National Facility for Imaging Nuclear Materials and Processes (INMaP)

Nigel D. Browning – Pacific Northwest National Laboratory

Summary

The goal of this proposal is to establish a national user facility to image radiation damage effects in materials on the fundamental atomic scale with the temporal resolution to image damage processes as they happen. This facility will allow direct observation of the critical materials damage events that occur in all nuclear energy options currently being considered, thereby providing unique insights into the design of materials and processes to accelerate the implementation of these new technologies (through both direct analysis and by providing benchmarks for the extensive simulations being used for nuclear energy). This level of structural analysis will be achieved by using recently developed state-of-the-art characterization tools (an ultrafast transmission electron microscope (UTEM), an aberration corrected and monochromated TEM and a compact X-ray light source (CXLS)) that are synergistically linked to multiple beam ion accelerators – providing direct multi-modal analyses of the atomic scale events occurring during the key damage cascades. These capabilities will build on the infrastructure at PNNL in nuclear materials and technologies, enhance existing and establish new partnerships with nuclear research programs in the US, and significantly leverage investments made in the laboratory Chemical Imaging Initiative (CII) to achieve success.

Introduction

The development of nuclear power systems for the immediate future (the next 10-20 years) presents severe challenges for materials scientists and engineers. This is in addition to the challenges facing the current nuclear industry that continue to evolve as new, unforeseen problems arise. As reactors designs are explored that push the limits of existing materials, it becomes increasingly important to develop new materials and processes to extend the operating envelop to higher temperatures and larger neutron fluences. This endeavor is hindered by two challenges unique to nuclear materials development. First, from start to finish neutron-irradiation experiments are difficult, expensive ($1-10M per experiment), and take years to complete in even the best of circumstances, and often produce highly radioactive materials that require expensive facilities to handle, store and characterize the materials. Secondly, and no less important, neutron irradiation produces continuous atomic scale structural damage events that take place in picoseconds, but then evolve over multiple temporal scales that include years of reactor operation, and even the millennia if one considers spent fuel storage where decay events continue to produce radiation damage. The radiation damage issues for fusion reactors are even more severe than fission reactors, and a viable fusion neutron source for materials irradiation experiments does not exist nor will it exist in the foreseeable future. To overcome these issues of extreme conditions and widely varying timescales, scientists have turned to computational methods to simulate the structural response to the radiation and temperature environment of a reactor. However, as has been found from other areas of materials science where experiments are easier to perform, without a benchmark set of measurements that can both validate existing simulations and provide unique insights for new materials and processes that can then be simulated, the advances that can be made are severely limited. Therefore, a critical national/global need exists to develop research facilities that will address the materials science and engineering issues associated with the future use of nuclear power in a more cost conscious and time sensitive manner. As the basic principle behind all materials development is a fundamental understanding of the structure-property relationships, this means that **state-of-the-art characterization facilities for nuclear materials are needed, one that can take advantage of highly activated neutron-irradiated materials.**

Characterization of radiation damage using in-situ transmission electron microscopes (TEM) coupled with ion accelerators has progressed through efforts at Argonne National Laboratory (ANL) in the US and at a few other institutions worldwide. However, the microscopes being used for this type of structural characterization are far from the current state-of-the-art (and the facility at ANL is scheduled to close permanently). Other than the highly visible structural characterization effort at ANL, much of the nuclear research in the US is in the National Laboratory system and is inextricably linked with the energy security, non-proliferation and stockpile stewardship missions of the Department of Energy. As these missions clearly have national security implications, the research is performed primarily behind a fence and the results are understandably not shared extensively with the scientific community. Therefore, although some basic characterization equipment does exist, it is far from ideal and very difficult to access. Conversely, modern instrumentation within the Basic Energy Sciences (BES) user facilities represents some of the most advanced characterization methods for materials science available anywhere in the world, but because of the contamination/certification issues for characterizing nuclear materials, very little nuclear materials characterization is performed in these facilities (which is understandable given the wide scientific research goals of these facilities). Hence, although the imaging/analysis technology exists, it is not being fully implemented to study nuclear materials. **For the rapid advances needed to realize next generation nuclear power, it is essential that a dedicated open access state-of-the-art facility be established to provide characterization for all areas of nuclear materials science.**

The imaging facility described here aims to distill the imaging and analysis technologies from within the multi-billion dollar Office of Science user facilities and to implement them (along with key recent advances) to the analysis of nuclear materials. Here the goal is to focus on creating and analyzing the critical damage phenomena that limits implementation of new nuclear facilities on the fundamental length and time scales. By providing a mechanism for experimental measurements not possible at other user facilities, this new National Facility for Imaging Nuclear Materials and Processes (INMaP) can rapidly accelerate the implementation of new reactor technologies.

Scientific Drivers

The DOE workshop on advanced nuclear energy systems held in July 2006 brought together 235 experts from 31 universities, 11 national laboratories, 6 industries and 3 government agencies with scientists from 11 different countries in attendance. This workshop defined nine priority research directions, of which six can be addressed either wholly or in part by the proposed imaging facility to study radioactive materials:

- Nanoscale Design of Materials and Interfaces that Radically Extend Performance Limits in Extreme Radiation Environments
- Microstructure and Property Stability under Extreme Conditions
- Fundamental Effects of Radiation and Radiolysis in Chemical Processes
- Exploiting Organization to Achieve Selectivity at Multiple Length Scales
- Predictive Multi-Scale Modeling of Materials and Chemical Phenomena in Multi-Component Systems under Extreme Conditions
- Physics and Chemistry of Actinide-Bearing Materials and the f-electron challenge

Each of these priority areas are associated with several aspects of a complete nuclear power plant – such as the basic physics and chemistry of the fissionable materials, the properties of the structural materials used to construct the reactor, the efficiency of the fuels used to power it, and the properties of the waste developed during its operation (which determine the environmental issues related to long-term storage). In terms of a research program that utilizes advanced imaging facilities to solve the particular materials challenges associated with nuclear power, it is easier to break the priority research areas described above into these four main areas of research; structural materials and cladding, fuels, waste immobilization and validation of theoretical models. Within these areas there are significant challenges that can be addressed by a dedicated characterization facility:

- The fate and transport of transmutation and implanted He in fusion and fission reactor structural materials is an issue of paramount concern. Imaging of He segregation at grain boundaries and interaction with other lattice defects during irradiation would make it possible to validate a tremendous amount of computational research.
- Use in-situ ion irradiation to study the details of defects, interfaces and phase evolution for different starting microstructural states, for example, follow the evolution of microstructure of unirradiated versus previously neutron-irradiated samples.

- The design of advanced nanoscale structural materials for radiation damage mitigation would benefit from imaging of defect localization at engineered nanoscale sinks and structural evolution of the sinks under irradiation. These materials include advanced ODS alloys, nanofeatured ferritic alloys, and nanolayered materials.
- Fundamentals of damage cascades in materials, including cascade morphology, cascade overlap, and retained post-cascade damage would benefit from UTEM images of cascade defect production, defect cluster formation, and initial stages of transport.
- The fate and transport of fission gases in nuclear fuels is a critical issue for fuels performance models and direct imaging of fission gas bubbles and related transport would facilitate the design of new, accident-tolerant fuels, as well as validate a number of fuel models.
- Fundamentals of environmental effects in nuclear reactor systems, including stress corrosion and environmental embrittlement, can be addressed in this facility by direct imaging of materials oxidation at high resolution and under irradiation. Additionally, in situ straining and high-resolution imaging can elucidate radiation hardening mechanisms, including dislocation channeling.

Instrumentation in the Facility

There are 5 main components to the proposed system: a specimen preparation section that features a shielded glove box linked to specimen preparation facilities and dedicated FIBs (total cost ~$20M), a multiple beam ion accelerator (total cost ~$15M) a double corrected cold field emission 300kV TEM/STEM with electron energy loss spectroscopy (EELS) and energy dispersive X-ray (EDS) capabilities (total cost ~$15M), an ultrafast TEM capable of ~picosecond temporal resolution ($25M), and a compact X-ray light source ($40M). This project is really an integration project rather than a construction project. In this section, therefore, a brief description of the capabilities of the components will be described – with the final system having the sum of the individual capabilities. The only part of the project where there is more design work is the shielded glove box/specimen prep facilities to deal with potentially highly radioactive samples.

The aberration corrected transmission electron microscope will be a high-tilt JEOL 80-300kV cold field emission double aberration corrected TEM/STEM (i.e. aberration corrected STEM and aberration corrected TEM on the same instrument). The microscope has a 0.3eV energy spread in the source which when coupled with the Quantum Gatan Imaging Filter (GIF) will allow high resolution EELS and energy filtered imaging to be performed. The microscope will provide 0.07nm imaging resolution in both bright field (phase contrast) and dark field (Z-contrast) STEM and TEM point to point resolution will be less than 0.1nm. For EDS the instrument will operate with high peak to background for a probe size of 1nm with 5nA beam current and the software will feature full mapping capabilities. The microscope voltage is variable between 80 and 300kV, allowing variability in the energy and dose of the electrons to be modified for each experiment. This microscope represents the highest resolution materials science instrument available and will have the potential to study nuclear materials and processes at unprecedented levels of spatial resolution. The ultrafast transmission electron microscope will be based on the microscope described above, but feature an optimized photo-emission source to allow a range of temporal resolution to be obtained (from microseconds to picoseconds). In addition to the modification of the source for the microscope, the instrument will feature an optimized set of temporal lenses (pulse compressors) to manipulate the beam to the critical temporal regime for the particular damage effect.

The compact X-ray light source (CXLS) will be based on ICS, where a high brightness electron beam collides with a high intensity laser pulse and upconverts the photons to X-rays. The starting point of the CXLS will involve a superconducting radio frequency (SRF) photo-injector to produce the high-brightness, high average current beam that is coupled to an SRF linac that accelerates the beam to tens of MeV. The X-rays are then created by the interaction of the electron beam with a high average power cryo-cooled infrared laser operating at ~1kW average power that is coupled into a coherent enhancement cavity to build up ~1MW of stored optical power. The flux that is produced in this way is ~1 micron in size with a relatively large divergence, which makes it ideal for coherent imaging. By coupling the optics under the right conditions, the ion beam (see next paragraph) can be used to stimulate pump-probe experiments over large areas with ~ picosecond resolution (making the CXLS comparable to UTEM in temporal resolution but covering a larger sample size). Together with the electron microscopes the CXLS will provide a complete set of imaging, diffraction and spectroscopic tools to study the progression of damage in radiation materials on its critical timescale.

The accelerators that will be linked to the microscopes and compact X-ray light source described above will enter the column at 30^o angle to ensure the maximum overlap with the electron beam. The two accelerators will be (or will be similar in performance to) the NEC Pelletron model 6SDH-2, which is designed for helium ion production and the NEC Pelletron model 5SDH-4 which is designed for heavy ion production. Included with the accelerators are the optical components necessary to bring the two ion beams into the column in the same guide tube. Both accelerators are designed for high current operation (i.e. implanting thick samples) making them easily capable of providing the necessary current for the in-situ TEM. This current will be measured for calibration purposes by a Faraday cup that can be inserted into the microscope/CXLS beam lines.

A glovebox will be utilized in order to protect researchers from airborne radiological contamination while performing sample preparation procedures on the radioactive specimens that will be examined in this facility. The primary features of the glovebox include a fully enclosed chamber with windows and gloves to retain airborne contamination, recirculating inert and HEPA filtered atmosphere for working with materials that are reactive to air, e.g. U and Pu; an airlock that will receive the tip end of a TEM and/or FIB specimen holder for transferring samples from the glovebox to the FIB/TEM/CXLS under sealed vacuum conditions and a fume hood and antechamber attachment for the safe transfer of materials and waste into and out of the glovebox. The glove box will be long and narrow in shape and will house all of the sample preparation instruments and supplies. The main issue for the proposed facility is the widely varying activities and radiation types for the elements that will be studied. For example, in the case of primary alpha emitters such as Pu239, thick rubber gloves, Plexiglas and thin stainless steel shielding around the glove box is sufficient to work safely. However, for beta emitters, such as Am241 lead shielding is needed. In the case of spent nuclear fuels there are some highly radioactive components with short half-lives that cannot be handled at all by a glovebox (these samples will require a cooling period and will be handled using facilities within PNNL Radiochemistry Laboratory and at collaborating institutions). The glovebox that is planned will have lead lined gloves, leaded glass and lead shielding that will allow the specimen preparation of radioactive samples at a level not possible in other academic institutions. All activities in the glovebox will be performed by the trained radiological workers at the McClellan facility and will follow the appropriate international guidelines for such systems and will be monitored and certified by the health physicists at UC-Davis. Once the thin samples have been prepared in the glovebox/FIB, the shielding in the vacuum transfer stage is sufficient and the loading into the microscope can be performed by the microscopy specialist operating the system.

Budget

The total funds required to establish the imaging facility described above is expected to be $105M, and require $15-20M per year to operate as a national user facility once established. The cost of the proposed instrumentation is based on a set of competitive quotes that have been obtained for the major components (Aberration corrected TEM, ultrafast TEM glove box and specimen preparation including FIB, CXLS and ion-accelerator). As the vast majority of the components used in the construction of the full system are based on tried and tested technologies (the UTEM and CXLS require minor upgrades that have already been defined), there is little risk associated with the construction, and the project is probably more accurately termed as an assembly or integration project. The only part of the project that is challenging is the integration of the ion accelerator into the various characterization tools. However, the risk in this process is not something that will result in additional costs – this is factored into the listed quotes for the components. It is conceivable that there may be a time delay in the installation (as is normal to encounter on any major installation), but experience with these systems suggests that a five year time period should be more than sufficient to move the facility from the design phase, through installation and into operation.