructure in the environment. Environmental, including biological, systems pose the special problem of physical and compositional heterogeneity coupled to typically dispersed and low concentrations of radionuclides. Questions needing answers that crosscut all areas are:

- How are radionuclides speciated over wide ranges of temperature and pressure?
- What is the supramolecular chemical behavior of radionuclides?
- How do the kinetics of reactions influence processes involving radionuclides?
- How can outstanding issues of sample size, radionuclide concentration, and reaction times be addressed experimentally and analytically?

Critical developments in synchrotron measurements are needed to answer these questions. New capabilities include detecting lower concentrations, probing smaller sample sizes independent of radioactivity level, measuring accurate reaction rates in real time, and engineering new forms of experimental and detector apparatus that have appropriate containment.

Detection of lower concentrations is essential for far-field characterization of radionuclide interactions with soils, sediments, water, and biota in the environment and for experimentation in separations technologies. Probing smaller samples via imaging, spectroscopy, and scattering is critical to the entire spectrum of processes — from development of waste glasses, to understanding oxidation state changes in solutions, to monitoring ion-exchange properties of separations materials, and to characterizing colloid and nanoparticle formation in the environment. Measuring reaction rates is of particular interest for monitoring oxidation state changes, characterizing ion-exchange on surfaces, forming and dissolving nanoparticles, and separating radionuclides using multiphase solutions.

Below are three representative sets of examples that focus on specific unresolved science issues that motivate upgrades in the APS facilities.

Speciation of Radionuclides in Solutions

There is a great lack of information on the association and molecular structure of trace and minor solutes in both traditional solvents such as water and the myriad of organic solvents including alkanes, ethers, and alcohols and in non-traditional liquids composed of molten salts, molten metals, and room-temperature ionic liquids. If we had knowledge of how these solutes are incorporated into this range of solutions, we would be able to better understand how to model the potential fate and transport of radionuclides in the environment, how to separate radionuclides from spent nuclear fuel, and better design functional high-performance materials for use in advanced nuclear energy systems and waste containment.

Because the partitioning of radionuclides and metals is driven by small energy differences and consequently can occur rapidly and at sharp boundaries, direct synchrotron-based structural and chemical probes are necessary to monitor when, where, and how the partitioning occurs in systems that span a broad temperature range from ambient to 800°C and pressure range from ambient to 100 GPa. X-ray absorption spectroscopy (XAS) has been used for a number of years to selectively probe actinide and fission product speciation in solutions at submillimolar concentrations, providing information on oxidation state and coordination environments [1]. Recently, high-energy X-ray scattering (>60 keV) has been used to provide more precise information on coordination environments in amorphous systems, including solutions [2]. These experiments require concentrations in the 10s of millimolal or higher, depending on the experiment. High-energy scattering experiments are providing new information that directly couples thermodynamic stabilities and free energies of solution complexes with their structures.

Radionuclide Separations

The second set of questions addresses the physics and chemistry of radionuclide separations. For example, the forces that favor and restrict potentially deleterious formation of a third phase in multiphase liquid-liquid extraction — a key process in the nuclear fuel cycle — are only just now coming into focus. Although there are clear correlations between temperature and solute concentration in the formation of a third phase, a significant gap in knowledge remains about the transition from a biphasic to a triphasic state. What is not yet clear is the effect of an externally-applied, hydrostatic pressure on this phenomenon. It would be desirable to observe if there is a "pressure-switch" to third-phase formation by conducting X-ray spectroscopy and scattering experiments on a biphasic system under modest applied pressures, for example from 1 GPa to 100 GPa. If so, this may provide a new entry into understanding an otherwise harmful phenomenon and create prospects for new approaches to separations science and technology.

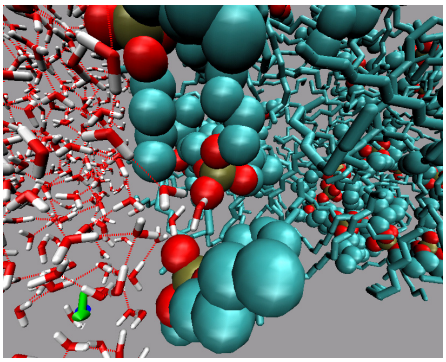
A second example in radionuclide separations has to do with understanding the basic physics of multiphase solutions now studied using centrifugal contactors, mixer-settlers, or pulse columns. The observed fluid dynamics in centrifugal contactors is a function of design, materials, physicochemical properties, and operating conditions. Much recent modeling has focused on the

nature of the interfaces formed between the aqueous and organic phases within the contactor because transfer of radionuclide species across the interface is central to the observed chemical behavior. Changes to the interfacial area are also critical to the separation and recovery of the products after reaction. Many factors play a role in the interfacial regime including bubble formation, size, and distribution; the rotor speed and geometry; the arrangement of vanes within the mixing zone; and the design of the exit ports within the rotor. Therefore, it would be highly advantageous to be able to image these fluid interactions *in situ* and link them to the design and operation of the machinery.

The third example is focused on the structural basis for the selectivity of ion exchange materials used to separate actinides from fission products. These materials change their structures when they adsorb a specific actinide or fission product ion and a fundamental understanding of this process requires knowing the effects of framework charge, the role of hydration/solvation on both the ion exchanger and sorbate ion, and the kinetics of the exchange reaction. Structural and chemical probes currently applied to non-radioactive materials at synchrotrons are appropriate tools for investigating these questions.

MOLECULAR-SCALE INTERACTIONS AT LIQUID-LIQUID INTERFACES

Progress and innovation in separation science and technology, in general, and advancements in actinide-lanthanide solvent extraction processes, in particular, can be driven by synchrotron radiation research in ways that have not yet been realized. The myriad fluid systems at the back end of the nuclear fuel cycle, where interactions between actinides, extractants, and solvents occur across a range of dimensionalities — from the molecular scale at the liquid-liquid interface (illustrated below) through to the supramolecular scale with the formation of reverse micelles and microemulsion — are wide open for exploration by X-ray scattering, spectroscopy, fluorescence, and imaging techniques. With regard to the latter, we envision a direct *in situ* entry to observe the distribution of actinide species as well as their fluid structures and dynamics in bulk media employed in centrifugal contractors. The APS would be ideal in this regard with the availability of hard X-rays corresponding the to K-edge energies of the 5f elements (109-129 keV for Th-Cm), for which the X-ray fluorescence yields are essentially unified and also of high energies (> 90 keV), providing penetration through process equipment.



Molecular dynamics simulation of the extractant tri-*n*-butyl phosphate at the water-*n*-dodecane interface. Water molecules (left) are colored red (O) and white (H). *N*-dodecane molecules (right) are illustrated by teal-colored linear chains and TBP molecules are shown with space-filling van Der Waals spheres. (K.E. Wardle, unpublished)

Radionuclides in the Environment

The third set of issues is exemplified by observations over the past decade showing that radionuclides can be distributed rapidly and broadly as colloids in environmental systems. Generation of colloids containing radionuclides has resulted from past waste disposal practices, underground nuclear tests, and mining/milling activities, and could result from failed repository storage containers in the future. At the Nevada Test Site [3] and Rocky Flats [4] the identification of colloidal forms of Pu in ground and surface waters, including Pu-oxides, other

metal oxides on which Pu adsorbs, and Pu complexed to dissolved natural organic matter has demonstrated a need for fundamental investigations of the chemical and physical behavior of these small particles.

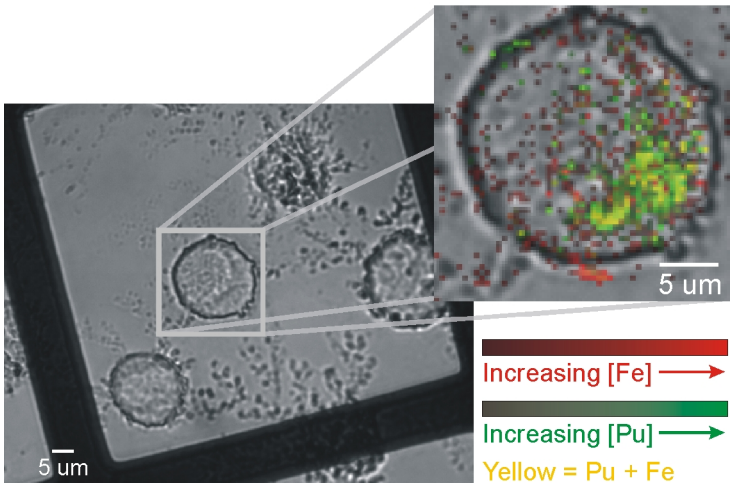
In these cases colloid formation accelerated Pu transport in ground and surface waters but in other cases, such as when high-level waste tanks leaked into the ground at the Hanford Site, formation of silicate colloids containing ^{137}Cs, ^{90}Sr, or U [5] appears to have immobilized or at least retarded transport. Colloids, which consist of nanoparticles and aggregates of nanoparticles or single particles approaching 1 micron in size, form relatively quickly in these environmental systems. They may be single phases, but more often are heterogeneous mixtures of phases; therefore, their identity, properties, and reactivity need to be characterized by *in situ* experimentation at the beamline.

The association of radionuclides with mineral surfaces in soils and sediments through adsorption of solutes or precipitation of surface species is critical information for modeling the future disposition of radionuclides in ground water. Some of this information is now obtained from synchrotron experiments and analyses using spectroscopy and scattering approaches.

The key issue is that the concentrations of targeted elements often have to be much higher than occur in the actual environment and this could lead to misunderstanding of the true controls on radionuclide immobilization mechanisms. The seminal work of Duff et al. [6] in which Pu was characterized as occurring as Pu(V) and associated with Mn-oxide and certain clay minerals in Yucca Mountain volcanic tuffs, was accomplished with synchrotron spectroscopic techniques but on samples that were doped with relatively high concentrations of Pu.

We need to understand the distribution of actinides and daughter products at lower concentrations and/or when localized in nanosized areas in heterogeneous environmental materials, ideally by sampling and analyzing contaminated materials directly using improved microprobe and nanoprobe (spectroscopy and diffraction) capabilities. These capabilities also would lead to increased understanding of the composition and distribution of radionuclides in tank sludges, still a significant problem at many DOE sites, as well as of the location of radionuclides in and mechanisms of transport through cellular tissues.

SUB-MICRON SCALE IMAGING OF Pu DISTRIBUTION IN MAMMALIAN CELL



Hard X-rays make sub-micron scale imaging of actinide-containing materials by X-ray fluorescence microscopy possible even for encapsulated samples. The optical micrograph above shows rat adrenal gland cells that were exposed to plutonium, deposited on electron microscopy grids, and encapsulated. Using on APS beamline 2-ID-D individual cells were mapped by rastering 18.1 keV X-rays focused in a 0.2 μm × 0.5 μm spot. This excites the L_3 edge of plutonium as well as the K or L edges of all the lighter elements, making it possible to simultaneously quantify elements from phosphorous (Z = 15) through plutonium (Z = 94). False color representations of the iron (red) and plutonium (green) distributions within a cell (above, right) demonstrate that iron and plutonium co-localize in the cytoplasm of these cells. Creation of this 26 μm × 26 μm elemental map with 0.5 μm resolution required 3 hours. (M.P. Jensen, unpublished)

APS Advances Necessary to Meeting Science Needs in Solution/Amorphous Studies

Current capabilities and planned upgrades to these capabilities at the APS are anticipated to be, for the most part, sufficient to meet the stated scientific challenges and opportunities. However, two enhancements are necessary to address the specific science questions related to advanced nuclear energy and it is considered that these improvements would apply to the full range of APS users. These are 1) increased stability of microbeams in current microprobes, and 2) development of the capability to access concentrations below 1 ppm for spectroscopy.

Additionally, new types of experimental and detecting equipment must be developed to make rapid progress in the fundamental science behind advanced nuclear energy systems. It is essential to construct 1) a modular experimental suite or set of modules for *in situ* studies, operando monitoring of reactions in real time, and sample manipulation; and 2) modular detectors for high-activity, irradiated, or activated samples. Such modular equipment will require a high level of design and engineering to achieve the science goals along with attention to proper containment and safety infrastructure.

Modular experimental suites would allow the necessary containment and encapsulation for a desired sample or type of experiment, providing the user with the ability to accomplish the required measurement as well as manipulate the sample with respect to temperature, pressure,

and chemical environment. Existing designs have already been demonstrated at the APS and copied at other synchrotron facilities; such as the *in situ* spectroelectrochemical cell for *in situ* measurement of solution electrochemical experiments involving actinide samples [7]. More advanced modules for imaging, surface studies, *in situ* hydrothermal and solvothermal investigations, and experiments requiring flowing liquids and reactive atmospheres are required.

Having modular equipment for experimentation will provide many advantages. The equipment will:

- Enable a wider community of users from the fields of chemistry, environmental science, materials science, biological science, and physics;
- Ensure the safest operation by providing input, experience, and lessons learned from a broad user group;
- Provide the capability of examining one experiment at multiple beamlines without dismantling an apparatus;
- Maintain a smaller nuclear footprint in the APS facility; and
- Provide the ability to exploit the best techniques at the most appropriate beamlines.

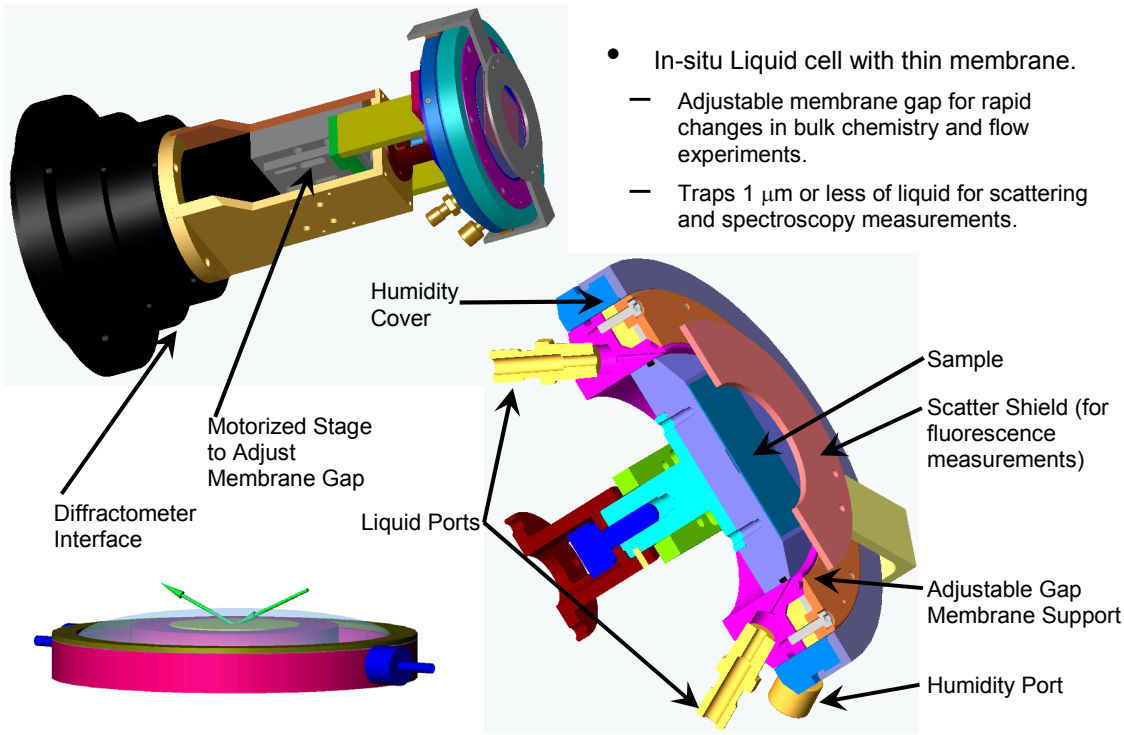
An example is the planned upgrade to X-ray Raman spectroscopy, which is a promising probe of low-energy edges (actinide O, oxygen K etc.) for high-activity samples.

Advantages of modular detectors are

- Improved performance under high-field radiation conditions, and
- Minimization or complete avoidance of induced radiation damage.

With modular equipment and detectors, a broad set of investigations under a diverse set of *in situ* conditions would be possible for the first time. These include monitoring reactions in supercritical solutions and during separations, formation, and dissolution of glasses at high temperatures; formation and dissolution of colloids at low temperatures; and electrochemical stimulation of oxidation state changes. Additionally, high-performance detectors and crystal analyzer systems would allow acquisition of data from high-activity samples where signal and detector performance are paramount due to the intrinsic sample properties.

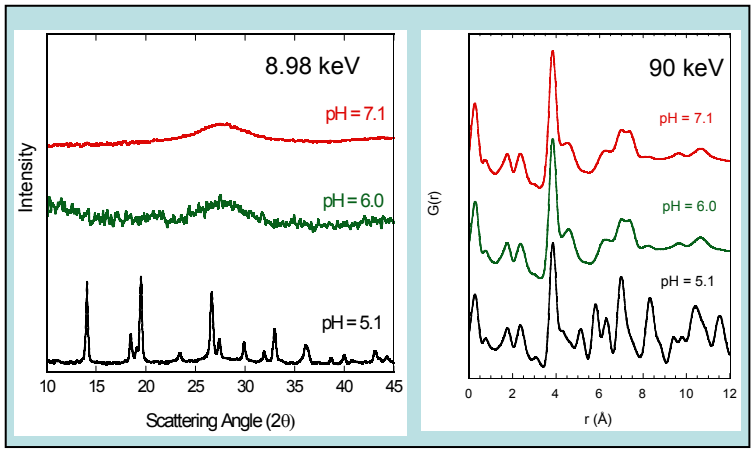
DEVELOPING A SURFACE AND INTERFACE SCATTERING SAMPLE CELL



- In-situ Liquid cell with thin membrane.
  - Adjustable membrane gap for rapid changes in bulk chemistry and flow experiments.
  - Traps 1 μm or less of liquid for scattering and spectroscopy measurements.

In situ surface scattering from solid-liquid interfaces involving radioactive samples requires engineered containment that benefits from both beamline staff and experimenters. (Peter Eng, unpublished)

CORRELATIONS IN AMORPHOUS AND SOLUTION SAMPLES



A comparison of X-ray data obtained with an in-house diffractometer using a Cu tube (left) with the Fourier transform of scattering data using 90 keV X-rays (right) from the APS Sector 11B. The data were obtained from the same series of uranyl silicate solutions and highlight the information available on atomic correlations in amorphous systems. Soderholm, et al., *Geochim. Cosmochim. Acta* **72** (2008) 140-150.

2. CORROSION AND INTERFACES

Interfaces play a crucial role in nuclear energy systems, ranging from corrosion of fuel cladding and structural materials and the stability of fuel cladding during and after operation, to separations of actinides species during fuel cycles, to the stability of waste forms in long term geological repositories, and to the environmental transport of actinides and fission products. These interfaces encompass solid-solid, solid-liquid, and liquid-liquid interfaces and, in many cases, their characteristics drive changes that propagate into the bulk, determining its final form. However, the role of interfaces in controlling reactions, transport, and phase stability is poorly understood, particularly in extreme radiation, temperature, and corrosive environments.

The photon science community, including the broader interfacial science community at the APS, recognizes the general feature and importance of interfacial science. The interfacial science community needs for research on nuclear energy systems has been met so far (at least in terms of available X-ray techniques for non-irradiated materials). Various researchers have taken advantage of the existing and expanding interface science expertise at the APS. To extend such work to irradiated solids, the use of radioactive isotopes, and corrosion under irradiation (radiolysis) requires different capabilities. Some of these new capabilities (such as in the study of sorption of radioactive isotopes in soils) can be accommodated in existing beamlines with appropriate sample containment, although some may require more extensive facilities and experienced personnel.

Scientific Challenges and Opportunities

Interfaces in materials play a crucial role in the stability and performance of almost every aspect of advanced nuclear energy systems, from fuel claddings and pressure vessels in reactors, to fuel reprocessing and separations, and ultimately to long-term waste storage. Next-generation advanced reactors may operate at considerably higher temperatures and pressures and their components may be exposed to higher burn-up than the current light water reactors.

Therefore, significant challenges associated with the degradation of the structural and fuel cladding alloy materials in the reactor core are expected because of increased radiation damage, corrosion, pellet-clad mechanical interaction, gas release and swelling, and hydriding.

Major contributors to the corrosion/hydriding are the higher expected temperatures and the long exposures in those reactors. In fact, the leading advanced reactor concepts do not presently have materials that can withstand their operation conditions. Revolutionary advances in structural and cladding materials resistant to the corrosive and irradiative environment are needed to significantly improve the lifespan of fuel and structural materials and reduce the operating cost.

One crucial common characteristic of the microstructure of nuclear materials when exposed to irradiation is their *heterogeneity*. Because of irradiation-induced microchemical evolution, radiation damage, and temperature and flux gradients, the microstructure of nuclear materials is far more heterogeneous than that of non-nuclear materials. For example, uranium dioxide fuel pellets undergoing light water reactor (LWR) irradiations contain many different regions with varying concentrations of fission gases, different shaped grains, varying porosity, and metallic

precipitates. This microstructure also evolves such that towards the end of the irradiation period a rim structure is formed at the edge of the pellet. The heterogeneity of such structures is responsible for many of its properties (for example, advanced alloys have dispersion strengthening oxide nanoparticles that provide high temperature strength and help anneal radiation damage). Heterogeneity implies the presence of interfaces and thus, investigating interfaces is a crucial aspect of investigating the behavior of materials under neutron irradiation in a corroding environment.

Such interfaces can be external (such as the metal-water interface during corrosion) or internal (such as the edge of the rim structure described above). Since chemical degradation of materials is initiated at their external surfaces or interfaces, it is imperative to hinder or control the degradation at the incipient stage on the surfaces or interfaces.

Therefore, it is important to understand fundamentally the oxidation and corrosion mechanisms and the microchemical and structural properties of the surfaces and interfaces. The fundamental understanding of the alteration mechanisms and product properties must be systematically extended from the un-irradiated surfaces and interfaces to severely irradiated surfaces and interfaces so that the behaviors of the surface and interfaces under relevant environments can be predicted, for example, in multiscale computer simulation using the experimental parameters as input values. The systematic fundamental understanding will also enable us to *engineer* materials suitable for the expected performance of the material-environment interface, instead of costly trial-and-error style developments. Moreover, it can eventually allow us to contemplate "self-healing" adaptable material interfaces capable of providing dynamic, universal stability over a wide range of anticipated (and unexpected) conditions of advanced nuclear energy systems [8].

Interfaces are critical components in design of radiation-resistant materials by nanophase self organization. Theory, modeling, and experiments have demonstrated that alloys can undergo self organization on a mesoscopic length scale when subjected to irradiation [9] and the resulting microstructure is in a steady state such that radiation resistance can be maintained indefinitely. A phase diagram of these irradiation stabilized steady states provides a similar utility as equilibrium phase diagrams to materials scientists and engineers. Experimental study of such steady-state phase diagrams requires prohibitively long and costly experiments for each alloy composition considered. However, the study can be significantly simplified and reliable if it can be performed *in situ.*

The importance of synchrotron radiation is recognized within the nascent community interested in developing a molecular-level understanding of interfacial processes and their role in nuclear separations processes. Whether for developing scenarios for reprocessing nuclear waste or modeling contaminant transport in the environment, molecular interactions at solid-liquid and liquid-liquid interfaces govern the observed thermodynamics and kinetics in ways that must be understood before significant progress can be made. X-ray surface scattering and reflectivity studies have been done that demonstrate the potential for elucidating mechanisms of adsorption and phase transfer [10, 11], but much more work is required.

Interfaces are also critical components in studying the disposal of radioactive wastes. Processes such as canister corrosion, radionuclide sorption, leaching and redox reactions of waste forms, and radionuclide migration are all interfacial processes that need to be understood for assessing

the performance of waste repositories. Interaction of actinides and fission products with mineral surfaces occurs at the solution-solid interfaces.

The fundamental chemical interactions concerning the low-temperature dissolution of waste forms, as well as the interaction of dissolved species with solid surfaces, are central to developing a firm scientific foundation for mitigating the possible environmental impacts of nuclear energy systems. Studies of waste form interactions with aqueous solutions (e.g. simulated ground water) can provide information concerning the processes and associated kinetics that control the long-term release of radionuclides. This would include leaching of particular waste form components, the evolution of the interfacial structure, and the formation of secondary phases, all of which could be studied in real time and as a function of solution conditions.

Direct observations of the interaction of dissolved species with mineral surfaces (with molecular-scale resolution and elemental specificity) will provide fundamental understanding of the processes that control the environmental transport through measurements of phases of adsorbed species (i.e., inner- vs. outer-sphere adsorption), adsorption thermodynamics, kinetics of adsorption/desorption reactions, and precipitation reactions. In particular, it will be necessary to understand the relationship between the chemical species of interest (oxidation state, and single ion vs. solution oligomers).

APS Advances Necessary to Meeting Science Needs in Surface/Interface Characterization

The main advantage of the APS X-ray techniques stems from the unique characteristics of brilliant synchrotron X-rays that can deeply penetrate most materials and environments without suffering from severe attenuation and yet provide sufficient sensitivity to the atomic structures and chemical states of the surface and interfaces. Although some surface-sensitive X-ray techniques were developed at second-generation synchrotron sources, advanced surface/interface X-ray techniques (e.g., for buried interfaces) have been fully developed at APS and other third-generation sources. Glancing-angle incidence techniques are extremely useful in depth-sensitive study of surfaces and buried interfaces (see Figure 1).

The APS X-ray beam is also significantly more coherent than the beams in previous synchrotron sources, which enables us to study dynamics of surface and interfaces instead of the traditional time-averaged states. Also, the tunablility over a wide range of X-ray energy enables us to study the chemical states of most atomic elements on the surface and buried interfaces. In addition, the high brilliance enables extremely tight focusing of X-ray beams from micron to nanometer scale. The micro- and nanofocused X-ray beams can be used for nondestructive microscopic imaging of the structural and chemical states of the surfaces, as has been done for non-irradiated materials [12]. It could be of great interest to perform similar work in irradiated materials to verify that the oxide growth mechanisms observed in the reactor irradiation are similar to those observed in autoclave tests. This can likely be performed in the context of the existing triple containment sample system.

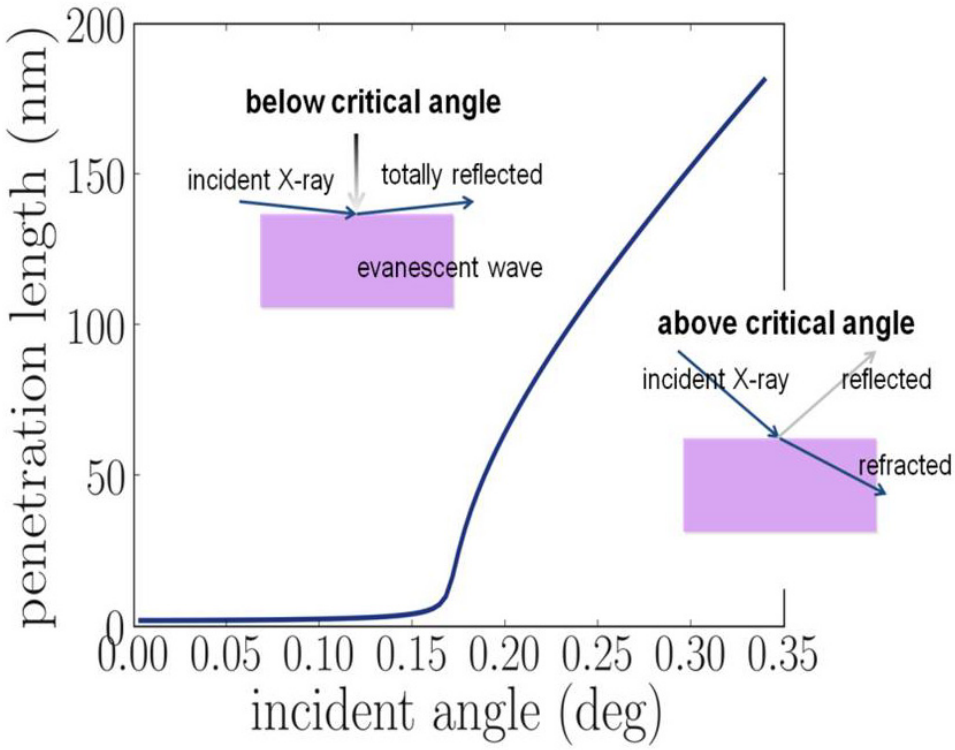


Figure 1. Depth vs. angle of incidence. The depth can be finely tuned in nanometer scale below critical angle and coarsely tuned above the critical angle to micron scale.

In situ **Surface Scattering**

The most powerful aspect of the APS X-ray techniques is certainly the *in situ* applicability in examination of the interfaces often buried under liquid, solid, or under extreme conditions of temperature, pressure, chemical, and irradiative conditions directly relevant to the advanced nuclear energy system. For example, we can examine corrosion reactions *as they occur*, including the influence of radiolysis to determine oxidation states, elemental distribution, secondary phases formed, etc. The X-ray techniques that can be used for *in situ* studies include surface scattering and diffraction, depth-sensitive fluorescence spectroscopy, and microscopy. The X-ray techniques suitable for the buried interfaces include glancing-incident X-ray diffraction, glancing-incident X-ray fluorescence absorption spectroscopy, and surface-enhanced resonance scattering. Although some intermediate resolution microscopy using a microfocused beam can be performed *in situ*, in general techniques based on micro- and nanofocused X-ray beams have limited *in situ* applicability due to the correspondingly short distance between the focal point and the sample.

In situ synchrotron studies focused on chemical speciation at solid-liquid and liquid-liquid interfaces are providing critical information to separations and environmental processes, but the extension of these studies samples involving radionuclides is significantly hampered by sample-containment issues. The APS state-of-the-art beamlines provide the photons and geometry necessary to carry out the experiments of interest. Unfortunately, strict experimental

requirements regarding surface positioning relative to both the incident beam and detector placement significantly impact the options for sample containment — a critical component of running experiments with radioactive samples on a general surface-scattering beamline.

A similar dilemma exists for surface studies involving corrosion and irradiated materials. It has already been shown that *in situ* monitoring of the important phenomena, such as interfacial uniform corrosion, can yield insights unobtainable in *ex situ* measurements (Figure 2) [13, 14]. *In situ* measurements often eliminate the guesswork associated with temperature and environmental cycles that, if performed *ex situ* can complicate the interpretation of the results.

To make the *in situ* study more realistic, we can simulate the chemical conditions of cooling fluids by intentionally adding radicals and chemically active elements known to exist in actual reactor operating conditions. Pristine samples to samples with varying degrees of radiation damage can also be examined for the corrosion process. The comparison of corrosion behavior between these samples should delineate the effect of fluid corrosiveness from the irradiation-induced corrosion.

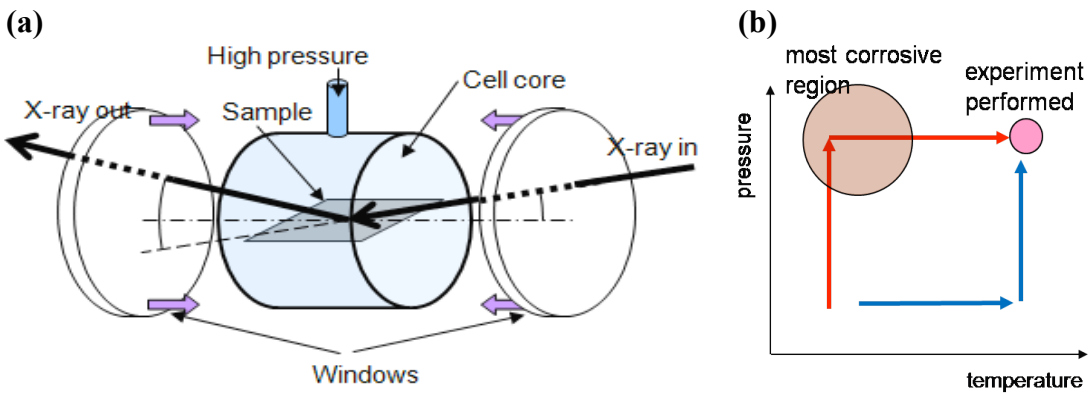
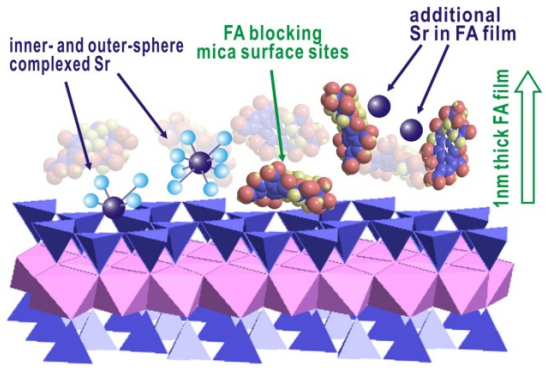


Figure 2. (a) Schematics of the high temperature/pressure cell for *in situ* interface X-ray scattering/spectroscopy studies. This cell was used to study surface corrosion of stainless steel in water at high pressure and temperature. (b) The cell was used to show that the corrosion can depend on the paths in P-T diagram and to identify where the corrosion takes place.

This area of research can take advantage of available surface-scattering beamlines but will require significant advancements in sample containment and the development of modular experimental suites designed with the geometrical considerations in mind.

QUANTIFYING INTERACTIONS AT MINERAL-WATER INTERFACES



Recent advances in interfacial X-ray characterization enable direct molecular-scale observations of how actinides and fission products interact with mineral surfaces in complex natural environments. Here, the competitive interactions between strontium (Sr^{2+}) and fulvic acids (FA, a form of natural organic matter) at charged muscovite-solution interface are revealed with resonant anomalous X-ray reflectivity. This work demonstrates a complex multisite adsorbed cation distribution and a nontrivial interplay between adsorption thermodynamics and kinetics. These factors ultimately will exert control over contaminant transport in the near-surface environment. The application of this approach to study actinide interactions with mineral surfaces is underway. (Lee et al., *Geochim. Cosmochim. Acta* (2010) **74,** 1762-1776)

Stress Corrosion Cracking (SCC)

In situ X-ray measurements may not be directly applicable for all corrosion problems. Many aspects of the SCC could be effectively studied *ex situ*. However, ex-core structural materials such as nickel-base alloys have suffered from SCC and finding structural materials resistant to SCC is a key to the materials improvements. For example, although a new material, alloy 690 (Ni-30Cr-9Fe), has shown excellent resistance to SCC it was found to be more susceptible to SCC in alkaline solution, particularly in the presence of Pb. *Ex situ* analyses (TEM, XPS, etc.) suggest that Pb dramatically alters the characteristics of the oxide layer formed on the alloy surface. The double oxide layer becomes single oxide layer so that $Ni(OH)_2$ and $Cr(OH)_3$ rather than NiO and Cr_2O_3 are formed at the surface. The hydroxides have less mechanical strength and therefore are less resistant to stress cracking. This is a pure interface phenomena which can be readily studied by the depth-sensitive *in-situ* X-ray fluorescence technique such as element-specific information at oxide/liquid interface and migration of Pb inside oxide layer.

Such measurements would require higher-energy X-rays, both for penetration into the bulk and for accessing the absorption edges of heavier elements including Pb. *In situ* measurements with the sample under load and at higher temperatures would be much more technically challenging and would require longer times in the beam. SCC experiments would require extensive sample-confinement engineering, regardless of where they would be undertaken. This is a clear example of the need for personnel experienced in SCC and with expertise in synchrotron experiments and radiological protection to be available to the user community.

Effect of Irradiation on Corrosion

The effect of irradiation on corrosion can be partly addressed by examining oxide layers formed under reactor conditions (and thus, under the influence of an irradiation flux). The oxide phases, microstructure, and phase separation can be compared to the out-of-pile condition to verify the specific influence of the irradiation environment. *In situ* studies on irradiation's influence of corrosion likely would only be possible using ion irradiation to simulate radiolysis or possibly using a radioactive sample to produce radicals in the water.

The effect of radiation damage on corrosion is another important area of research. Again, we can design experiments to simulate irradiation using ion sources, much like a recent investigation employing light reflectivity using a laser in a vacuum chamber with ion source [15]. We can examine the surface and subsurface *in situ* during irradiation using depth-sensitive off-specular diffuse scattering, known as Huang scattering [16], as the surface and subsurface evolve from the pristine, to irradiated, and finally to a steady-state irradiated condition. Off-specular diffuse scattering is particularly sensitive to second-phase formation in alloys, which is known to strongly influence corrosion behavior. Once set up, this type of *in situ* measurements can yield a steady-state phase diagram as a function of sample temperature (or average size of the mesoscale pattern formation) vs. irradiation amount, which is generally difficult to obtain by any other means.

Surface-enhanced X-ray fluorescence technique can be used to examine the elemental and chemical composition of the interface as a function of depth, ranging from nanometer to micron, in addition to the structure information. These measurements can be combined with other characterization such as transport measurements during irradiation [17, 18]. The samples irradiated and characterized can now be placed back into the fluid cell discussed above and examined for irradiation-induced corrosion.

Micro- or nanofocusing of X-ray beams became possible at APS and other third-generation sources due to new X-ray lenses, such as zone plates, refraction lenses, and high-quality mirrors. The focused beam is powerful for spatially resolved nondestructive imaging of the heterogeneity of surfaces and interfaces with respect to elemental composition, structural composition, and oxidation state. It can also be used to image the second-phase formation and follow its evolution during irradiation.

Examination of Fuel

The focused beam can have many other applications. Cross sections of fuel pellets can be examined for the elemental, oxidation-state, and structural distributions using a microfocused X-ray beam and can correlate such distribution to fission gas release and fuel performance at high burn-up. Previous work with synchrotron X-ray microanalysis of corrosion in commercial spent nuclear fuel has concentrated on questions important to licensing the Yucca Mountain repository — primarily the fates of Np and Tc in the case of water infiltration resulting from a breached storage container.

However, in this study the original locations of the particles in the fuel pellet were unknown. Given the very different microstructure and composition as a function of radial distance from the center of a fuel pellet, the study of fuel-pellet cross sections using X-ray microscopy, X-ray microdiffraction, and X-ray microspectroscopy would yield a wealth of information that would help improve the fabrication and performance of nuclear fuels. The availability of high-energy X-rays would significantly broaden this opportunity. In addition, with appropriate sample encapsulation and handling facilities at the APS, the fuel pellets could be studied from fabrication though high burn-up and beyond to accelerated corrosion.

A broad range of length scales are important to the fuel structure evolution; in order to study the interface between the fuel and the cladding or the corrosion at the cladding-coolant interface, focused-beam techniques are necessary. Grain size and orientation can be studied with X-ray microdiffraction, while very sensitive elemental distribution maps can be generated with X-ray fluorescence and chemical species and oxidations state are measurable from X-ray spectroscopy — all as a function of position. The examination of fuel could strain the APS capabilities in terms of radiological limits, and ability to contain and handle radioactive samples.

The combination of sample transportation requirements and the scheduling of synchrotron beamtime call for a dedicated facility to enable and enhance our abilities to study these hazardous samples. In many of the interface studies mentioned above, improved radiological sample handling is required. Many, but not all of the experiments discussed above can be done on existing beamlines.

However, enhanced sample handling capabilities are required to perform experiments on samples containing radionuclides under extreme conditions including, as examples, elevated temperatures, *in situ* irradiation, and harsh chemical conditions. *In situ* mechanical testing, such as tensile strength and bending of irradiated samples on radioactive specimens, would add enormously to the development of new models to simulate materials behaviors in advanced nuclear energy systems. A purpose-built facility and personnel familiar with the experimental problems associated with handling and encapsulation of radioactive materials would open up a new user community eager to take advantage of synchrotron radiation and the information it brings to their problems.

3. RADIATION DAMAGE AND EFFECTS

Adequate response to the significant demands on fuel and structural materials is critical to the performance of any nuclear system, either fission- or fusion-based. Improvements in materials science and technology will support the continued safe performance and extended lifetimes of existing light water reactors (LWR)s; enable the construction of new extended-life LWRs, advanced fission concepts, or fusion-based systems; and support a sustainable closed fuel cycle [8, 19, 20]. Operations to extend reactor lifetimes present challenges to materials which must withstand many degradation mechanisms including irradiation creep, low-temperature embrittlement, void swelling, irradiation-assisted stress corrosion cracking, and fuel clad chemical interaction to doses up to 400 displacements per atom (dpa).

Scientific Challenges and Opportunities

The Challenge of Predicting Material Performance in Nuclear Systems

The combined irradiation field and high temperatures under which nuclear systems operate can lead to changes in toughness, strength, ductility, fatigue, and creep resistance that shrinks the range of allowable temperature and stress in which materials performance is adequate (See Figure 3). Additionally, materials can be attacked, leading to cracking and mass loss in a corrosive environment. The specific effects depend on the exact combination of temperature, stress, irradiation flux, and the environment (corroding media) (See Figure 4).

For fuel, structural materials, and waste forms, the key microstructural feature that leads to degradation is a system of nanometer-sized features *distributed* within micron-dimension grains. These features initiate with nm-sized displacement cascades leading to enhanced diffusion and a non-equilibrium microstructure.

The degradation mechanisms that occur under radiation are a function of temperature (relative to the melting temperature Tm), damage rate, and total dose, and fall into five categories as described by Zinkle [21]:

- Radiation hardening and embrittlement ($<0.4\ T_M$, >0.1 dpa)
- Phase instabilities from radiation-induced precipitation ($0.3\text{-}0.6\ T_M$, >10 dpa)
- Irradiation creep ($<0.45\ T_M$, >10 dpa)
- Volumetric swelling from void formation ($0.3\text{-}0.6\ T_M$, >10 dpa)
- High-temperature He embrittlement ($>0.5\ T_M$, >10 dpa)

It is not possible to do experiments that simulate 80-years of irradiation damage and corrosion in a practical time frame. Approaches that accelerate the transition from observation and validation of performance to the prediction and control of functionality through materials discovery and science-based certification remain grand challenges. A particular challenge is the ability to make (and model/interpret) measurements of relevant phenomena in irradiation environments of interest, thereby advancing fundamental understanding and predictability beyond traditional approaches.

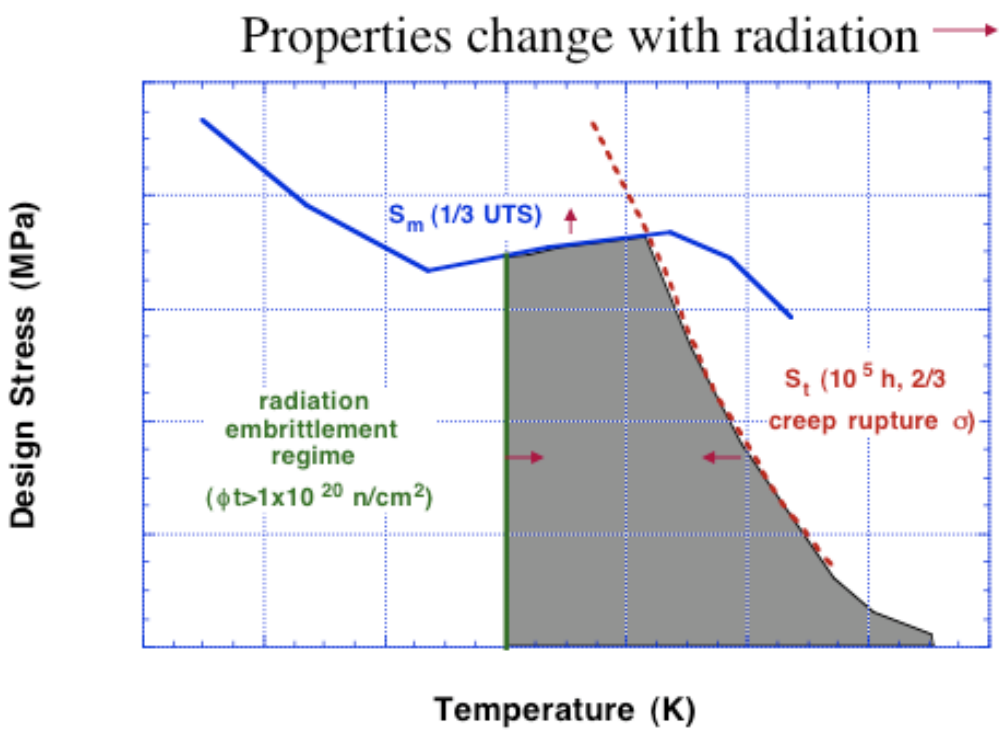


Figure 3. Each material has properties that allow it to be used in certain temperature and stress ranges and these are changed by irradiation. (Adapted from *S.J. Zinkle et al., STAIF 2002, Ed. M.S. El-Genk, AIP Conf. Proc. 608 (Am. Inst. Phys. 2002) p. 1063*)

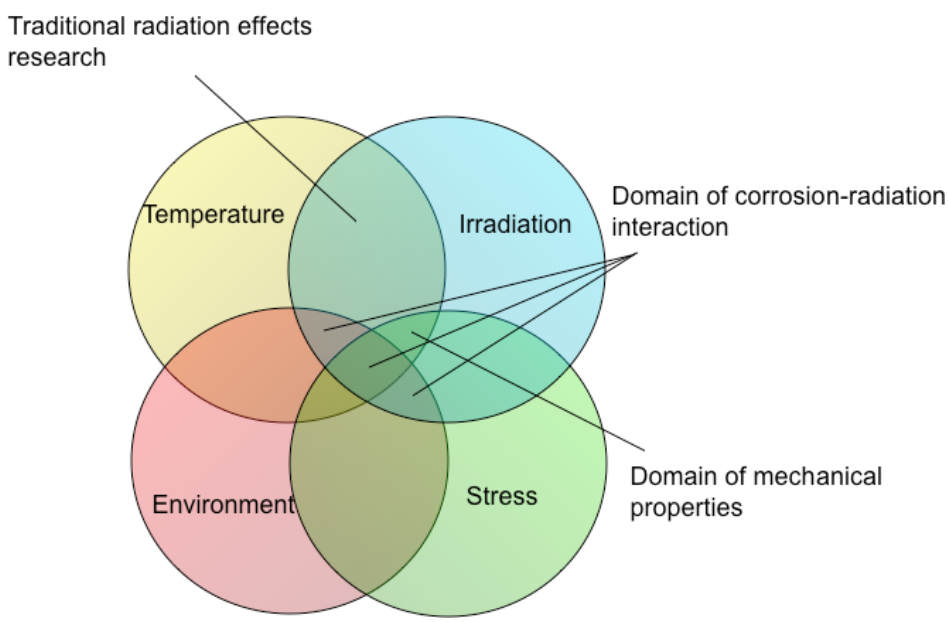


Figure 4. Radiation effects are a unique combination of temperature, stress, irradiation, and environment.

Therefore, materials scientists will need to rely on theory, simulation, and modeling, validated by well-posed experiments to predict material behavior. As presented in Figure 5, this theory, simulation, and modeling must be developed on three general scales: 1) bulk material properties (strength, corrosion resistance, thermal conductivity, dimensional stability); 2) microstructure; and 3) atomic-scale unit processes. Information from the atomic-scale processes is required to understand the development of microstructures and the specifics of the microstructures determine the bulk properties. Developing the ability to predict and control the material response at each level would allow for the extrapolation of material properties beyond the existing database and control of materials properties through informed design.

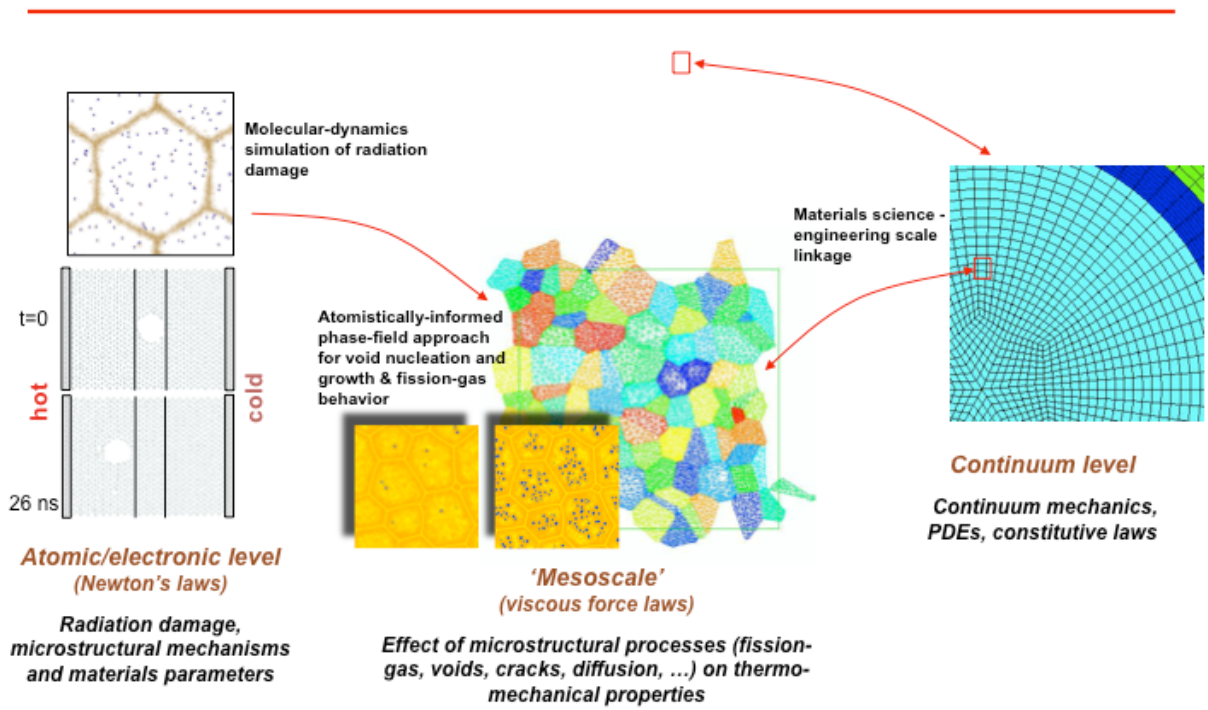


Figure 5. Material properties (continuum level) are determined by microstructural features (mesoscale) which form due to interactions at the atomic and electronic level.

Examples of Degradation Due to Radiation

Void Swelling

Void swelling is a dimensional change that occurs under radiation at intermediate temperatures (roughly 30-50% of the melting temperature of a material) (See Figure 6 left image) and must be managed either through material replacement or material improvements. The amount of swelling is controlled by microstructural developments; specifically the nucleation and growth of voids and dislocation loops at the mesoscale (See Figure 6, right image).

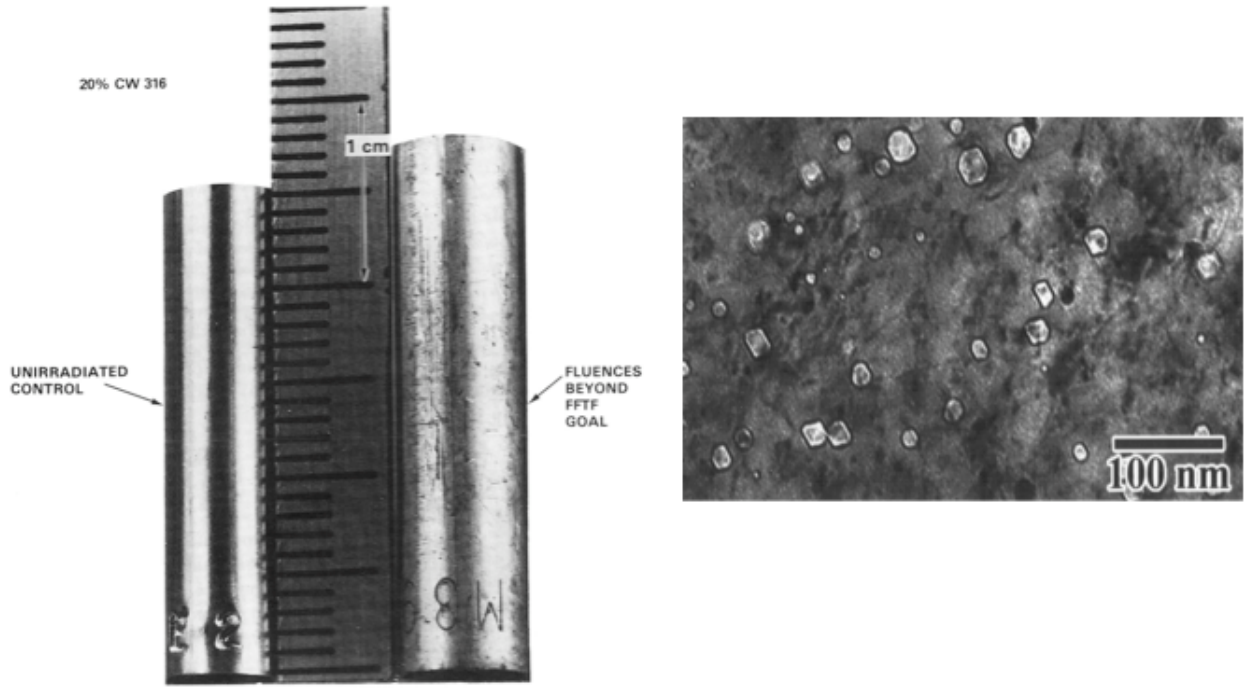


Figure 6. Swelling of an irradiated material (left) and the underlying microstructure. A collection of voids (right) that lead to the swelling [22].

Accurate prediction of the microstructural development requires a knowledge of many key processes that occur at the atomic level, including the creation of radiation-produced point defects; transport of defects and atoms through the material; nucleation and growth of mesoscale defects; and interactions with sinks, as shown schematically in Figure 7 for the phenomena of radiation embrittlement and stress corrosion cracking. Understanding many of the phenomena shown in Figure 7, such as point defect and atomic transport and radiation-induced segregation, is also critical in predicting other radiation-induced effects such as creep or void swelling.

For example, experiments and modeling have shown that void growth can be changed by radiation-induced segregation to the void surface and that radiation-induced segregation in austenitic iron-based materials is a strong function of bulk composition [23, 24]. Therefore, to properly model radiation effects requires an understanding of how structure, composition, and strain affect activation barriers in non-dilute, multi-component, and crystalline solids. This is necessary to determine the complex interplay between structure and composition. Critically, all these atomic-scale effects (unit processes) occur concurrently, so establishing methods for understanding the concurrent development of many unit processes is critical to understanding the microstructure and thus, the bulk properties. Material solutions need to be designed to control deleterious radiation response.

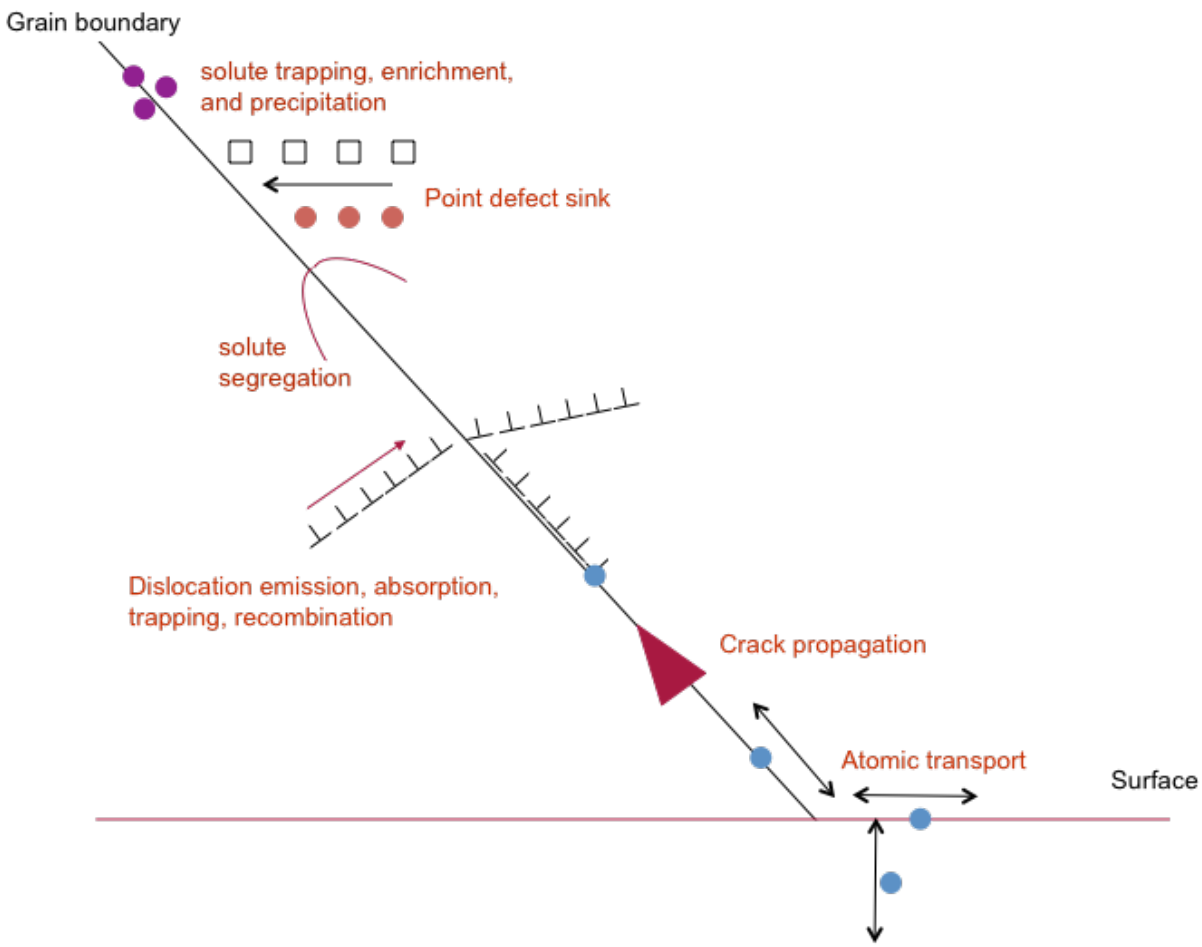


Figure 7. Many atomic-scale interactions occur that lead to changes in microstructure [8].

One possible design to control radiation response is the use of selected interfaces to mitigate radiation damage [25-28]. Interfaces with low formation energy of vacancies and interstitials, high excess atomic volume, and high density of misfit dislocation intersections are expected to be good sinks for radiation-induced defects. Thus, it should be possible to design materials with a high density of special "super-sink" interfaces in order to have a tailored response in extreme conditions of irradiation, stress, and temperature.

Precipitate Stability

Although many improvements were made to the design of austenitic steels in order to mitigate void swelling, the final engineering response was to switch to body-centered cubic (BCC) steels which inherently have a better response to swelling. Unfortunately, these BCC steels have lower high-temperature strength, thus bringing their own engineering limits. One method to increasing the strength of BCC steels is the addition of nanometer-sized Y-Ti-O particles (See Figure 8), which leads to the concept of oxide-dispersion-strengthened (ODS) steels. The stability of these nanoclusters under radiation is critical to the performance of the alloy.

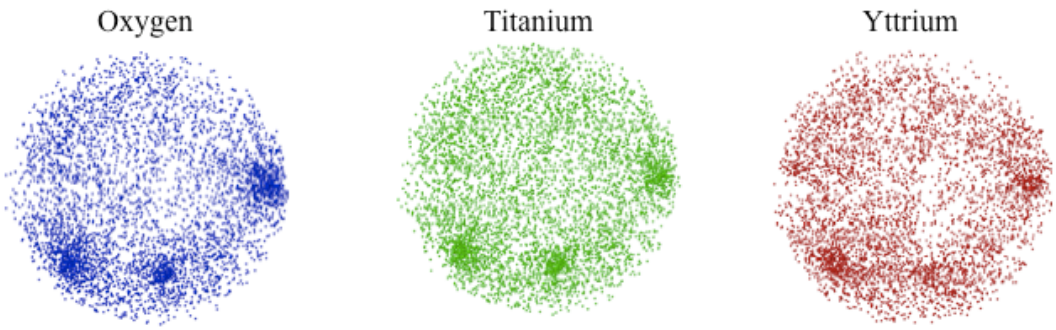


Figure 8. Atom maps from an oxide-dispersion-strengthened steel [29].

Understanding the performance of an ODS steel requires knowledge about size distributions as a function of radiation conditions (dose, dose rate, and temperature). The mechanisms of particle dissolution or stability depend on local (5-10 nm) concentration gradients at particle surface within micron-sized grains, the composition and structure of particles as a function of size, and the nature of the coherency of particles and matrix.

Irradiated Nuclear Fuels

Under reactor operating conditions, metallic-alloy fuels undergo significant chemical and material changes that are a major factor in limiting their operational lifetime. Material modifications include phase and elemental segregation, microscopic re-crystallization, stress, fission gas production, and bubble formation. Concentration variations, as shown in Figure 9, are of significant concern. Driven in part by the thermal gradients within the nuclear fuel, these variations can affect reactor performance and fuel burn-up levels. Similar gradients were observed in samples that were not irradiated but underwent thermal gradient treatments. [30, 31] From measurements such as these, kinetic parameters (such as effective inter-diffusion coefficients) were derived. The amount of such experimental data is very limited. Interaction of the fuel constituents with cladding and coolant are also important.

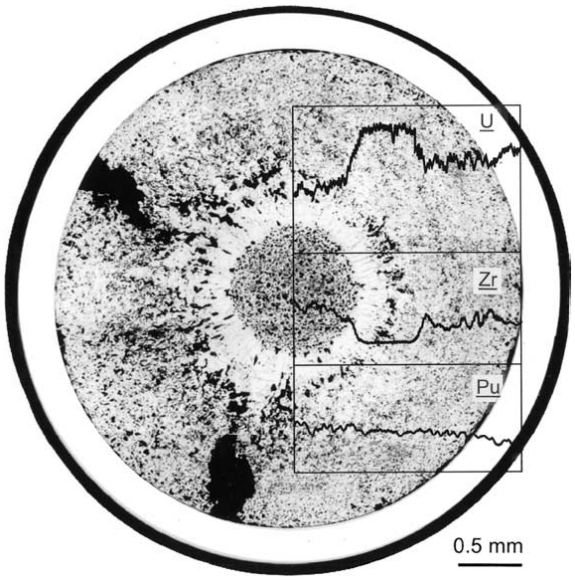


Figure 9. Past studies of the ternary nuclear fuel UPuZr have demonstrated constituent redistribution when irradiated or with thermal treatment. [30, 31] Post-irradiation optical metallography and measured constituent redistributions in T179 fuel at 1.9 at.% burn-up. Note the U depletion and Zr enrichment in the center zone and U enrichment and Zr depletion in the intermediate zone. Energy Dispersive Spectroscopy (EDS) within a scanning electron microscope was used to determine the concentration variations.

Tools for Studying Radiation Matter Interactions for Fission and Fusion

Researchers need the capability to expose materials to well-characterized and controlled extreme environments while simultaneously probing, measuring, and manipulating interfacial reaction kinetics, transformations, and mechanisms associated with their performance. These data and tools are essential to replacing empirical- and database-driven predictive models with knowledge-based models for design, prediction, and control of performance, and prognostic health monitoring of materials targeted for use in extreme environments.

Unit processes, such as defect-interface interactions, strongly influence the material properties and irradiation stability of materials. For example, interfaces with nanometer-scale spacing block slip and lead to unusually high strengths in materials. Interfaces also act as sinks for radiation-induced point defects and impurity atoms such as helium. The atomic structure of the interface is crucial in determining the number density of sites that are traps for defects and the formation energy of point defects at interfaces.

Understanding either a unit process or collective effect requires the proper tools to interrogate the proper length and time scale. Observation *in situ* could significantly improve the ability to observe effects like defect production, diffusion, and nucleation. Recent advances in capability are greatly improving the ability to provide quantitative information on smaller length and time scales [32].

While many radiation effects of concern are initiated by collision between lattice atoms and high-energy neutrons, a significant amount of radiation damage in solids occurs between charged particles. Atom-atom collisions in damage cascades or interactions with fission products are two examples. Additionally, radiation interactions in liquid media can also be initiated from high-energy photons (gamma particles). Therefore, an understanding of radiation effects requires multiple tools including test reactors, ion and electron beam facilities, and photon sources.

APS Advances Necessary to Meeting Science Needs in Surface/Interface Characterization

Uses of Synchrotron Facilities in Studying Radiation-induced Degradation

The problems enumerated above are all amenable to study using synchrotron radiation techniques. Synchrotron techniques can be used prior to irradiation to determine how structures, such as precipitates and defect traps, form during the manufacturing process. These features can also be studied as a function of manufacturing technique such a cold rolling, hot isostatic pressing, etc., prior to irradiation. Finally, evolution of defects, structural changes in precipitates, and the interaction of defects with traps can be tracked after irradiation using synchrotron radiation techniques.

In order to understand these mechanisms, the capability is needed to determine structural correlations over a broad range of length scales, from the atomic (1 Å) through to the macroscopic (mm). The structural and materials problems encountered in studying stress- or radiation-damaged materials would benefit from most of the currently available synchrotron

techniques which are broadly divided into three categories: imaging, scattering, and spectroscopy.

X-ray Imaging

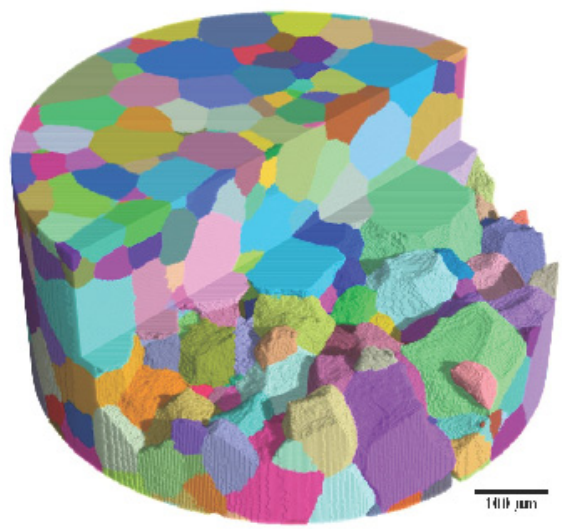
In X-ray imaging, two different approaches can be used to acquire images of the sample. In full-field methods, an extended field of view on the sample is illuminated and the whole area imaged, typically in one exposure. Images are often recorded with scintillator crystals and optical cameras. Spatial resolutions down to below 1 micron can be achieved with direct imaging; resolutions down to 30 nm can be achieved with additional X-ray magnification (Transmission X-ray Microscopy).

In scanning approaches hard X-rays are focused into a small spot through which the specimen is scanned and the image is built up sequentially. The achievable resolution is 100-200 nm in the hard X-ray range. In addition to use of the transmitted X-ray signal (absorption and phase contrast), one can make use of other contrast mechanisms such as X-ray fluorescence for element detection/identification/chemical state and X-ray diffraction for reciprocal-space information.

In the soft X-ray region, current tomography with resolution on the order of 50 nm is possible [33]. Unfortunately, soft X-rays do not have the penetration depth necessary for studies of either irradiated structure materials or fuels. In the hard X-ray region, sub-100 nm reconstruction is possible [34] at 9 keV with the possibility of pushing the resolution to 15 nm with much lower efficiency. Even at 9 keV, the attenuation length of the X-rays is on the order of 10 μm. This probe depth limitation precludes the study of bulk materials such as clad fuel pellets at these resolutions. The study of bulk irradiated materials requires incident photon energies between 30 and 100 keV, where the attenuation length can extend to 650 μ for spent fuel and as high as 3,400 μ for steels associated with reactor components.

This application requires the development of new optics that focus the incident X-ray beam to 10 nm. The need for high-energy tomography has broad interest and is not limited solely to applications in nuclear energy systems.

HARD X-RAY IMAGING



A rendition, obtained by Diffraction Contrast Tomography (DCT), of the three dimensional grain structure of a cylindrical specimen composed of 1008 individual beta-Ti grains. Whereas this image was obtained using 40 keV X-rays, samples such as spent fuel, which contain heavier atoms and are denser, would require even harder X-rays. H. Poulsen et al., Rev. Sci. Instr. 80 (2009) 33905.

Another complication of tomographic imaging of irradiated materials is the gamma ray emission from the sample. X-ray tomography is often performed with the detector in direct line of sight with the sample. The gamma emission reaching the detector has the potential of saturating the detector making imaging impossible. Recent advances coupling analyzer crystals [35] with tomographic imaging systems may allow the detector to be shielded, thereby making measurements on irradiated samples much more practical.

X-ray Scattering

X-rays elastically scatter off the electrons in a sample, a phenomenon that is dependent on the wavelength of the X-rays with an intensity that is proportional to the electron density. The scattered X-rays from one atom's electrons act as waves that interact with those scattered from a neighboring atom to set up interference patterns. The general result from X-ray scattering is pair correlations that can be interpreted in terms of species and relative location of the atomic pairs. For the general condition, as found in amorphous samples including solutions, the scattering data can be Fourier-transformed to reveal pair distribution functions that describe atomic correlations.

Small-angle X-ray Scattering (SAXS)

In addition to interference patterns set up by variations in atomic electron density, similar patterns are also set up by contrasts in electron density that occur over much larger length scales in a sample. Intensity variations caused by this scattering — observable at very low scattering angles — are valuable for determining grain and precipitate sizes. Of particular interest is information obtainable about bubble and void sizes on the order of 1-1000 nm.

The contrast in electron density seen by the X-rays occurs between the sample and the void, the latter of which has no electrons within its space. It is also possible to use incident X-ray energies just above and below element absorption edges. By using changes in absorption for a specific element, which selectively affects the scattered intensity arising from that element, anomalous small-angle X-ray scattering (ASAXS) provides information on the chemical composition of the precipitates or grains.

ASAX has been used to probe irradiated samples, specifically looking at chromium-phase segregation as a function of dose [36]. As recently demonstrated on cladding samples [37], the extension of this technique to higher energies can be used to probe the bulk of the sample. The use of high-energy incident photons would also permit ASAXS on higher Z materials, notably the actinide K-edges which begin at 110 keV for Th and increase monotonically with increasing atomic number.

SAXS measurements are collected using a two-dimensional imaging detector. These detectors, as were the tomography detectors, are located in direct line of site to the sample. Thus, it is possible for the two-dimensional detector to see γ-emission from the sample. Unlike the case in tomography, analyzer crystals cannot be used to separate the elastically scattered X-rays from the γ background. Detection will benefit from experiment geometry, in which the detector is located at significant distance from the sample. The collimation of the transmitted X-ray beam will

improve detection of the signal over background. These experiments would also benefit from higher count rate detectors and the possible development of imaging analyzer crystals.

X-ray Diffraction (XRD)

For crystalline systems, the long-range atomic order results in planes of electron density that coherently scatter to produce peaks in detected intensity as a function of the angle of incident to scattered X-rays. Following crystal-symmetry rules, the peak positions and their intensities can be analyzed to produce a detailed understanding of the atomic arrangement within the solid. This information can be compared to a database of known compounds to provide a chemical analysis of the studied material. Any structural changes due to irradiation such as phase segregation, phase change, and amorphitization [38] can be observed with XRD. Its sensitivity to small changes in long-range atomic order (>0.001 Å) can be used to measure the effects of residual stress [39] or under an applied stress [40] in any given material.

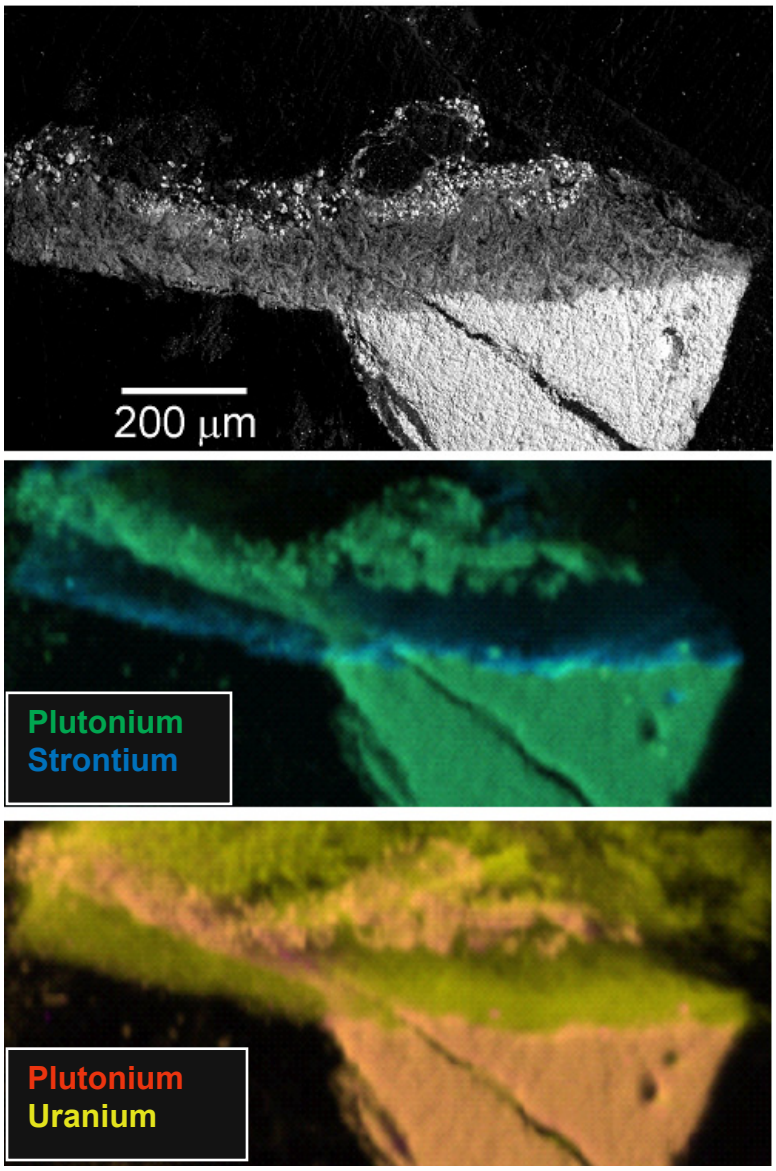
For example, residual stress from the manufacture of a clad fuel pellet may negatively impact its performance under irradiation. XRD measurements have the ability to measure and to map this residual stress to submillimeter resolution. Stress could arise during the irradiation process from differential swelling in clad materials. Measurement on irradiated materials will allow the quantification of any residual stress arising from the irradiation. Stress-strain measurements can be used to determine loss of structural integrity post-irradiation. Annealing during these measurements can be used to show when the initial structure [36] of the material is regained.

X-ray scattering measurements can be made with either a point detector or two-dimensional imaging detectors. The point detection measurements can be made with an analyzer crystal. The analyzer crystal can be used to prevent the sample's γ-emission from reaching the detector, which allows for higher activity samples to be measured. The two-dimensional array detectors are similar to the SAXS detectors and it is unclear how these detectors can be protected from gamma emission from the sample.

X-ray Spectroscopies

X-ray absorption spectroscopy (XAS or XAFS), comprised of both XANES and EXAFS, can be used to simultaneously measure the chemical state of atoms in a solid, liquid, or gas, and the local atomic structure around the absorbing atom. XAS can be used to reveal the average speciation of a targeted (absorbing) element at low concentrations (ppm) in complex materials such as those encountered during nuclear energy systems, including radiation damaged materials such as spent nuclear fuels, their claddings, and reactor components [41]. The influence of temperature, stress, and local compositions on variation of local chemical environments can be measured *in situ* by XAS. However, because the X-ray energy is constrained to the absorption edge of interest with a practical high-energy limit of about 40 keV, the radiation does not penetrate deep into the sample and consequently the technique is often restricted to near-surface studies. With this caveat, XAS spectra have been shown to be very sensitive to the effects of radiation damage [42].

PROBING AQUEOUS CORROSION OF IRRADIATED OXIDE NUCLEAR FUEL



Specimens of irradiated oxide nuclear fuel subjected to aqueous corrosion tests for 10 years were epoxy-embedded and cross-sectioned in a hot cell for this investigation. A small core (plug) was extracted and re-embedded in epoxy to form a specimen that was small enough to handle safely. A backscatter SEM micrograph (top) revealed intact fuel (light colored) with a 100-micrometer corrosion layer of uranyl silicate (gray). X-ray fluorescence mapping and edge spectroscopy were employed to investigate the local chemical environment of key elemental components. X-ray imaging of plutonium, strontium, and uranium (center and bottom) show their relative distribution near the oxidation front [41].

XAS can be measured with a variety of monitoring methods including electron-, fluorescence-, and Auger-yield measurements. Many of these detectors are less susceptible to saturation from γ photons from the sample. Fluorescence measurements can be collected using a Bent Laue detector [43] which uses a bent silicon crystal as an analyzer to prevent the photon detector from seeing the sample directly.

The Bulk Properties breakout session also suggested that soft X-rays could be used for "spectroscopic calorimetry" with the goal of improved simulation of nuclear materials via experimental benchmarking. Examples would include issues such as thermodynamic instabilities in nuclear fuels under irradiation, such as the determination of heats of solution from experimentally determined, elementally specific, binding-energy shifts [44].

In a break with past paradigms, the U.S. Department of Energy has proposed a novel approach to the development of advanced nuclear fuels — predictive numerical simulation [44]. The advent of massively parallel computing and other improvements in computation capabilities has opened the door to the possibility of simulating much of the work that would have necessarily been determined empirically in the past. Nevertheless, these simulations and projections require the input of fundamental physical parameters that are experimentally generated or at the very least benchmarked. In particular, there is a dearth of fundamental thermodynamic information.

To remedy this, a radical departure from past practices of calorimetry was proposed during the workshop. Using the techniques first proposed by Martensson and Johansson [45] and then validated by Steiner et al [46], it is possible to use spectroscopically determined core level shifts to benchmark the computationally generated heats of solution. These measurements will be compared directly to the predictions of heats of solution from *ab initio* and CALPHAD calculations being conducted in complementary projects.

Owing to resolution (100 meV bandpass) and intensity arguments (1% alloy compositions), only soft X-ray (500-1000 eV) photoelectron spectroscopy will work. Furthermore, this requires "exposed samples" where triple containment is impossible.

Summary

Synchrotron radiation-based research programs may have a significant scientific and technological impact on the development of advanced materials and fuels for nuclear systems. This includes the promising applications of synchrotron X-ray techniques in characterizing microstructural evolution and associated physical and mechanical properties of materials under irradiation.

Needs for a Separate Irradiation Station and Potential Challenges in Using Synchrotron User Facilities to Examine Radiation Damage

In many instances, the APS is adequate to answer important scientific questions related to radiation damage as long as appropriate radiation protection protocols can be established. Participants noted a few cases where a dedicated beamline may be necessary, specifically:

- High-temperature and tensile testing with environmental control, high-pressure and autoclave testing which may be incompatible with encapsulation rules; and
- Isolation of very high radiation levels or potential releasable contamination (tritium, fission pressure gas) from the main beamline is of too high a probability.

In some cases, an advantage of an isolated beamline is the improved imaging, space, and angular separation that would be a side benefit provided by a separate building.

The facility would necessarily have to operate in a manner different from the other APS beamlines. The complex radiation handling requirements of the irradiated specimens make it difficult for researchers unfamiliar with the facility to operate the equipment without assistance. One of several possible scenarios has beamline scientists and technical staff members operating the beamline equipment for the general user community.

This would not be a facility designed for the study of environmental radioactive samples and low-activity samples currently studied at the APS. Instead, a dedicated facility would focus on higher-activity samples and those with special containment issues, including heavy shielding.

Challenges to Routine Examination of Radioactive Material in APS

A number of challenges to the use of APS for the examination of radioactive material were identified:

1) Current plans limit radioisotope quantities for synchrotron experiments to those defined under DOE regulations for a Radiological Facility. There needs to be a comprehensive risk management strategy that is *attractive to users (e.g., well understood safety envelopes for specific sample types)*.

2) The facility needs to improve its ability to handle increasingly more complex samples with higher levels of radioactivity or samples that are difficult to encapsulate and still do the required study, such as the challenges presented by *in situ* wet chemistry experiments.

3) In order to analyze highly radioactive samples with synchrotron radiation techniques, a program to develop detectors that are robust under radiation is required.

4) There is no apparent formal connection to a facility that can section small pieces of radioactive material.

5) In many cases, size reduction to lower radiation levels is possible; it is not always possible to trade size for activity.

ASSESSMENT OF THE APS' RADIOACTIVE SAMPLE QUANTITY LIMITS

A major discussion point during the workshop and in the subsequent Breakout Session reports focused on the physical quantity of radiological samples permitted by the APS for a single experiment. The concern arises from the DOE Hazard Categorization regulations for a Radiological Facility, which are described in the DOE document DOE-STD-1027-92 (available on the web). The participants were greatly concerned that limiting the APS to less than a Hazard Category III Nuclear Facility would significantly impact the experiments that could be performed and underlie arguments in favor of constructing a Nuclear Facility in a building adjacent to the APS. The discussion included the possibility of a beamline extending from the APS ring to the adjacent building where the experimental hutch would be located. This arrangement would permit working with higher levels of activity while limiting the APS' liability in the event of a contamination incident.

Primarily, it was the participants wishing to study spent nuclear fuel who expressed these facility-limit concerns. The specific activities of their samples combined with the expressed needs for macroscopic quantities suggest that these experiments may constitute the limiting example. With these concerns in mind, calculations were performed to assess the impact of the APS restrictions on spent-fuel sample sizes to less than Category III quantities. A description of the samples chosen for these calculations and the results are discussed below.

A scenario is presented that quantifies sample limits imposed by such a restriction. This exercise involves the calculation of the radioisotopes present in a spent-fuel pellet. Specifically, the computer code ORIGEN was used to simulate the isotopic distribution [47] in a one-gram fuel pellet with starting composition of UO_2 that had been enriched by the addition of either 5% or 10% of ^{235}U. The scenario included burn-ups to 2%, 5%, or 10% assuming light-water reactor conditions. After irradiation, the code produced a list of the isotopes and their quantities that would be present in the spent fuel as the irradiated fuel was left to cool for specified times. These calculations assumed retention in the sample of all isotopes produced, including gases such as tritium and Xe that could be expected to be volatile. The isotopes and their relative quantities determined by these simulations were then multiplied by their Hazard Category III threshold values as described in DOE-STD-1027-92 and these values summed to give the Hazard Category ratios listed in Table 1. It is understood that users may wish to bring samples to the APS that are very different from the example chosen here and that new calculations will be needed. This example is considered to be a high-risk scenario for demonstration purposes only.

The numbers in Table 1 represent the ratios calculated for one gram of spent fuel as discussed above. Overall, a Radiological Facility cannot exceed one for the sum of the ratios of all the samples within its boundaries at any given time. For example, such a facility could accommodate simultaneously two one-gram samples, both starting with 10% enrichment, one irradiated to 5% burn-up and the second to 2% burn-up after both were allowed to cool for 30 days. The sum of ratios, 0.4216 + 0.4955, is less than 1.0. From an alternative perspective, the Facility could accommodate a single sample representative of 129 grams of UO_2 fuel originally 5% enriched in ^{235}U, consumed in the reactor to 2% burn-up, and then cooled for 10 years.

Table 1. Hazard Category III Ratio for a pellet initially containing one gram of UO_2 fuel at the specified enrichment and burn-up percentages. The contribution from the cladding (simple Zircaloy in this calculation) is not included in these calculations and at times beyond one year does not contribute significantly to these values. DOE regulations require that the ratio summed over all samples in a Radiological Facility cannot exceed one [48].

Fuel Enrichment Burn-up	Post-irradiation Cool Down			
	30 days	1 year	10 years	20 years
10:10	0.3323	0.1259	0.0611	0.05300
10:5	0.4216	0.05158	0.01789	0.01517
10:2	0.4955	0.01998	0.006051	0.00496
5:2	0.4912	0.02443	0.007726	0.00680

Following on with this specific example, the next question to be addressed is whether restricting this spent-fuel sample to 129 grams would compromise the synchrotron experiment. This question is approached by factoring in the amount of sample that can be accessed in an experiment. X-rays are attenuated as they pass through materials. Depending on details of the experiment (reflection of transmission geometry, for example) the interior of a sample may not contribute to the observed signal. The attenuation of X-rays is dependent on their energy and the electron density through which they pass and, in turn, depends on the elemental constituents of the sample and its density. The density of unirradiated UO_2 is 10.96 g/cm^3. The attenuation length — the distance an X-ray beam travels through a sample before losing 67% of its initial intensity — is strongly dependent on energy.

As shown in Table 2, a 5 keV X-ray beam has an attenuation length of 1.2 microns; 650 microns are required for the same attenuation at 100 keV. Assuming a cylinder 5λ in height (0.325 cm) because a thicker sample would not be probed by the X-ray beam (liberal estimate) and applying $V = \pi r^2 h$, a radiological facility could accommodate a cross-section sample with a diameter of 3.42 cm. This diameter is much larger than the footprint of the X-ray beam on sample, even for glancing-incident surface experiments. It is also larger than the diameter expected for a spent-fuel pellet, even from some of the new fuel designs under consideration from an advanced reactor design. Thus, this calculation indicates that a section from the spent-fuel pellet used in this example could be accommodated at the APS without exceeding the DOE Hazard Category III threshold limit.

Our one gram reference UO_2 sample with 5% enrichment and 2% burn-up that was allowed to cool for 10 years emits 4.09×10^9 photons per second, approximately 110 mCi of gamma-ray and X-ray activity. The 129-gram sample permitted under the Hazard Categorization criteria would have approximately 14 Ci (14,000 mCi) of gamma-ray and X-ray activity.

Table 2. Selected attenuation length (λ)'s representing the penetration depth of an X-ray into the material before the intensity is reduced to 1/e of its intensity at the surface. Densities used for calculations are 10.96 g/cm^3 for UO_2 and 7.86 g/cm^3 for stainless steel. Data obtained from http://physics.nist.gov/PhysRefData/FFast/html/form.html

Material	λ(microns) 5 kev X-rays	λ(microns) 50 kev X-rays	λ(microns) 100 kev X-rays
UO_2	1.2	100	650
Stainless steel	6.5	700	3400

The software code MicroShield was used to estimate the amount of Pb shielding required to reduce the estimated radiation field to less than 5 mR/hr at a distance of 30 cm, the DOE limit above which the enclosure housing the sample and its shielding must be posted as a Radiation Area [49]. The calculations indicated that 8 to 17 cm of Pb would be required, with the numbers corresponding to the limiting cases of 1 and 129 grams of sample respectively. Assuming spherical symmetry the shielding would weigh approximately 25 to 230 kg.

These examples are meant to represent spent fuel in general and demonstrate that the primary problem facing an experimenter is not the constraints of a DOE Category III Nuclear Facility but instead is the necessity of shielding the radiation emanating from such a sample. This problem takes three forms:

1. Protecting the individual from a radiation dose. The amount of shielding required to limit the dose poses significant handling and safety issues because of its weight,

2. Designing shielding with appropriated located window to allow the synchrotron radiation to impinge on the sample and be detected appropriately while still shielding the experimenter. Associated with this issue is the design of shielding with remotely removable windows.

3. Discriminating the synchrotron radiation associated with the experiment from the background radiation. Because the X-rays from the sample have such a broad energy distribution this will not be a trivial problem and will require detector development.

WORKSHOP CONSIDERATIONS AND RECOMMENDATIONS

As demonstrated in this report, the community interested in advanced nuclear energy systems is very diverse, ranging from nuclear engineers to environmental geochemists. This is reflected in the wide range of interest expressed for a diversity of synchrotron experiments.

Many workshop participants have had considerable experience working at a synchrotron while an almost equal number have interest but little experience. For a significant number of the latter group, their inexperience arises in part because their experimental needs cannot currently be met by the capabilities of APS or any other synchrotron. This group includes those interested in studying materials properties of reactor materials, used fuels, and waste forms, samples of which may be highly radioactive and/or be highly dense.

Currently, X-ray availabilities for radioactive samples are practically limited to energies less than ~35 keV. Because of the high absorption of these X-rays, they effectively probe only near-surface sample volumes. However, some of the most pressing questions now being posed by the nuclear energy systems community involve the need to probe varying length scales in the bulk. In some cases this difficulty can be resolved by preparing thin-sections but in so doing concerns are raised about the effects of sample preparation on the features of interest. The extension to X-ray energies greater than 60 keV would result in increased X-ray penetration lengths that would probe the bulk sample, diminishing the need for extensive and costly sample preparation. Such extended capabilities are particularly important when studying materials issues such as void formation, stress corrosion cracking, and sample inhomogeneities, including grain boundaries. Extensive sample preparation in these cases can alter the materials properties of interest.

Regardless of participant experience level, there is unanimous agreement that synchrotron studies hold enormous potential to answer questions central to all aspects of nuclear energy production: from validation of simulations, to quantifying reactor materials problems, to environmental contamination and remediation. Specific considerations and recommendations follow.

CONSIDERATIONS

Sample Size Restrictions

The nuclear community has expressed concern that the current limits on sample quantities will adversely impact the kinds of experiments that are possible at the APS. The possibility of the construction of a DOE Nuclear Facility was discussed as a route to enabling the larger sample sizes thought to be necessary to run some experiments. This possibility was particularly noted with respect to samples involving radiation damage and spent fuel. Unfortunately, at the time of the meeting there was no clear understanding of DOE Hazard Categorization restrictions on sample sizes and their impact on experiments.

Subsequent calculations on a model system (UO_2 spent fuel from an LWR), which are detailed in this report, indicate that sample size is not expected to be a limiting factor from this perspective.

However, it is conceivable that a single sample could significantly impact the ability of the facility to accept other samples, thereby requiring care in the scheduling at the APS to assure that Radiological Facility limits are not exceeded.

Not discussed at the workshop is the clear hazard such a spent-fuel sample would pose in terms of the radiation field in its proximity and accumulation of radiation doses above reasonable ALARA limits for those in the vicinity, whether or not they are directly involved in the experiment. Appropriate shielding can mitigate this dose, although calculations suggest that it may need to be very heavy, requiring cranes or other mechanical assistance to move. This would be difficult to accommodate on some beamlines. In addition, the design of shielding with appropriately removable windows would require significant engineering.

The construction of a Nuclear Facility is not thought to provide the highest impact for resource allocation given the enormous costs and regulatory burdens associated with operating a Nuclear Facility and because a clear need was not demonstrated for sample sizes that exceed the restrictions imposed by a DOE Radiological Facility. The accompanying recommendation is that the current APS experimental safety protocols be made more accommodating for larger sample sizes. This could be accomplished in part through an enhancement in capabilities and technical management within the restrictions imposed by a Radiological Facility.

Current APS Capabilities for Sample Handling

The APS requires that radioactive samples be adequately contained at all times while in the experimental facility. The determination of what constitutes adequate containment is decided by APS safety staff and by a consideration of the isotope, its quantity, the form of the sample (solid, liquid), and ALARA criteria. Samples can be mailed to the APS, where packages are opened and containment checked for external contamination before they are forwarded to the beamline. After the experiment, all samples must be returned to the APS for shipment back to the home institution; there is no option available for sample storage. APS categorizes as user support the on-site sample handling and transport, as well as packaging for off-site transport. This work is done by APS support personnel and is paid for by the APS. Prior to sample delivery, arrangements can also be made for sample disposal by Argonne following the experiment. However this cost, together with the cost of off-site transport, must be borne by the user.

Currently, arrangements can be made through the APS to breach containment by transporting the samples to the Argonne's Actinide Facility located in Building 200. This facility includes a suite of radiological laboratories fully equipped to handle open samples and safely deal with loose activity. If the need arises, more extensive preparatory work can be done the Actinide Facility after prearrangement. Actinide Facility support can include including sample preparation; wet chemistry; crystal re-orientation; containment evacuation; and more detailed characterization such as optical spectroscopy or in-house crystallographic work such as single crystal alignment. The facility and preparatory work is provided free of charge and is widely used by the research community.

There are two major drawbacks to using the Actinide Facility:

- Its services are available only during regular working hours. Additionally, Argonne's Materials Controls and Accountability (MCA) Group must transport samples to and from the Actinide Facility. The MCA Group's availability can further restrict sample transport hours. This restricts any APS user who may have an urgent need for manipulating or repackaging the primary sample.

- Its laboratories are aging and, past a 5-year timeframe, it is unclear how long their capabilities will be available to the APS users. Considerations of alternative options for handling radioactive samples at the APS should be underway in order to maintain this capability into the future.

Radioactive Sample Capabilities at Other Synchrotron Facilities

Experimental capabilities currently exist for performing X-ray experiments at a number of other national and international synchrotron facilities. Within the U.S., the major hard X-ray synchrotron light sources — Stanford Synchrotron Radiation Lightsource (SSRL) and National Synchrotron Light Source (NSLS) — also permit radioactive samples on the experimental floor and like the APS, require that they are encapsulated at all times. The SSRL has a large program in synchrotron research on nuclear materials and will permit significant quantities of materials to enter the facility. Neither of these light sources have on-site capabilities available for handling open samples. Because of technical considerations associated with X-ray energies, the Advanced Light Source (ALS) permits experiments on un-encapsulated radioactive samples but has significant safety protocols for sample handling as well as restrictions on the activity, size, and form of the samples that all must be individually determined through an appropriate safety review.

Internationally, there are several synchrotron facilities that permit radionuclides on their experimental floors, with beamlines specifically designed for use with radionuclides. One facility, the European Synchrotron Research Facility (ESRF) has a beamline that is optimized for spectroscopy in the energy range appropriate for work on Th-Pu. Although ESRF has a glove box associated with the beamline, un-encapsulated samples are not permitted in the facility and the capability to handle open samples is not available even in the glovebox. The Source Optimisée de Lumière d'Énergie Intermédiaire du LURE (SOLEIL) facility, located near Paris, has recently opened MARS, a beamline for multidisciplinary research on radioactive matter. This medium energy beamline will permit macroscopic sample sizes on the order of those permitted in a DOE Radiological Facility. With the exception of samples associated with cryogenic experiments that have to be placed in special sample containers and backfilled with inert gas, MARS does not have the capability to handle open samples. These three international lightsources all have available X-ray energies within the range of about 5-35 keV.

RECOMMENDATIONS

Continued Use of APS Beamlines

The user community expressed an overwhelming priority for the continued full access to the APS' suite of state-of-the-art beamlines. Stated another way, a single "radioactive beamline" or even a "radioactive sector" meant to accommodate all nuclear samples would not meet the needs of this community. The present APS operating mode works on a "distributed model," in which access is granted to all beamlines through the General User Proposal system. This system considers the technical merit of the proposal and assigns beam time in a process independent of safety aspects of the proposed work. In this way, radiological work is given equitable access to the facilities and containment efforts can be focused on approved proposals. At the time of the workshop, about 1,000 experimental safety protocols have been approved for work with radiological samples on approximately two-thirds of the operating beamlines. The resulting data acquisition has had a significant impact over the broad scope of materials, physical, and chemical problems relevant to the community. Continuing unfettered beamline access will allow nuclear systems to continue advancing as a component of the national energy systems portfolio.

Safety Reviews for Radiological Samples

There is widespread confusion about what is and is not acceptable in terms of allowable isotopes, quantities, and encapsulation for radioactive samples. The user community comment that improved communication with the APS is necessary in this area. Suggestions include an improved web site with scenario guidelines, example safety envelopes, and a more detailed description of the risk-based approach used to determine safety protocols. The community also voiced a need for knowledgeable, readily identifiable APS staff with whom they can interact to design an experiment that will meet with APS approval.

Sample Containment and Hardware Development

All interest groups cited sample handling and containment assistance as critical to overcoming the administrative and technical hurdles of working with radiological samples in a DOE facility. At Argonne, sample handling and encapsulation requirements necessitate that APS provides assistance to meet its safety requirements while permitting the data acquisition defined in the approved user proposal. The user community rates as a high priority the investment of resources in the design, engineering, and construction of standardized sample containment vessels that are preapproved and available for use.

With its technical expertise and access to the broad user community, the APS is in a unique position to lead the development of standardized sample containment and shielding. There is an unmet need for leadership in a team approach to overcoming technical obstacles. An example is the recent collaborative design work done by the geochemical community on the *in-situ* surface cell (see Developing a Surface and Interface Scattering Sample Cell, Page 9) This was handled under an *ad hoc* arrangement involving a group of users and lead by ANL safety staff that could

have been significantly strengthened and streamlined by a formalized APS involvement and leadership.

The overall issue of sample containment and safety protocol management is one of the major stumbling blocks for researchers wishing to access synchrotron user facilities. The user community sees as a high-impact recommendation the direct involvement and application of resources for sample manipulation such as Argonne's Actinide Facility (a BES Chemical Sciences funded effort that assists users in running radioactive samples at the APS), the Irradiated Materials Laboratory, and other on-site radiological facilities. Assistance from APS in overcoming obstacles is expected to result in an influx of researchers anxious to take advantage of the opportunities afforded to their scientific and engineering programs by synchrotron light.

Effectively shielding the experimenter and the detectors from the sample's radiation is also part of the containment problem. Effective shielding can be critical to the success of an experiment, particularly those involving irradiated fuel samples. Detectors can be swamped by stray radiation that affects their linear response and, in some cases, prevents the acquisition of data with adequate statistical quality. By involving APS engineering and beamline staff familiar with the proposed experiments, the development of effective shielding could be greatly enhanced.

The user community also recommends the development of advanced detector hardware. Although many experiments can be performed in ways that shield detectors from sample-induced radiation, there remain experiments — notably transmission imaging — that require advanced detector development before they can be performed on radioactive samples such as spent fuel. Necessary improvements to the detectors include increased count rates, higher dynamic range, and improved resolution.

Dedicated Facility

Based on the considerations and recommendations listed above, the community argues for a dedicated DOE Radiological Facility located at Argonne. This facility would enable experiments on advanced nuclear energy systems, increase sample accessibility, and encourage new users by enhancing and securing sample handling capabilities into the future. Argonne has extensive experience and training in handling and managing radioactive materials and experiments, the required radioisotope permits and shipping certifications, an experienced health physics organization, and an infrastructure for monitoring and tracking radiological materials. As part of the APS user support, these capabilities should be made available to the experimenter through the existing infrastructure at Argonne.

Optimally, the dedicated facility would consist of a stand-alone, independent building located adjacent to the APS. This location would permit sample transfers between the two buildings without the need for special transportation while minimizing the risk of radioactive contamination within the APS itself. A stand-alone facility would reduce experiment-scheduling problems by removing from the experimental floor the sample packaging and handling; creating two independent facilities and thereby increasing the total permissible radioactive inventory. The goal would be access to sample-handling capabilities, not only during normal working hours, but also during the evenings and on weekends — viewed as a critical need for all APS experiments.

The capability of handling and moving heavy samples in their containment should be a key part of the facility. This capability would be required for experiments on irradiated fuel samples that need extensive shielding to meet ALARA safety regulations.

For users taking advantage of any of the APS beamlines, the new facility should provide the opportunity to perform simple chemistry and sample loading into primary containment. For example, the loading of radiological materials into cells designed for mineral-water surface scattering studies (see Developing a Surface and Interface Scattering Sample Cell, Page 9). These capabilities are not available elsewhere and should open new opportunities for experiments that cannot be prepackaged and transported to the synchrotron, such as *in situ* surface studies, electrochemical experiments, and time-sensitive chemistry studies including kinetics measurements.

The majority of the workshop participants recommend that all extensive sample preparations — including the cutting and polishing of spent-fuel samples — be performed prior to transport to this facility. Such preparative procedures would have a significant burden on the new facility infrastructure without a commensurate benefit. Other DOE facilities, including some located at Argonne, are able to perform such procedures and may enter into an agreement with APS for doing so. There are no clear experimental benefits for locating these specialized capabilities next to the APS.

A new dedicated facility would serve as a home for the advanced nuclear energy systems community; this would be amongst its most important functions. The very diverse user community shares many complex problems associated with conducting challenging experiments on intractable samples. A common goal is overcoming experimental obstacles including transportation requirements, assessing sample quantities, suitable materials for encapsulation, encapsulation requirements imposed by the APS, sample-containment engineering, experimental geometry optimization, and detector issues. A facility having experienced staff knowledgeable in radioactive sample management within the specialized context of synchrotron experiments would be of enormous assistance to this community — particularly to new users who face these complications in addition to the usual hurdles involved in accessing a big user facility. Synchrotron radiation's potential for providing otherwise unavailable information to address problems of importance to the nuclear energy community is clear. The localization of a knowledgeable staff in a facility dedicated to helping the user community overcome their unique technical and administrative hurdles is a critical need.

Specialized Beamlines

The community interested in performing experiments on radioactive samples associated with advanced nuclear energy systems has expressed interest and need in a wide variety of the capabilities and techniques offered by synchrotron experiments. Many of these experiments are possible on more traditional samples but require added engineering before they can be performed on radioactive samples. However, some problems and questions posed by advanced nuclear energy systems cannot be addressed at the APS or at any other synchrotron at this time.

Many of these problems are associated with fuels, structural materials, and wasteforms exposed to extreme conditions of temperature, thermal gradients, and radiation fields. Current approaches to understanding nuclear energy related materials' behaviors involve multiple tools, including test reactors, ion and electron beam facilities, and techniques such as electron microscopy and atom probe tomography. With notable exceptions, studies at a synchrotron have been limited to unirradiated samples, despite the obvious opportunity and potential for the validation of ongoing theory and modeling efforts.

As discussed above, while part of the reason for the absence of synchrotron techniques in the nuclear energy portfolio is the problems with sample handling, the more fundamental issue concerns the absence of experiments that can adequately probe the bulk of the sample without the need for extensive sample preparation. The particular challenge for much of the nuclear energy work on irradiated samples or wasteforms is their primary composition of densely packed, high Z (atomic number) elements that strongly absorb X-rays. High-energy photons would be better able to probe the bulk of the material to provide *in situ* characterization of voids, cracks, inhomogeneities, interfaces, and chemical segregation. Transmission experiments could take advantage of the use of larger (thicker) samples, which implies heavier shielding around the sample with engineered windows for the synchrotron beam.

Some capabilities currently exist for using high-energy X-rays to probe samples of interest to this community. These include stations at Sector 11 that are currently being used for obtaining pair distributions used in studying solution and amorphous solids and largely associated with separations and environmentally relevant problems. Also, Sectors 1 and 6 have some high-energy capabilities that include pair distribution and small angle scattering capabilities. The work on these beamlines has been more focused on materials-related concerns.

The availability of the APS' higher energy X-rays (>60 keV) provides an unrealized opportunity to address many of the issues involving sample inhomogeneties at varying length scales — from scattering experiments that look at atomistic correlations, to SAXS probing bubble formation, and to tomography and imaging studies. Some of these techniques are of interest to other materials science communities as well and are being addressed as new opportunities in the APS upgrade. This communities needs focus on high-energy tomography, full-field imaging, spectromicroscopy, fluorescence imaging, and high-energy wide- and small-angle scattering. The high radiation fields emitted by many of the samples will require an insertion device to provide sufficient X-rays to overcome issues with counting statistics. The combined need for a high intensity, high energy beam with some tunability argues for a superconducting undulator.

There can be an argument made, based on the specialized equipment needed to handle and position these heavier samples, for the construction of high-energy beamlines within the dedicated facility that would be focused on a subset of the experiments on nuclear energy materials. The construction of such beamlines within the facility would accommodate relevant requirements associated with nuclear materials handling (radiation levels, safety, and security) and would provide capabilities unavailable anywhere else in the world. The proposed high-energy capabilities are expected to be consistent with the overall upgrade plans of the APS and would serve to highlight their strengths to a broader community.

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APPENDIX I: WORKSHOP PROGRAM

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems

27 to 28 January 2010
at Argonne's Advance Photon Source

Scope

Third-generation synchrotron facilities offer unprecedented opportunities for ultra-small, ultra-fast and *in situ* measurements with high-energy, high-brilliance, x-ray beams. The capabilities provided by synchrotron radiation have had an enormous impact on resolving forefront scientific issues in the area of advanced nuclear energy systems, despite the technical difficulties in working radiological samples at a synchrotron facility. This workshop will bring together leading researchers in the field, with the following objectives:

- To discuss important scientific and technological issues in the all areas of nuclear-energy systems, including fundamental electronic properties, radiological materials characterization, radiation damage studies, solution and separation chemistries, and radionuclide behavior relevant to the geosphere, that require synchrotron radiation techniques, and the potential of synchrotron radiation to address these issues.
- To encourage communication between nuclear-energy researchers and synchrotron experts to optimize the use of synchrotron techniques for discovery in areas related to advanced nuclear energy systems.
- To identify the user-community's future needs for synchrotron radiation facilities, address scientific questions of importance in nuclear-energy technology and examine the adequacy of current capabilities and the need for new facilities.

This workshop will provides an important opportunity to assist the APS with identifying their current and future synchrotron capabilities in the context of solving the pressing scientific and technological problems in advanced nuclear energy systems.

Argonne NATIONAL LABORATORY

U.S. DEPARTMENT OF ENERGY

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems

27 to 28 January 2010
at Argonne's Advance Photon Source

Technical Program and Themes

Keynote Speakers

Gerry Lander (ITM, retired) – Probing Condensed Matter

Steve Zinkle (ORNL) – Radiation Damage in Fuels and Structural Materials

Mark Antonio (ANL/CSE) – Solution and Separations Studies

Topical areas include

- Basic actinide and fission product chemistry
- Electronic properties
- Materials characterization
- Pressure studies
- Radiation damage studies
- Separation chemistry
- Radionuclides in the environment

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APPENDIX II: WORKSHOP AGENDA

Workshop on the Role of Synchrotron Radiation in Solving Scientific Challenges in Advanced Nuclear Energy Systems

27 to 28 January 2010

at Argonne's Advanced Photon Source

27 January 2009

7:45	Registration APS Lobby; Continental Breakfast
8:15 – 8:25	**Al Sattelberger** (Associate Laboratory Director ESE) Welcome
8:25 – 9:00	**Murray Gibson** (Associate Laboratory Director APS) Welcome **Denny Mills** (APS, ANL) Introduction to the APS
9:00 – 9:50	**Gerry Lander** (ITU, retired) Research on transuranium systems: present capabilities with large central facilities
9:50 – 10:10	Break
10:10 – 11:00	**Steve Zinkle** (ORNL) Radiation effects in nuclear structural materials and advanced fuels
11:00 – 11:50	**Mark Antonio** (ANL) Probing structural, morphological, and electrochemical aspects of actinide speciation through synchrotron studies

11:50 – 12:00	**Charge to working groups**
12:00 – 1:00	Working Lunch
1:00 – 1:40	**Henning Paulsen** (DTU-Risø) 4D tools for studying microstructure dynamics
1:40 – 2:20	**Gene Ice** (ORNL) Microbeam, timing, and wavelength dispersive studies of nuclear materials using 3rd and 4th generation synchrotron sources
2:20 – 3:00	**Philippe Martin** (CEA, France) XAS characterization of nuclear fuels
3:00 – 3:20	Break
3:20 – 4:00	**Jon Carmack** (INL) Fuel Cycle R & D Advanced Fuel Development
4:00 – 4:40	**Paul Fenter** (ANL) In-situ imaging of structures and processes at interfaces with x-rays
4:45 – 5:15	**Breakout Session**
5:30	**Poster Session** – Gallery

28 January 2009

7:30	Continental Breakfast
8:00 – 8:40	**Todd Allen** (U. Wisconsin, ATR/INL) Radiation effect studies for ANS systems and the role of national user facilities
8:40 – 9:20	**James Tobin** (LLNL) A path forward to advanced nuclear fuels
9:20 – 9:50	**Lynda Soderholm** (ANL) Experiments with radioactive samples at the APS
9:50 – 10:00	Break
10:00 – 11:00	**Panel Discussion**
11:00 – 3:30	**Breakout Session**
	Working Lunch
3:30 – 4:00	**Report Summary**
4:00	Close

APPENDIX III: WORKSHOP ATTENDEES

First Name	Last Name	Organization
Todd	Allen	U. Wisconsin
Mark	Antonio	Argonne
Motta	Arthur	Penn State University
Chi Bum	Bahn	Argonne
Mark	Bourke	LANL
Don	Brown	LANL
Wei	Bu	U. Illinois, Chicago
William (Jon)	Carmack	INL
Carlos	Castano	Missouri University of Science and technology
Yiren	Chen	Argonne
Wei-Ying	Chen	U. Illinois
Xiang	Chen	U. Illinois
Lidens	Cheng	Illinois Institute of Technology
Rob	Crowell	Brookhaven National Laboratory
Csilla	Czako	Illinois Institute of Technology
Robert	Daum	EPRI
Norman	Edelstein	LBNL
Mohamed	Elbakhshwan	U. Illinois
Paul	Fenter	Argonne
Kondev	Filip	Argonne
Paul	Forster	U. Nevada, Las Vegas
Jeffrey	Fortner	Argonne
Lynn	Francesconi	Hunter College of the City University of New York
Ben	Freemire	Illinois Institute of Technology
Hasitha	Ganegoda	Illinois Institute of Technology
Murry	Gibson	Argonne
Bruce	Glagola	Argonne
Mike	Goff	DOE-NE/INL
Chris	Grandy	Argonne
Dean	Haeffner	Argonne

First Name	Last Name	Organization
Daniel	Haskel	Argonne
Steve	Heald	Argonne
Brent	Heuser	U. Illinois
David	Hobbs	Savannah River National Laboratory
Jim	Ibers	Northwestern University
Gene	Ice	ORNL
Thomas	Irving	Illinois Institute of Technology
Chris	Jacobsen	Argonne
Mark	Jensen	Argonne
Qingying	Jia	Illinois Institute of Technology
Geng Bang	Jin	Argonne
Almer	Jon	Argonne
John	Joyce	LANL
Michael	Kaminski	Argonne
Djamel	Kaoumi	U. South Carolina
William	Kerlin	U. Nevada
Hussein	Khalil	Argonne
Marquis	Kirk	Argonne
Lukasz	Koscielski	Northwestern University
Jeremy	Kropf	Argonne
Gerard	Lander	ITU
Maik	Lang	U. Michigan
Sang Soo	Lee	Argonne
Meimei	Li	Argonne
Shanshan	Liu	Illinois Institute of Technology
Guangming	Luo	Argonne
Philippe	Martin	CEA
Mitch	Meyer	INL
Dennis	Mills	Argonne
Kun	Mo	U. Illinois
Lester	Morss	US DOE BES
Raluca	Musat	Brookhaven National Laboratory
Kathryn	Nagy	U. Illinois at Chicago

First Name	Last Name	Organization
Ken	Natesan	Argonne
Maria	Okuniewski	INL
Daniel	Olive	Illinois Institute of Technology
Luis	Ortega	Argonne
Alena	Paulenova	Oregon State University
Candido	Pereira	Argonne
Frederic	Poineau	U. Nevada, Las Vegas
Chrystian	Posada	Missouri University of Science and technology
Henning Friis	Poulsen	Risoe National Lab for Sustainable Energy
Andrew	Quicksall	U. Notre Dame
John	Quintana	Argonne
Donald	Reed	LANL
Daniel	Rego	U. Nevada
Michael	Ricnmann	LANL
Jessika Viviana	Rojas Marin	Missouri University of Science and technology
Tarik	Saleh	LANL
Al	Sattelberger	Argonne
Moritz	Schmidt	Argonne
Lee	Schroeder	LBNL and TechSource
Carlo	Segre	Illinois Institute of Technology
Brandon	Seilhan	Illinois Institute of Technology
Guoyin	Shen	Carnegie Institution of Washington
S.	Skanthakumar	Argonne
James	Smith	INL
Lynda	Soderholm	Argonne

First Name	Last Name	Organization
Tanju	Sofu	Argonne
Marius	Stan	LANL
William	Stirling	CEA/ESRF
Stephen	Streiffer	Argonne
James	Stubbins	U. Illinois
Jeff	Terry	Illinois Institute of Technology
Marvin	Tetenbaum	Argonne
James	Tobin	LLNL
Yulia	Trenikhina	Illinois Institute of Technology
Paul	Tumaneng	Illinois Institute of Technology
HSIAO-MING	Tung	U. Illinois
Richard	Wilson	Argonne
Eric	Wisniewski	Illinois Institute of Technology
Michael	Wojcik	Illinois Institute of Technology
Dieter	Wolf	INL
Art	Wright	Argonne
Yuming	Xiao	Argonne
Peng	Xu	U. Wisconsin
Abdellatif	Yacout	Argonne
Yong	Yan	Argonne
Yong	Yang	U. Wisconsin
Bei	Ye	U. Illinois
Hoydoo	You	Argonne
Linda	Young	Argonne
Di	Yun	U. Illinois
Andrej	Zeman	IAEA
Gary	Zeman	Argonne
Jiaming	Zhang	U. Michigan
Steven	Zinkle	ORNL
Paul	Zschack	Argonne

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