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Accelerated Nuclear Energy Materials Development with Multiple Ion Beams

M. J. Fluss, G. Bench

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Accelerated Nuclear Energy Materials Development with Multiple Ion Beams: *On the Path to a hybrid*

Michael Fluss and Graham Bench
(Lawrence Livermore National Laboratory)

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Background

A fundamental issue in nuclear energy, fission, fusion, and fusion-fission hybrids, is the changes in material properties as a consequence of time, temperature, and neutron fluence. Usually, candidate materials for nuclear energy applications are tested in nuclear reactors to understand and model the changes that arise from a combination of atomic displacements, helium and hydrogen production, and other nuclear transmutations (e.g. fission and the production of fission products). Experiments may be carried out under neutron irradiation conditions in existing nuclear materials test reactors (at rates of 10 to 20 displacements per atom (DPA) per year or burn-up rates of a few percent per year for fertile fuels), but such an approach takes much too long for many high neutron fluence scenarios (300 DPA for example) expected in reactors of the next generation. Indeed it is reasonable to say that there are no neutron sources available today to accomplish sufficiently rapid accelerated aging let alone also provide the temperature and spectral characteristics of future fast spectrum nuclear energy systems (fusion and fission both). Consequently, materials research and development progress continues to be severely limited by this bottleneck.

The Role of Ion-Beams

Alternatively, irradiation with ions can be utilized to initiate the processes of radiation damage similar to that expected in nuclear energy systems of the next generation, but at much higher rates ($\sim 10^3$). In particular, ion-beam irradiation brings several advantageous characteristics to the problem of understanding radiation damage in materials, the evolution of fertile and fissile fuels, and performing critical scientific studies on reasonable time scales. While it is generally accepted that ion beam irradiation of materials can be utilized to simulate reactor irradiation with neutrons, [*Averback et al., 1978, Averback, 1949; Ullmaier,* 1984] the simulation of the simultaneous production of helium through (n, α) reactions in concert with displacement damage from ion recoils only became important when it was finally understood that helium plays a significant role in determining the incubation period for void swelling through its ability to thermodynamically stabilize small vacancy clusters [*Lévy, et al., 1985.*]. More recently it has been realized that the harder neutron spectra of fast reactors and fusion sources introduces the additional issue of hydrogen production through (n, p) reactions. Two countries involved in developing Generation IV (GEN IV) reactors and fusion energy systems have implemented "triple-beam" radiation facilities to simulate the synergies of ion displacements in the presence of hydrogen and helium production as well as other transmutants. Japan has a facility called TIARA (Takasaki Ion Accelerators for Advanced Radiation Applications) and France is completing the JANNuS (Joint Accelerators for Nano-science and Nuclear Simulation) triple beam system at CEA-Saclay in late 2009.

These facilities, with simultaneous tri-beam ion irradiations, are/will be able to mimic critical parameters of the aging of materials, corresponding to reactor irradiations of decades, in a matter of days and will be at the forefront of efforts to investigate the physics of materials developed specifically for ultra high radiation tolerance. Such facilities will be a cost effective and time effective platform for understanding ultra high DPA responses of materials, and in this role will stimulate the search and development of new classes of nano-phase, interfacial, and micro-structurally engineered materials that exhibit radiation tolerance.

The US role in Nuclear Energy Materials Research and Development with Ion-Beams

On April 6–7, 2009, members of the U.S. scientific community interested in irradiation effects on materials gathered at a workshop, "Workshop for Science Applications of a Triple Beam Capability for Advanced Nuclear Energy Materials" held at Lawrence Livermore National Laboratory. A repot of this workshop is available on-line (*M. Fluss and W. King*, *LLNL 2009*, ***http://tinyurl.com/triplebeamworkshop***). Their purpose was to discuss the need for a U.S. triple ion-beam irradiation capability to advance the science for nuclear energy materials. The motivation for developing such a capability lies in the complex, highly non-equilibrated situations that are observed in the range of high-radiation environments associated with advanced nuclear energy systems, including fission, fusion, and nuclear waste environments. In these environments, materials not only undergo radiation damage but also experience changes in their chemistry as the concentration of hydrogen, helium, fission products, and of other nuclear processes increases.

Ion beams are of great use in experimentally simulating complex, high-radiation environments. Among their many advantages is that they greatly accelerate the damage process compared to available neutron sources or the self-radiation damage in nuclear waste materials, while not usually producing additional radioactivity. This enables scientists to effectively use ion beams to understand the fundamental radiation effects mechanisms of complex nuclear system environments. However, there is evidence that *sequentially* irradiating materials with heavy ions (to create radiation damage), protons (to inject hydrogen), and He- does not produce conditions that behave linearly when damage and the build-up of H and He occur *simultaneously.* The occurrence of possible non-linear synergistic effects has prompted scientists in France, Japan, and the Ukraine to develop the capability to simultaneously irradiate materials with two or three ion beams: heavy ions, and one or two other energetic beams representing the transmutants.

There were four major outcomes of the workshop:

1. Participants agreed that a triple ion-beam facility would play an important role in training the scientific leaders of the future. Additionally, sample space in neutron sources is quite precious and can be difficult to obtain. Irradiations can take months or years to reach desired doses. Finally, samples are radioactive when removed from the neutron source and may need to cool for months before experiments can be carried out. In contrast, ion irradiation has the advantage of high displacement and simulated "transmutation" rates with little to no activation. Experiments can be carried out very efficiently and samples can be characterized in situ or immediately after irradiation.
2. Participants clearly stated the important role that ion beams play in advancing our fundamental knowledge of irradiation effects. Ion beams are invaluable because they provide a versatile and flexible research platform for investigating the fundamental mechanism underpinning radiation effects in nuclear energy systems. They provide control of variables that will allow investigation of unit mechanisms. Hence, new and important basic research would be catalyzed by the existence of a U.S. triple ion-beam facility.
3. The community is keenly interested in experiments where mechanisms can be probed in

real time, in situ. This included a strong endorsement of the capability for in-situ, multiple-ion-beam irradiation in an electron microscope, as well as the usual ion-beam techniques(channeling, PIXE, ERDA, NRA, RBS, etc.).

What LLNL Proposes: ***Upgrade of the LLNL Center for Accelerator Mass Spectrometry***

LLNL is well positioned to develop and apply unique capabilities in materials and solid-state theory, simulation and modeling, coupled with ion-beam accelerators and a full spectrum of advanced materials characterization instruments to enable fundamental materials research for the range of fusion and fission nuclear energy systems. One important capability is embodied in the development of a high energy (MeV) ion implantation facility at the multi-user Center for Accelerator Mass Spectrometry (CAMS). The center's main instrument is a 10 MeV FN tandem accelerator that is connected to four ion sources. As the ion sources are external to the tandem accelerator they enable CAMS to pursue both "routine user" and "state of the art" ion beam analysis applications. The accelerator and associated ion sources can produce ion beams with a Z of 1 through 94. Beam energies from the 10 MeV Tandem are sufficiently high that heavy ion implants to a depth of 5 to 10 micron can readily be achieved (e.g., 50 MeV to 90 MeV Fe ions) - sufficient to observe hydrogen and helium distribution over the depth of a grain size in high-tech steels rather than near surface implantations obtained with heavy ion beam energies of a few MeV or less.

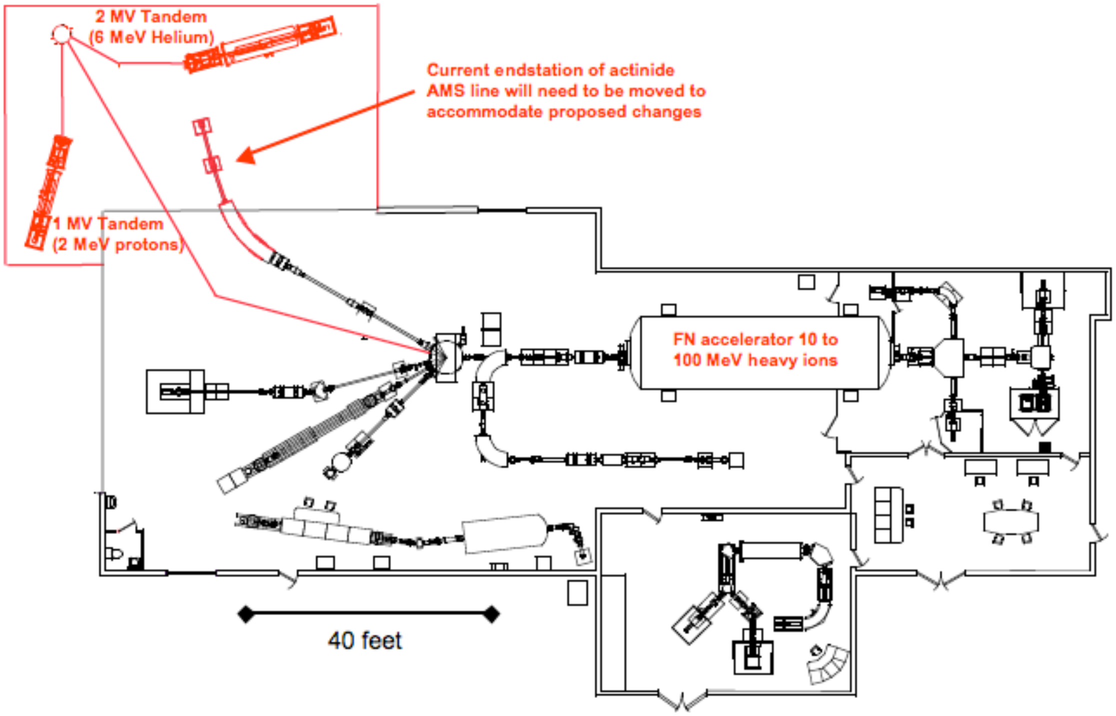

Figure 1: Conceptual changes to CAMS accelerator facility for a multi beam implantation capability. Building expansion, triple ion beam accelerators and beam lines and modifications to existing infrastructure are outlined in red

We propose to build a simultaneous triple ion beam capability centered on the 10 MeV FN tandem accelerator. For a simultaneous triple ion beam capability we will need hydrogen and helium ion beams of a few MeV energy, beam currents up to a few hundred nanoamperes, and heavy ion beams of energy 10 to 100 MeV with particle currents up to a few microamperes. Consequently, we will:

1. Use the current 10 MeV FN tandem to provide heavy ion beams up 100 MeV,
2. Procure a 1 MeV accelerator to provide up to 2 MeV protons, and
3. Procure a 2 MeV accelerator to provide up to 6 MeV alpha particles.
4. Design and construct a specialized environmental chamber for triple-beam irradiations

The existing accelerator building will have to be expanded by ~2000 square feet (Figure 1) for installation of the triple beam facility. A facility growth plan will also be developed that will likely include increased access with the purchase of an ECR source 3.5 MeV accelerator for heavy ion irradiation, and plans for a multi-ion-beam TEM integrated station. With institutional priority that "fast tracks" the building addition we estimate the cost and time of $20M and a timeline between 18 to 36 months. A preliminary spending and build out time schedule is schematically represented in Figure 2 below.

Operations and Accessibility

Once complete it is envisaged the triple ion beam capability will utilize 25% to 50% of the FN accelerator beam-time to investigate the materials physics of nuclear energy materials developed specifically for ultra high radiation tolerance or deep burn. Concurrent with this capability, improvements in theory, simulation, and modeling to develop techniques to model advanced materials at rates and time scales consistent with both accelerated aging and with reactor environments are being undertaken.

We also anticipate creating the required framework for support of the triple beam facility as a National (and International) User Facility. We will initiate an education and outreach effort to obtain interest and commitments from other National Laboratories, Universities, and Industry. Such a facility will provide opportunities to train and support leading scientists of the future.

CAMS has an outstanding record of outreach and currently has over 100 university, DOE and private sector collaborations and has hosted over 1,000 faculty & student visitors in the past decade, resulting in more than 125 Masters and Ph.D. theses. These interactions have led to outstanding broad scientific impact with CAMS scientists authoring 18 and 13% of LLNL's *Science* and *Nature* articles, respectively, over the past decade and CAMS research featured on covers of 10 journals in the last 5 years. We propose a structure that will take advantage of and build upon the history of successful collaborative research and development between CAMS and the outside community. Our scientific goal is to facilitate, formalize, and substantially expand collaborations based around the CAMS facility that will take advantage of high energy ion implantation capabilities for development of new classes of nano-phase, interfacial, and micro-structurally engineered materials that exhibit radiation tolerance.

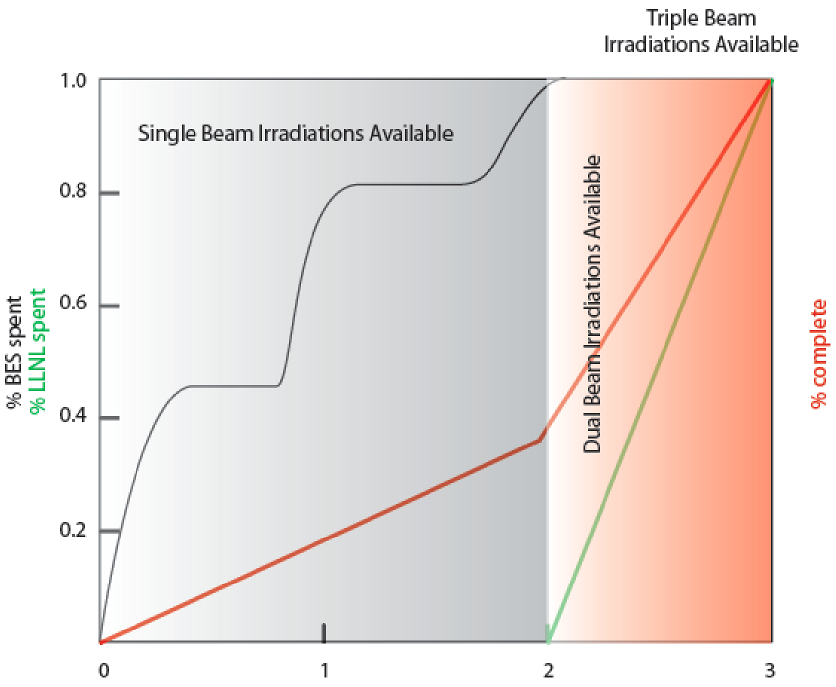

Figure 2: Preliminary spending and build out plan for upgrading the CAMS FN Tandem to a Triple Beam Ion-Beam Facility.

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