LA-UR- 09-07527

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Title:	Research Needs and Opportunities for Characterization of Activated Samples at x-ray and Neutron User Facilities
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Intended for:	Word-wide web

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Workshop Summary

Research Needs and Opportunities for Characterization of Activated Samples

at x-ray and Neutron User Facilities

Santa Fe, September, 2009

Abstract

A new generation of science-based models and high performance simulations, coupled with credible uncertainty quantification, could significantly reduce the time and cost required for nuclear plant licensing and fuel qualification. Because the complementary and necessary irradiations and characterization of activated specimens are expensive, the intelligent application of innovative experiments is important to realizing the potential savings. X-ray and neutron facilities are relevant because they provide measurement capabilities over wider time and length scales than typically are available in lab scale testing and have been fruitfully applied to wide range of materials challenges. However realizing the potential of these large-scale user facilities will require cultural changes at many facilities because of the requirements and infrastructure associated with safe handling of activated material. The provision of instrumentation and procedures necessary for efficient handling include sample preparation areas. Bringing to bear the full range of diffraction and spectroscopic techniques on smaller and smaller samples will be crucial. Particular areas of opportunity include interfaces and surface properties. Ultimately exciting scientific rewards can be realized by making measurements *in situ* in a radiation environment.

Preamble

In September, 2009, approximately fifty international researchers met for two days at a workshop entitled "Research Needs and Opportunities for Characterization of Activated Samples at X-ray and Neutron User Facilities." Attendees came from national laboratories, academia, EPRI and the NRC. The workshop was motivated by a belief that new tools at user facilities could proffer unique advances in our understanding of radiation damage but are underutilized. If they could be more widely applied their insights might accelerate our understanding and certification of materials used in nuclear applications. A subsidiary motivation for the workshopwas the desire of Los Alamos National laboratory to explore current and future facility plans that would complement its "MaRIE" signature facility concept. The discussion was informed by three Office of Science workshops: Basic Research Needs for Advanced Nuclear Energy Systems,Basic Research Needs for Materials Under Extreme Environments and Next Generation Photon Sources. Specifically this workshop focused on user facilities and addressed problems that warrant examination, current activity at user facilities, opportunities that can be realized in the next few years, insights that could be realized by new diagnostics and experimental methods, and requirements/desired attributes for new facilities of decadal scope.

Problems that warrant examination

The range of irradiated and activated samples that can benefit from examination at user facilities is considerable. Candidates run the gamut from fundamental materials science studies to fuels to structural materials all over a range of dose, rate and temperature conditions. Problems encompass immediate issues such as fuel pool liners to future issues implicit in conceptual fusion first wall materials. The capability to characterize activated samples in hot cells, while inevitably limited, is robust (at least internationally).Thus, the present workshop focused on opportunities enabled at user facilities which provide intense sources of neutron or X-rays. These can offer unique non destructive insights, often, more efficiently than lab scale characterization tools. Noting the current advocacy for greater impact of modeling to certification and discovery, the meeting focused on tools that would contribute to the perennial desire to link models over comprehensive length and temporal scales. Currently many models are not hardware limited, but algorithm limited in developing physically-validated methodologies for predicting for example microstructural evolution or tensile stress-strain behavior using mesoscale (dislocation dynamics) models. For this reason much debate focused on measurements pertinent to atomistic and molecular dynamics calculations which were considered most in need of validation.

Current activity

In-hot-cell characterization measurements typically apply the full range of lab scale techniques such as TEM, SEM Auger PAS, Raman, mechanical testing etc. to irradiated samples with the requisite remote handling infrastructure (and limitations). Often the focus is on surveillance coupons and on properties pertinent to engineering: hardness, tensile properties, toughness, residual stress, texture, yield strength, strain hardening, corrosion & oxidation rates, and materials compatibility. X-ray and neutron user facilities often provide opportunities to examine the same, and in some cases different, phenomena non-destructively on smaller samples with greater precision, spatial and temporal resolution. Unfortunately the willingness of existing facilities to accept radioactive samples is variable. For example the Diamond facility in the UK currently imposes limits which essentially preclude examination of activated samples. However, there are counter examples such as the Stanford Synchrotron Radiation Lightsource which accepts samples up to 10GBq or the Chalk River facility where shielded casks have been used to make neutron diffraction measurements on samples up to 20 000R/hour! Over the course of the workshop the logistics challenges of making measurements at neutron and synchrotron facilities were discussed. Success stories included measurements of residual stress, texture, solute clustering and void nucleation by diffraction and small angle scattering.

Near-term Opportunities Developments at 3rd and 4th generation light sources as well as at new neutron sources such as SNS or JPARC hold potential for unprecedented insights into fundamental processes that dictate radiation damage. Workshop participants identified several specific areas of opportunity: diffraction measurements during loading of archival samples; residual stress measurements of structural welds; full suite of property determination of irradiated ODS / nano structured ferritic materials; measurements on samples of atypical isotopic composition (e.g. inelastic neutron scattering on Pu 242); resonant inelastic X-ray scattering fluorescence measurements (e.g. on Americium at high pressure); and development of advanced neutron focusing optics (e.g. for studies of ion beam irradiated layers). One recurrent theme was the widespread availability of material, already irradiated under a wide range of conditions.

Opportunities implicit in the Linac Coherent Light Source, LCLS, were singled out for discussion because of its potential to observe with pico second resolution, defects, their interactions and dynamics, thereby elucidating atomistic phenomena. This may lead to better charting of damage pathways and thus to the development of improved materials. The high-intensity femtosecond photon pulses offer experimental potential that complements high-performance computing and advanced models. Realizing this potential will require the development of techniques that can use such radiation sources to measure nucleation of defect clusters, bubbles/voids, image dislocations, grain boundaries and precipitates, and demonstrate the interaction of dislocations with defect clusters, bubbles and voids.

Methods for transformational insights

Much of what we think we know about primary damage formation comes from molecular dynamics simulations. However much experimental effort misses the spatial and temporal regimes most pertinent to MD by focusing on long range or bulk average phenomena such as resistivity, temperature, swelling , constitutive response and corrosion rates. Thus new tools which can provide 3-D spatial distributions of defect and chemical distributions with atomistic resolution could prove invaluable, especially if they also had temporal resolution consistent with their dynamic evolution. Moreover, collecting a database of such information could inform the many calculations which currently predict the behavior of point defects and clusters, location of specific nucleation centers, defect formation and recombination, nucleation transformation and migration phenomena.

There is also a compelling need for engineering studies using new tools that can operate under extreme irradiation environments. For fuels, issues of specific interest include melt temperature as a function of actinide composition and chemistry; dimensional stability, thermal properties, and material diffusion as a function of chemistry, temperature and microstructure; heat generation from nuclear processes; fission product accumulation and gas release. For cladding, issues include strength and ductility as a function of microstructure, temperature, and chemistry; actinide and fission product diffusion; chemical reactions at fuel-clad interface. In both cases the potential to make measurements during simulated failures such as loss of coolant accidents would allow failure margins to be explored.

Requirements in a decadal future facility

At the end of the workshop the participants considered what capabilities might be required in a facility that could juxtapose irradiation capability with advanced probes a decade in the future. There was no immediate consensus answer to this question or indeed on the likelihood of science based certification superceding the existing cook and look paradigm. However there was consensus that complementing a new neutron irradiation facility with intense neutron and X-ray sources for characterization purposes would be valuable. The capability to irradiate under a variety of conditions (fission reactors, ion beams, spallation sources, etc.) was considered essential. Material test reactors, spallation sources and ion beams were all discussed.

Regardless of the irradiation source, desirable functionality in a decadal future facility included in situ and ex situ measurements of a range of phenomena, e.g., diffraction during loading; examination of individual grains in activated samples; handling and characterization of "large" components; in situ creep properties with helium; defect kinetics measurements with resolution than overlap with models and spent fuel characterization characterization of spent fuels. Engineering requirements will likely be dominated by need for insight, particularly under high burn up conditions, on new fuel types such as Triso, MOX, or Thoria, of which we have comparatively little experience.

Broader cross-cutting themes

During breakout discussions, three broader themes arose that merit attention in any exploration of advanced measurement capabilities. First, it was noted that the nuclear power industry does not appear to be advocating long term research. The general assumption was that short-term commercial imperatives focused industry attention on the immediacy of existing light water reactor recertifications. The participants spent some time discussing this issue with solutions ranging from more aggressive participation of the research community at

EPRI and NRC meetings; education of the user communities on the way safety case certification occurs now; definition of a DOE champion; efforts to bridge the gaps between the different modus operandi of the Office of Science and the Office of Nuclear Energy; and addressing the issue of conflict of interest and intellectual property issues.

A second theme pertained to the engagement of theorists. There was some belief that, even with new validation opportunities, the promise implicit in new generations of models would not be realized without complementary investment in what was called a "virtual computational end station". Essentially this theme advocated significant investment in developing the computational infrastructure necessary for the community to take advantage of insights that could be achieved by bringing light and neutron source characterizations to bear on activated materials.

Finally, a philosophical theme arose concerning the question of whether emerging nano/microscale testing can provide relevant certification data. This question is similar to that of whether science based models, perhaps selectively validated under non representative conditions, can make substantial inroads towards certification, when compared to long term irradiation under representative conditions.

Conclusion

If the promise of the so-called nuclear renaissance is to be realized, especially in the United States, the breadth and depth of the nuclear science and engineering community must be enhanced substantially. In particular there is a need to revitalize the materials science of radiation damage. It was clear to workshop participants that x-ray and neutron sources at national user facilities have an important role to play in this endeavor. Further, in addition to cultural changes to allow the full exploitation of currently available tools and techniques, new capabilities need to be developed if science-based certification is to play a role in the resurgence of nuclear energy. Finally, given the magnitude and urgency of the need for carbon-neutral energy, approaches must be found to reduce the time and cost associated with licensing and certification.