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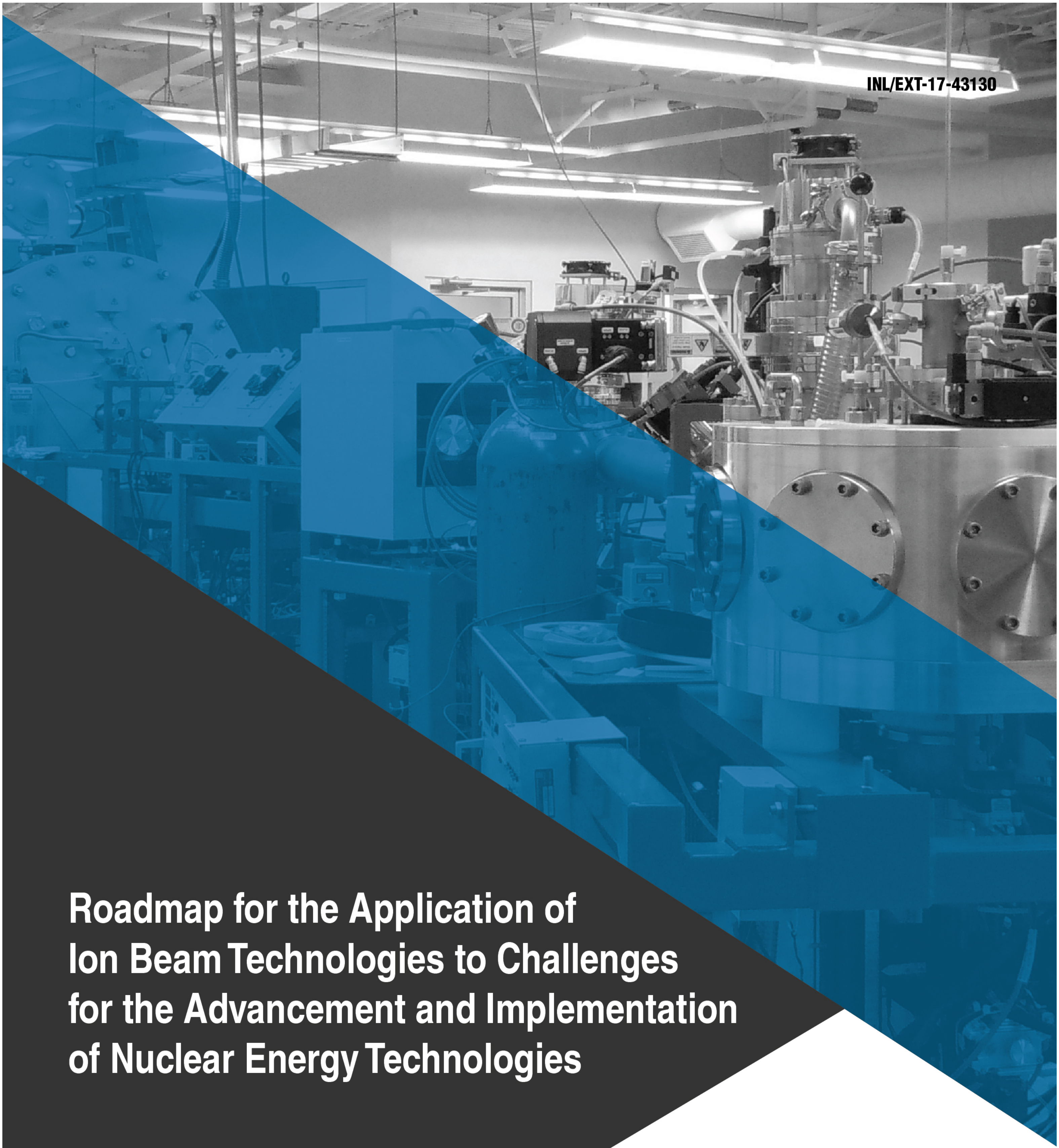

Roadmap for the Application of Ion Beam Technologies to Challenges for the Advancement and Implementation of Nuclear Energy Technologies

NSUF Nuclear Science User Facilities

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Prepared for the

U.S. Department of Energy
Office of Nuclear Energy
Under DOE Idaho Operations
Office
Contract DE-AC07-05ID14517

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Executive Summary

The use of ion beams in interrogating materials performance for nuclear energy applications is advancing at a rapid rate. Ion irradiations have been proven to produce radiation effects data that are of direct relevance for understanding neutron-induced displacement damage. This development reflects significant and continued investment in infrastructure by the U.S. Department of Energy (DOE) as well as support of science and engineering research at universities and national laboratories. Application of ion beam irradiation methods to enhance the deployment of materials in reactors is in the spirit of the Gateway for Accelerated Innovation in Nuclear (GAIN) initiative sponsored by DOE to accelerate the deployment of innovation in the U.S. nuclear industry.

Ion beam irradiation shows considerable promise for assisting in down selecting candidate materials for use in both current and advanced nuclear energy systems. Ion beams allow rapid achievement of materials damage levels not accessible by neutron irradiation in test reactors due to cost and time constraints. Ion beam studies provide data on the effects of irradiation under very specific conditions of temperature, radiation dose, and radiation dose rate that are difficult or impossible to achieve via reactor irradiation experiments. This data can be used to develop and validate detailed predictive radiation effects models for use in reactor materials research and development.

Despite the current high state of the art and the maturity of quality assurance, there are still challenges to the deployment of ion beam data in support of reactor materials qualification. For the most part, these challenges arise from the lack of a detailed mechanistic understanding of potential differences between ion induced and neutron induced materials damage, which is exacerbated by difficulties in obtaining bulk physical and mechanical properties data from the micron-scale heavy ion irradiation depths. In particular, improved understanding of dose rate effects, as a function of temperature, are needed.

This document offers a roadmap for the development and enhancement of current U.S. ion beam irradiation technologies within university and national laboratory settings, and especially for the deployment of new highly controlled in situ interrogation of materials during irradiation to provide dynamic and mechanistic data for model development. The status and capabilities of relevant U.S. ion beam facilities are summarized and recommended “best practices” for performing ion irradiations are described. The potential role of ion beam irradiations to assist the development and deployment of reactor materials is outlined. Key principles include developing methods for rapid and cost-effective materials selection and development, characterizing fundamental material response under irradiation, and developing a robust mechanistic understanding of microstructure evolution under irradiation (including development and validation of reliable predictive models for microstructure evolution).

Ion beams are anticipated to provide an increasingly important role in the down selection of candidate materials, providing crucial data for the development of validated radiation effects models, and allowing confirmatory reactor irradiation tests to focus on a subset of key irradiation conditions. Ion irradiations have proven highly valuable for understanding complex phenomena such as radiation induced solute segregation, irradiated microstructure evolution, irradiation hardening, and irradiation assisted stress corrosion crack initiation, thereby providing important predictive information on likelihood of failure of key reactor components and the potential impact of operational changes such as power uprates. Several challenges that currently inhibit full implementation of ion beam research to accelerate the development and qualification of nuclear reactor materials are discussed. The challenges represent potentially valuable near term research and development focus areas.

List of Acronyms

ANL	Argonne National Laboratory
BNL	Brookhaven National Laboratory
DOE	U.S. Department of Energy
DOE-NE	U.S. Department of Energy Office of Nuclear Energy
EPRI	Electric Power Research Institute
GAIN	Gateway for Accelerated Innovation in Nuclear
HBS	high burn up structure
IASCC	irradiation assisted stress corrosion crack
IBIOW	Ion Beam Investment Options Workshop
IBIR	Ion Beam Irradiation Roadmap
INL	Idaho National Laboratory
IVEM-TUF	Intermediate Voltage Electron Microscope–Tandem User Facility
LANL	Los Alamos National Laboratory
LWR	light water reactor
NE-5	Office of the Deputy Assistant Secretary for Nuclear Technology Demonstration and Deployment
NEAMS	Nuclear Energy Advanced Modeling and Simulation
NEC	National Electrostatics Corp.
NRC	Nuclear Regulatory Commission
NSLS-II	National Synchrotron Light Source-II
NSUF	Nuclear Science User Facilities
RD&D	research, development, and demonstration
SNICS	source of negative ions by cesium sputtering
SRIM	Stopping and Range of Ions in Matter
TAMU	Texas A&M University
TEM	transmission electron microscopy

1 Introduction

Accelerator-based ion irradiation has been proven to produce radiation effects data that are directly relevant for interpreting neutron-induced damage. Such irradiation experiments, although conducted with dpa (displacements per atom – one dpa means that on average, every atom in the lattice is displaced one time) rates with orders of magnitudes higher than the neutron damage rates in a reactor, offer the most credible method for materials screening (i.e., comparing the relative response of individual alloys, even though a one-to-one correlation between ion irradiation and neutron irradiation in a reactor has not yet been provided).

The purpose of this roadmap document is to:

1. summarize the current status and state-of-art techniques employed in ion simulation of neutron-induced damage,
2. develop "best practice" recommendations towards standardization of testing procedures,
3. identify issues and challenges for emulation of neutron damage,
4. propose techniques to overcome technological bottlenecks in the near future, and
5. propose long-term strategic plans for research and development needs.

Ultimately, these goals will provide (i) guidance for research groups in their efforts to mesh fundamental nuclear energy science and engineering studies with nuclear industry needs, and (ii) the will to speed up the acceptance of accelerator testing data in nuclear reactor regulation, design, and licensing processes.

1.1 Role of Ion Beam Systems in Addressing the Materials Challenges in Nuclear Energy

Developing and supporting methods for accelerated materials qualification with respect to licensing current and advanced reactor technologies

It is well known that the time for innovation to have impact in the nuclear industry is too long. The rapid development and uptake of ideas is hampered by many challenges, especially the difficulty of accessing and the high cost of using nuclear research, development, and demonstration (RD&D) facilities. In addition, regulatory processes are complex and difficult to overcome for small-scale innovators and entrepreneurs.

To meet U.S. energy security and climate protection goals, it is now recognized that the U.S. Department of Energy (DOE) needs to accelerate the commercialization of innovation in the nuclear industry by providing both a single point of access to the DOE research complex for innovators and investors, and focused research opportunities, as well as working with the Nuclear Regulatory Commission (NRC) to facilitate communication.

DOE has launched the Gateway for Accelerated Innovation in Nuclear (GAIN) initiative to meet this objective.[1] GAIN is a public-private partnership that is dedicated to increasing uptake of new and novel ideas and to accelerating deployment of innovative nuclear technologies. It will focus "relevant, federally-

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[1] http://gain.inl.gov, accessed May 21, 2017.

funded nuclear energy RD&D programs" to provide access to technical, regulatory, and financial support to accelerate innovation to commercial deployment in the nuclear industry in a cost-effective way.

The GAIN initiative has five themes:

1. Modeling and simulation
2. Cross-cutting design support
3. NRC interface
4. Base reactor and fuel cycle RD&D programs
5. Experimentation.

Provision of world-leading, state of the art capabilities to provide innovation in materials development, selection, and validation for their use in (potentially extreme) irradiation environments at reactor operating conditions of high temperature and pressure is a central component of the last of these five areas. Ion beam irradiation offers the opportunity to interrogate the performance of materials under very specific and defined conditions. Furthermore, ion beam technologies allow the measurement of the deleterious effects of radiation on an accelerated timescale compared to neutron irradiation, allowing significantly faster testing and a more rapid qualification judgement.

For conventional materials used in existing nuclear power plant and fuels, ion beam experiments provide data allowing a detail mechanistic understanding of the damage and corrosion processes leading to material deterioration and failure. This information, along with the output from the associated models, will enable better predictions of performance and integrity, and therefore, allow potential lifetime extension due to overly conservative assumptions made during construction and licensing.

For advanced, new, and novel materials, ion beam studies will enable the down-selection of candidate materials based on actual performance under irradiation and will give detailed data for the development of models and simulation techniques that may ultimately be used for design and qualification of materials.

Development of a deeper understanding of factors that control the evolution of microstructure and properties under irradiation

Unlike reactor environments in which temperature, neutron flux, and sometimes stress are correlated with each other and frequently experience large fluctuations throughout the reactor operational history, ion irradiation can be performed under well-controlled and relatively invariant conditions. Major irradiation parameters can be separated from each other for studying the dependence of damage phenomena on single variables. Furthermore, ion irradiations can be extended to conditions beyond typical reactor operation conditions (e.g., dpa levels >200).

The effect of temperature, dpa level, and dpa rate are major factors controlling the structural evolution of materials, with stress playing a secondary role. The use of the displaced atom concept has been widely accepted as a damage equivalency parameter to convert neutron damage to ion damage, but dpa equivalency in itself is insufficient: it does not consider processes that arise from the very large differences in dpa rate between the two types of irradiation. Other important factors include primary knock-on atom energy spectra (light ions vs. heavy ions), effects of electronic ionization on defect development (electronic stopping vs. nuclear stopping), electron-phonon coupling (heating of electron subsystem and its conversion to local heating), damage cascade overlapping (non-linear displacement creation in damage overlapped regions), and Coulomb explosion (lattice atom displacement in electron depleted zones under extremely high electron stopping). Additionally, the effect of phase evolution may also be sensitive to time and/or displacement rate. The effect of phase evolution often precedes the development of irradiation creep and void swelling, thereby influencing the overall radiation response.

Development and validation of fuels and materials models

Multiscale modeling conducted via a combination of *ab initio* first principle calculations, molecular dynamics simulations, Monte Carlo simulations, kinetic Monte Carlo simulations, rate theory, phase field theory, and Monte Carlo-based defect cluster dynamics simulations has been actively pursued in the radiation damage community. This approach seeks to link atomic scale details to mesoscale behavior that is linkable to experimental observation, but the gap between modeling and experimental observation is still large and very visible. Issues involving computation costs, integration of various codes over varied length and time scales, accuracy/uncertainty, and data communication are still to be solved.

For both fuel cladding materials and fuels, ion irradiation experiments can contribute to modeling validation and verification under precisely controlled irradiation conditions. However, due to the limitation of ion penetration depths it is difficult to simulate fuel reconstructing since rim structural formation, fuel densification, and fuel grain growth occur at much larger length scales. Ion irradiation is valuable to simulate localized structural changes such as fission gas bubble formation. Through a combination of ion implantation techniques, it is also feasible to study fundamental fission product transport behaviors. For fuel cladding materials, microstructural evolution is dominated by defect clusters of typical sizes up to a few nanometers, which can be well simulated by ion irradiation. Ion irradiation to study radiation effects on surface oxidation and surface cracking, processes that typically occur over much larger length and time scales, is more problematic.

1.2 Development of Ion Beam Systems as a Tool

2016 Ion Investment Options Beam Workshop Summary

The Nuclear Science User Facilities (NSUF) Ion Beam Investment Options Workshop (IBIOW) was held to develop a set of recommendations (i.e., a priority list) for supporting domestic ion beam irradiation capabilities available to researchers. These capabilities are focused on the support of nuclear energy research, development, and deployment. The recommendations are intended for use by the U.S. Department of Energy Office of Nuclear Energy (DOE-NE) when faced with funding decisions about investments in ion beam support, instruments, and facilities. Recommendations developed during the IBIOW are provided in the *Supplement to the NSUF Ion Beam Investment Options Report: Initial Results and Recommendations* [1].

As part of their initial discussions of potential future funding scenarios, IBIOW participants considered input submitted through DOE-NE Request for Information DE-SOL-0008318, "University, National Laboratory, Industry and International Input on Potential Office of Nuclear Energy Infrastructure Investments (April 13, 2015)." Discussions and presentations of other input, whether specific or general in scope, were also welcomed. Also included was user input, including input regarding DOE-NE program interests and ion irradiation research, development, and deployment needs. The workshop was held

March 22–24, 2016, at the Idaho National Laboratory (INL) Meeting Center in the Energy Innovation Laboratory in Idaho Falls, Idaho.

Workshop participants were selected from various sources (i.e., request for information respondents, Nuclear Energy University Program/Nuclear Energy Enabling Technology infrastructure applicants, universities with known expertise in nuclear engineering and materials science, and other developed sources). Thirty-three members of the ion beam community attended the workshop, including 15 representatives of ion beam facilities, six representatives of DOE-NE research and development programs, an industry representative from the Electric Power Research Institute (EPRI), and the chairs of the NSUF Users' Organization and the NSUF Scientific Review Board. Four ion beam users attended as advisors to the process, but did not participate in the options assessment. Three members of the sponsoring agency, the DOE Office of Nuclear Energy, Office of the Deputy Assistant Secretary for Nuclear Technology Demonstration and Deployment (NE-5), also attended the workshop.

The NSUF IBIOW process began in December 2015 by soliciting interest in participating in the workshop from the various U.S. ion beam facility owners (universities and national laboratories). This solicitation was followed in January and February 2016 by official invitations to the workshop. The participants were asked to become involved in an ongoing process to define and weight criteria that could be used to judge the options available to DOE-NE to support and to expand domestic ion beam irradiation capabilities. The assessment process started informally, but later transitioned to the ThinkTank collaboration software.

As the goal of the workshop was to provide recommendations to DOE-NE, a data driven process was designed with the assistance of the INL's systems engineering division. ThinkTank collaborations software was selected as the tool to gather the data and link the workshop participants together. ThinkTank has been used successfully in a wide variety of government projects, notably the Nuclear Innovation Workshops held in March 2015.

The workshop participants developed a weighted list of criteria to compare the various ion beam facilities and estimated the need for future investment based on 15 criteria generated by NSUF managers as a starting point for the discussion. The final set of 10 criteria agreed upon by the workshop participants were

1. Viability for the capability to extend our understanding towards accurately simulating nuclear irradiation conditions (neutrons or fission fragments).
2. Ability of the facility to produce results that meet the needs of the DOE-NE (including cross-cutting programs) and the nuclear energy industry.
3. Ability of the facility to provide a variety of well-controlled target environments and conditions.
4. Ability of the facility to handle radioactive materials (structural materials and/or fuels) in the beams and elsewhere onsite.
5. Ability of the facility to collect and analyze materials properties and/or perform microstructural characterization data in situ.
6. Ability of the facility to produce quality-level data that can support licensing as well as verification and validation of modeling and simulation.

7. Ability of the facility to provide a variety of ion irradiations (ion types, energies, multiple beams, etc.).
8. Unique capabilities of the facility including any new technology that has the capability to close technological gaps.
9. Current or potential productivity of the facility (e.g., fewer high-impact experiments or high-volume sample throughput).
10. Ability of the facility to collect and analyze materials properties and/or perform microstructural characterization data onsite.

In addition to developing and weighting criteria, workshop participants viewed presentations from ion beam users and DOE-NE research and development programs as well as the ion beam facility representatives. Following the presentations, the workshop participants assessed each ion beam facility against each of the final 10 criteria. This exercise was performed individually, although discussions and questions were allowed. ThinkTank software was used to collect the data from the assessments. Table 1 lists the presenting facilities from the workshop.

Table 1. Facilities attending the FY 2016 Ion Beam Investment Options Workshop.

Institution	Facility
Argonne National Laboratory (ANL)	Intermediate Voltage Electron Microscope-Tandem User Facility (IVEM-TUF)
	Extreme Materials Beam Line (proposed)
Idaho State University	Idaho Accelerator Facility
Brookhaven National Laboratory (BNL)	Brookhaven Linear Isotope Producer / Brookhaven Linear Accelerator Irradiation Test Facility (proposed)
	Ion X-Ray Beam (proposed)
Los Alamos National Laboratory	Ion Beam Materials Laboratory
Lawrence Livermore National Laboratory	Center for Accelerator Mass Spectrometry
Massachusetts Institute of Technology	Nuclear Materials Laboratory (proposed)
Ohio University	Edwards Accelerator Laboratory
Purdue University	Center for Materials under Extreme Environment Facility
Sandia National Laboratories	In situ Ion Irradiation Transmission Electron Microscope
Texas A&M University (TAMU)	Ion Beam Laboratory
University of Michigan	Ion Beam Laboratory
University of Tennessee	Ion Beam Materials Laboratory
University of Wisconsin	Ion Beam Laboratory

Of the 15 facilities that were considered, only 11 were operational at the time of the workshop with four facilities proposed for construction in the future. It should be noted that the facilities are not all focused on the same research objectives and deliverable specifications, and therefore, have significantly different designs. Eight of the currently operating facilities provide vital support to nuclear materials researchers, yet the individual capabilities of these eight facilities differ based on their particular missions.

Additionally, the facility at Purdue University focuses on surface science of materials and utilizes much lower energy ions than the other facilities. The Edwards Accelerator Laboratory at Ohio University is primarily engaged in nuclear data measurement and not on the effects of ion irradiation on materials. The Idaho Accelerator Laboratory at Idaho State University is a multipurpose facility that supports a wide variety of research endeavors. These last three facilities should not be assessed in the same manner as the others. Three of the assessed facilities have (or will soon have) in situ characterization capabilities that combine ion irradiation with a transmission electron microscope, while one of the proposed facilities seeks to provide in situ characterization with an X-ray source.

Following the workshop, DOE NE-5 provided infrastructure improvement funds to two facilities: the IVEM-TUF at ANL, and the Michigan Ion Beam Laboratory at the University of Michigan. In addition, the NSUF began the partnership/affiliation process with several facilities: the IVEM-TUF, the Center for Accelerator Mass Spectroscopy at Lawrence Livermore National Laboratory, the In situ Ion Irradiation Transmission Electron Microscope at Sandia National Laboratories, and the Ion Beam Laboratory at TAMU. Two reports were issued in 2016 *Supplement to the NSUF Ion Beam Investment Options Report: Initial Results and Recommendations* [1] and *NSUF Ion Beam Investment Options Workshop* [2].

Additionally, the workshop participants collected a large quantity of data about the capabilities of their facilities for inclusion in the Nuclear Energy Infrastructure Database [3].

Method development and technology enhancement

Various experimental techniques have been developed to probe the roles of the various components of the neutron irradiation environment (including transmutation effects), including methods to increase atomic scale characterization resolution, to develop site-selective mechanical/thermal property measurement, and for in situ characterization to study dynamic response and details on the atomic scale and micron scale. For example:

- For ion irradiation, simultaneous dual-beam or triple-beam irradiations have been utilized to investigate simultaneous effects of neutron recoil damage and transmutation gas creation. Gas injection plays an important role to influence the swelling incubation period.
- For material characterization, the focused ion beam technique has greatly improved the efficiency of transmission electron microscopy (TEM) specimen preparation. The emerging technique of aberration-corrected TEM further improves imaging resolution and provides additional atomic scale details.
- Ion irradiation and in situ TEM characterization has greatly advanced the study of fundamental dynamics in radiation materials science.
- Compression/fracture testing and in situ TEM have been used to test micrometer-sized specimens within a TEM. Simultaneous TEM characterization and force loading are able to reveal dynamic structural changes, which are important to understand mechanical responses.
- The focused ion beam technique also makes it feasible to prepare specimens from the ion irradiated region only, allowing for pillar testing and fracture testing. Hence, radiation effects on mechanical property changes can be locally studied.

Motivation for standardization of techniques and methods for quality assurance

The challenge of successfully generating credible neutron-relevant data via ion beam irradiation depends on two primary principles. First, all neutron-atypical aspects of ion beam irradiation must be identified, quantified, and mitigated to the greatest extent possible. Second, a set of "best practices" for the ion beam technique must be compiled to avoid the possibility that many small, seemingly inconsequential variations in irradiation and data extraction procedures may generate significant differences in the final experimental product. Such variations, if not recognized, impair the credibility of the simulation procedure. Therefore, it is important that comparative studies be conducted to seek out the optimum and correct procedures, leading to standards and respected quality assurance.

1.3 Historical Contribution of Ion Beams to Nuclear Energy Research and Development

Although ion beam radiation effects research on materials dates back to the 1940s or earlier [4, 5, 6], ion beam irradiations for nuclear materials research and development rose into prominence in the 1970s in connection with high displacement damage studies for the liquid metal fast breeder reactor program in the U.S. and elsewhere [7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18]. In the early- to mid-1960s, the most severe radiation effects phenomena in structural materials were considered to be radiation hardening and embrittlement at low temperatures and helium embrittlement of grain boundaries at high temperatures. For the former case, the remedy was to perform irradiations at higher temperatures such as envisioned for sodium-cooled fast reactors; whereas, the high temperature helium (He) embrittlement was considered to be controllable by limiting the amount of impurities such as boron, which were known to have high thermal neutron-induced He production cross sections. Following the discovery of void formation in neutron irradiated stainless steel in the late 1960s and the prediction and verification of several other higher dose (>1–10 dpa) degradation phenomena such as radiation induced solute segregation and precipitation and irradiation creep, it became imperative to employ ion irradiation sources to provide insight on the unanticipated complex dose- and temperature-dependent radiation damage evolution in structural materials, due to the relatively slow damage accumulation rates, access limitations, and high cost of neutron irradiation experiments.

Research performed using ion beams was instrumental in providing crucial scientific insight and quantitative experimental data on numerous radiation effects phenomena on candidate structural alloys for the liquid metal fast breeder reactor program. For example, ion irradiations were used to rapidly screen numerous metals and alloys to provide initial scientific understanding of void swelling dependence on temperature and microstructural features. Ion irradiations provided some of the earliest confirmation of the high swelling resistance of titanium-modified austenitic stainless steel and 9–12% chromium (Cr) ferritic/martensitic steel, which are the reference structural materials for sodium cooled fast reactor cladding and wrap/duct designs to this day. Similarly, ion irradiations provided some of the earliest and most comprehensive experimental confirmation of radiation-induced solute segregation near interfaces such as grain boundaries and surfaces. These data provide much of the experimental foundation for our current understanding of radiation induced solute segregation. The competing influences on the stability of small precipitates due to ballistic dissolution by energetic displacement cascades and accelerated re-nucleation and growth via radiation enhanced diffusion were extensively investigated using ion beams, which has led to improved fundamental understanding of the irradiation conditions that produce stable vs. unstable precipitates. Ion irradiations also provided key early experimental research on a variety of fundamental irradiation effects parameters such as defect production efficiency (relative to the Norgett-Robinson-Torrens displacements per atom model) and direct in-cascade defect cluster production as a function of primary knock-on atom energy. Many of the advantages and limitations of ion beams for correlating neutron radiation effects in materials were summarized in a comprehensive U.S. community workshop report in 1976 [19].

A vast majority of ion beam use in the nuclear energy industry has been in the area of materials and fuels research, either as a surrogate for neutron damage (ion irradiation) or to understand the response of fuel materials during fission. The utility of ion beams for irradiation studies stems from the relatively inexpensive and rapid production of damage that allows material screening under damage conditions relevant to those in a neutron environment as well as the ability to isolate variables allowing separate effects testing. Fundamental understanding of materials behavior under long-term irradiation has been gained in many programs of interest to DOE. In the 1990s and early 2000s, the Cooperative Irradiation Assisted Stress Corrosion Crack (IASCC) Research program utilized proton irradiation to identify the most promising alloys to be irradiated in reactor for time consuming and expensive crack growth rate testing. More recently, the Advanced Radiation Resistant Materials program is utilizing proton irradiation and heavy ion irradiation to screen candidate alloys for potential replacement of alloys currently used in core internal components that are susceptible to IASCC. Currently, ion irradiation is being used to evaluate the susceptibility of accident tolerant fuel candidate claddings for their robustness under combined corrosion and irradiation that will be experienced in reactor, to solve the shadow corrosion problem in boiling water reactors and to solve the IASCC problem in light water reactors (LWRs), more generally.

There are other, less common applications of ion beams such as in sputter deposition and cleaning for materials surface chemistry manipulation and ion induced modifications in high-atomic number refractory metals for nuclear fusion applications at Purdue University’s Center for Materials Under eXtreme Environments (CMUXE) facility. Another use to date is at the Edwards Accelerator Laboratory, where ion beams are used in neutron time of flight studies and calibration of neutron detectors.

1.4 Areas of Application in Nuclear Energy Research and Development

Ion beams will most likely play an increasingly important role in the development of new materials for nuclear energy systems. They are proving to be very valuable in emulating the irradiated microstructure and mechanical properties of reactor irradiations at a fraction of the cost and time. As such, they can compress the time for radiation effects experiments by orders of magnitude, producing a comparable reduction in time to development of new materials. Applications include accident tolerant fuels testing, acceleration of screening and qualification of additively manufactured materials (or other types of advanced manufacturing) for use in advanced reactors, and material surface enhancement. Ion beams will likely find an increasing role in understanding processes in fuels. Use of cyclotrons to produce higher energy ions and consequent determination of material irradiation effects in larger and presumably more representative samples may be an additional future opportunity. Cyclotrons have been profitably used in the past. Advances in compact cyclotron accelerator technology are proposed by Massachusetts Institute of Technology and is currently in the planning stages.

1.5 Roadmap Organization

The NSUF Ion Beam Irradiation Roadmap (IBIR) Committee was established to provide to DOE-NE a report describing current and potential future contributions of ion beam technologies to address the technical and regulatory challenges of the nuclear energy community for the advancement and implementation of nuclear energy technologies that are part of the mission of DOE-NE.

1.5.1 Roadmap Report

The IBIR Committee provided a report titled *Roadmap for the Application of Ion Beam Technologies to Challenges for the Advancement and Implementation of Nuclear Energy Technologies*, whose scope is informational in essence and includes technical descriptions of ion beam technologies and their applications, including both current and future aspects, as they relate to particular areas of DOE-NE's mission. The report establishes recommendations and their impacts for DOE-NE and its programs to use at their discretion in establishing future directives and priorities. This report is not intended to have any binding authority. Table 2 shows the chapters included in the report.

1.5.2 Organization

The NSUF IBIR Committee was comprised of ion beam facility operators, subject matter experts, experimental and computational users, and programmatic, industry, and regulatory representatives. Table 2 shows the membership of the committee and their roles within the organization.

Members of the committee were welcome to engage any and all external resources or expertise to complete their writing assignments. The IBIR Committee met together on March 8, 2017, at INL. All other work on the roadmap was performed remotely. The Executive Committee gathered the chapter drafts from the leads, wrote the executive summary, and compiled the final report, which was sent to the NSUF for submission to DOE-NE by August 1, 2017.

Table 2. Ion Beam Investment Options Workshop committee membership and organization.

Area	Role	Member
NSUF Coordinator		Brenden Heidrich (NSUF/INL)
Executive Committee		Steve Zinkle (University of Tennessee)
		Gary Was (University of Michigan)
		Simon Pimblott (University of Manchester)
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		Simon Pimblott (University of Manchester)
		Gary Was (University of Michigan)
		Steve Zinkle (University of Tennessee)
Chapter 2: Current State of Ion Beam Technology	Chapter Lead	Lin Shao (TAMU)
	Chapter Contributor	Gary Was (University of Michigan)
Chapter 3: Challenges in Nuclear Energy Development	Chapter co-Lead	Stu Maloy (LANL)
	Chapter co-Lead	Blas Uberuaga (LANL)
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		Daniel Schwen (INL)
		Cem Tobasi (EPRI)
		Yongfeng Zhang (INL)
Chapter 4: Application to the advancement and implementation of nuclear energy technologies	Chapter co-Lead	Todd Allen (University of Wisconsin)
	Chapter co-Lead	Lance Snead (Stony Brook University)
	Chapter Contributors	Lynne Ecker (BNL/NSLS-II)
		Meimei Li (ANL/IVEM-TUF)
		Ming Tang (LANL)
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Chapter 5: The Path Forward	Chapter Contributors	Gary Was (University of Michigan)
		Simon Pimblott (University of Manchester)
		Steve Zinkle (University of Tennessee)
		Michelle Bales (NRC)

2 Current State of Ion Beam Technology

2.1 Types of Ion Beam Capabilities

Globally, more than 35,000 particle accelerators have been built over the past 60 years for industry processing, medical therapy, and materials research. During the past half century, the role of particle accelerators in nuclear materials research has been very important, with ~200 accelerators currently being used for such research [20]. Various ion beam research laboratories in the U.S. are active in this area and are continuously being upgraded to meet the needs for radiation studies of nuclear materials. Table 3 summarizes the facilities and capabilities of selected groups in U.S. national laboratories and universities.

Table 3. Facilities attending the FY 2016 Ion Beam Investment Options Workshop.

Institution/Facilities	Accelerators	Capabilities
ANL/IVEM-TUF	2 MV Tandem 500 kV implanter	Up to 1573K, radioactive materials allowed, in situ TEM during irradiation
Idaho State University/Idaho Accelerator Laboratory	4.0 MV Tandem	Radioactive materials allowed
LANL/Ion Beam Materials Laboratory	6 MV Van de Graaff Vertical 9 MV Tandem	Up to 1473K, corrosion studies in liquid metal and molten salts, radioactive materials allowed
LLNL/Center for Accelerator Mass Spectrometry	FN 10 MV Tandem Van de Graff 1 MV Tandem 1.7 MV Tandem	Up to 1023K, radioactive materials allowed
Ohio University/Edwards Accelerator Laboratory	4.5 MV Tandem Van de Graaff	
Sandia National Laboratories/In situ Ion Irradiation Transmission Electron Microscope (I3TEM)	6 MV Tandem 1 MV Tandem 3 MV Pelletron Implanter 100 kV Nanoimplanter 10 kV Colutron	Up to 1473K, in situ TEM during irradiation, radioactive materials allowed
TAMU/Accelerator Laboratory	10 kV Implanter 140 kV Accelerator 400 kV Van de Graaff 1.7 MV Tandem 3 MV Tandem	Up to 1273K, radioactive materials allowed, dual beam irradiation allowed, corrosion in high pressure high temperature water (planned)
University of Michigan/Ion Beam Laboratory	3 MV Tandem 1.7 MV Tandem 400 kV Implanter	Up to 1500K, corrosion testing in high pressure, high temperature water, radioactive materials allowed, dual beam and triple beam irradiation
University of Tennessee/Ion Beam Materials Laboratory	3.0 MV Tandem	Up to 1475K
University of Wisconsin/Tandem Accelerator Ion Beam Laboratory	1.7 MV Tandem	Up to 1073K, radioactive materials allowed, corrosion in molten salt (planned)

2.1.1 Current State-of-the-Art/State-of-the-Practice

Various ion beam capabilities, either for ultra-low energy implanters with beam energies of <1 kilo-electron volt (keV) or high energy implanters with beam energies of a few mega-electron volts (MeV), have been developed during the past half century largely for doping of semiconductor devices. Energy contamination (spreading of beam energy), space charge effects (beam loss due to beam expansion upon transport), and fluence control (errors due to charge neutralization upon transport) represent three outstanding issues driving technology improvement.

Technology upgrades of accelerators for semiconductor doping care are directed primarily toward beam quality and throughput. Typical beam currents are larger than 1mA and target areas can be as large as 12-inch diameter (under target rotation). In comparison, accelerators for nuclear materials studies do not require a large beam spot and complicated target cooling, and most ion beam facilities use research type accelerators provided by National Electrostatics Corp. (NEC) or High Voltage Engineering, Inc. Additionally, some low or medium energy industrial-type implanters have been utilized in several labs for implantation and irradiation studies. High-energy implanters are not used at this point but they are available.

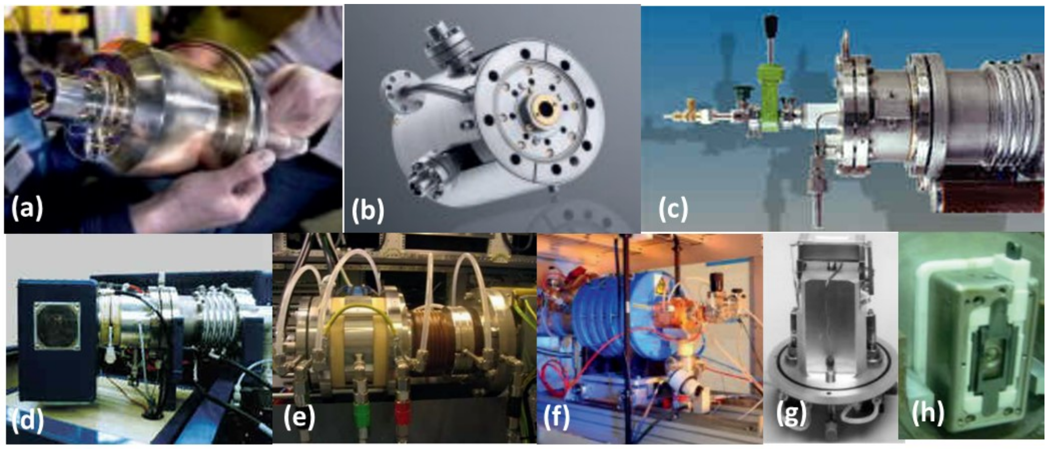

Figure 1. Ion sources of various types including (a) Duoplasmatron, (b) Penning, (c) source of negative ions by cesium sputtering, (d) radiofrequency, (e) Chordis, (f) electron cyclotron resonance, (g) Freeman, and (h) Magnetron [21].

As the key component that determines the beam current, a variety of ion sources have been developed that are based on electron bombardment, ion bombardment, plasma discharge, radiofrequency discharge, microwave and electron cyclotron resonance, and laser driven techniques. Figure 1 shows typical types including Duoplasmatron, Penning, source of negative ions by cesium sputtering (SNICS), radiofrequency, Chordis, electron cyclotron resonance, Freeman, and Magnetron sources.

In a recent technology improvement, NEC has commercialized its Multi-Cathode SNICS source (Figure 2a) to allow cathode replacement without disturbing the vacuum, in addition to incorporating significant modification of internal components to increase the beam current by a factor of three. NEC has developed the toroidal volume ion source (Figure 2b) for high current hydrogen (H) (>100 µA for >1,000 hours) and He (>20 µA for >1,000 hours) beams.

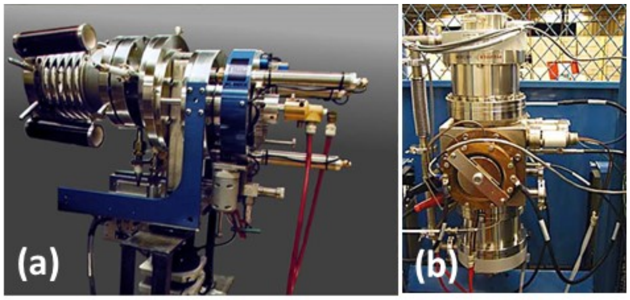

Figure 2. (a) Multi-Cathode SNICS source and (b) toroidal volume ion source recently developed by NEC.

Depending on the method of acceleration, particle accelerators can be divided into electrostatic accelerators and oscillating field accelerators. In addition, depending on whether the initial ion charge is positive or negative, accelerators can be divided into singled-ended accelerators and tandem accelerators. Main categories of accelerators include Van de Graaff, Cockcroft-Walton, Linear Accelerator, Cyclotron, Betatron, Microtron, Synchrocyclotron, and Synchrotron.

For nuclear materials research, proton irradiation is primarily used for simulation of boundary segregation and irradiation-assisted stress corrosion cracking while self-ion or heavy-ion irradiation is primarily used for simulation of neutron induced damage cascade creation. In both cases, ion beam energies are typically in the region of 1–7 MeV. Consequently, the major types of accelerators currently in use are the High Voltage Engineering, Inc. Tandetron accelerator based on the Cockcroft-Walton design and the NEC tandem accelerator based on the Van de Graaff design.

In comparison with industrial implanters, research accelerators have not dramatically evolved during the past three decades. The ion source, vacuum, and acceleration and target chambers have largely followed the original design concepts. On the other hand, available accelerator designs largely meet currently defined research needs. Therefore, there is no critical need to call for new accelerator concepts in the energy region that is most interesting to nuclear materials research.

2.1.2 New and Promising Technologies

For nuclear materials application, current acceleration techniques are largely satisfactory, especially considering the low cost and reasonable size of accelerators in the energy region <10 MV. New concepts and designs, however, always drive high-energy particle physics research to reduce costs. A few cost-effective concepts have been proposed and are currently being explored.

For instance, in the laser-driven plasma accelerator design, an intense laser pulse is introduced into a plasma consisting of equal numbers of electrons and ions. Under the strong electric field caused by the laser, the lighter plasma electrons are separated from the more massive positive ions, hence creating a trailing longitudinal density wave with very high charge separation that propagates through the plasma. The charge separation creates a strong electric field and particles injected into the correct phase of plasma waves can be locked and accelerated to energy of order 1 giga-electron volt (GeV) over a few millimeters. Figure 3 (a-c) shows schematics of plasma density waves (blue) induced by different laser pulse fields in a “laser wakefield accelerator,” a “plasma beat-wave accelerator,” and a “self-modulated laser wakefield accelerator," respectively. This technique opens various possibilities ranging from low cost colliders to high-speed electron diffraction and medical imaging devices [22].

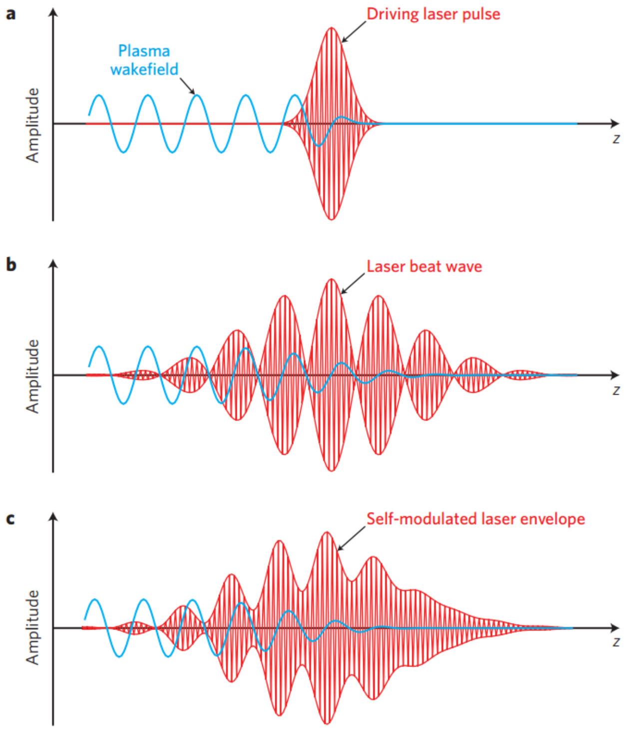

Figure 3. Schematics of driving laser field (red) and created electron density waves (blue) for the cases of (a) a single laser pulse, (b) beating of two laser fields with a frequency difference equal to the plasma frequency, and (c) self-modulation of along laser pulse by its interaction with the plasma.

2.2 Ion Beams for Nuclear Research and Development

2.2.1 Motivation

Most irradiation consequences that the nuclear community is dealing with today were unknown when the current LWR fleet was being built and the early advanced reactors were being designed. Up to ~1970, designers were unaware of radiation-induced segregation or IASCC. The first observation of voids was in 1967 [23] and this single discovery significantly slowed the progress of fast reactor development worldwide. The concern over void swelling triggered the initiation of a 20-year, $150M in 1980 dollars (about $380M in 2013 dollars) [24] research effort aimed at developing alloys that were resistant to void formation. Most of the program resources were aimed at optimizing a single alloy – austenitic stainless steel. Today, a number of additional degradation modes have surfaced including radiation induced precipitate stability, radiation induced segregation, irradiation creep, irradiation hardening and embrittlement (reduction in fracture toughness), and IASCC.

Traditionally, research to understand radiation-induced changes in materials is conducted via radiation effects experiments in test reactors, followed by a comprehensive post-irradiation characterization plan. Modeling of the radiation damage process helps to reduce the need for experiments covering the entire parameter space by providing predictive capabilities. Both fast and thermal spectrum reactors have been used for radiation damage studies. In either reactor type, the damage rate is at best only slightly faster than that in a commercial reactor. Thus, the capability to advance our understanding of radiation effects has been impeded by the emergence of several barriers. First, test reactors cannot create radiation damage significantly faster than that in commercial reactors, meaning that radiation damage research often cannot "get ahead" of problems discovered during operation. Second, there is a paucity of test reactor capability in the world, and especially in the U.S., for addressing known issues in advanced reactor concepts. Finally, the cost of conducting such experiments has risen dramatically in the past several decades.

The U.S. has only two high-power test reactors (1) High Flux Isotope Reactor at Oak Ridge National Laboratory and (2) the Advanced Test Reactor at INL and both are thermal reactors. To address the issue of life extension of light water reactors would require more than one decade of irradiation to reach the damage levels of interest as commercial reactor components receive in the 40–60 year time frame. There are *no* fast spectrum test reactors in the U.S (though High Flux Isotope Reactor and Advanced Test Reactor can approximate a fast spectrum by shielding the thermal and epithermal flux). Worldwide, only the BOR-60 fast reactor in Russia is currently active and accessible for radiation effects experiments.

Light water reactor core components will see damage levels of 100 dpa after 40–60 years and well above that value with life extension. Fast reactor core components will likely see damage levels of 200 dpa. The components of the newest reactor concept that is designed to substantially eliminate nuclear proliferation risk and significantly reduce radioactive waste production (traveling wave reactor) must withstand ~600 dpa. Thermal test reactors can generate damage in materials at a rate of 3–10 dpa/yr and accessible, fast spectrum test reactors can reach 20 dpa per year (Figure 4), so to utilize existing test reactors for certification of materials integrity through the end of life of any reactor type would require one or more decades of irradiation, followed by expensive hot cell analysis due to the high level of radioactivity of the irradiated samples. Neutron irradiation and testing facilities to test fuels and materials out to the damage level required to certify their operation are extremely limited in the U.S. and elsewhere.

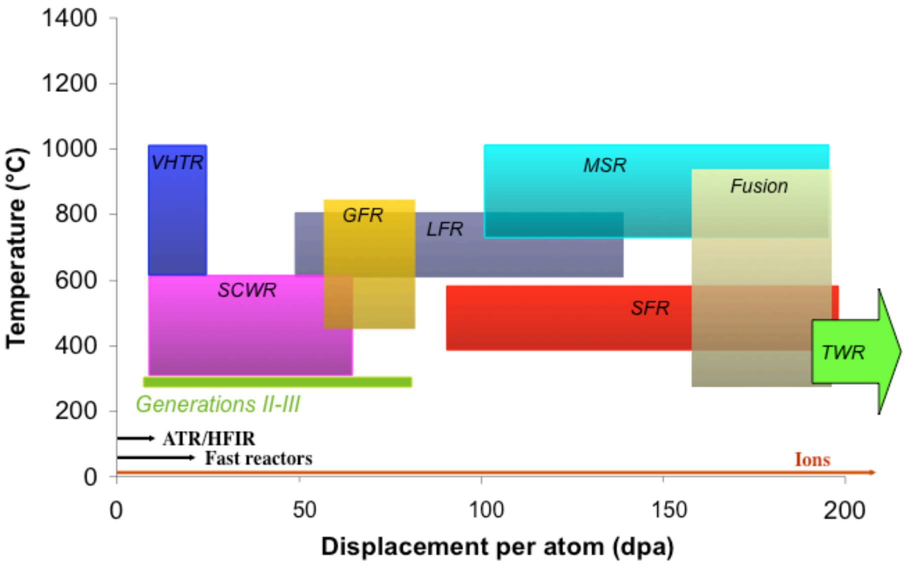

Figure 4. Schematic of the temperature-dpa requirements for various reactor concepts and the achievable annual damage rates in different test reactors and with ion irradiation.

2.2.2 Advantages and Limitations

The advantages of ion irradiation are many. Dose rates (typically 10^{-3} to 10^{-4} dpa/s) are much higher than under neutron irradiation (10^{-7} to 10^{-8} dpa/s), which means that 200 dpa can be reached in days or weeks instead of decades. Because there is little activation, the ion beam irradiated samples are not highly radioactive. Control of ion irradiation experiments is typically much better than experiments in reactor. Measurement of temperature, damage rate and damage level is difficult in reactor, often resulting in reliance on calculations to determine the total dose, and estimate irradiation temperature. By contrast, ion irradiations have been developed to the point where temperature can be extremely well controlled and monitored, and damage rate and total damage can be also measured continuously throughout the irradiation and with great accuracy.

The high rate at which ion irradiations can be conducted coupled with the absence of residual radioactivity are the key attributes that make this route to advancing our understanding of radiation effects so attractive. High damage rates mean that the radiation effects arising from 20 years in reactor can be achieved in two days in an accelerator. The lack of activation means that samples can be handled as if they were unirradiated; eliminating the need for the extremely high investment in time and cost associated with the use of hot cells and dedicated "active" characterization instrumentation. Further, the optimum voltage for accelerator irradiation experiments is in the few MV range, making such experiments accessible by many laboratories. The net result is that ion irradiation is 10–1,000 times less costly and 10–100 times quicker than test reactor irradiation.

Ion irradiation has several potential limitations; in particular, the limited volume of irradiated material, the effect of high damage rate on the resulting microstructure versus reactor-relevant damage rates, and the need to account for important transmutation reactions that occur in reactor, such as the production of He and H. Understanding and modeling the microstructure-property relationship, and the development of micro-sample fabrication and testing, while not a replacement for bulk property determination, hold the promise for minimizing the drawback of limited irradiated volume [25, 26, 27]. The extent to which high damage rates can produce microstructures relevant to reactor conditions is a major challenge, but significant progress is being made to address this issue [28, 29, 30]. The importance of He and H production through transmutation is evident in the large impact of He on processes such as void swelling [31]. The solution to this problem is simultaneous irradiation of a target with self-ions to cause damage, and with He and/or H to emulate transmutation using multiple accelerators.

To qualify ion irradiation to study neutron irradiation, it is necessary to reproduce as best as possible both the neutron irradiated microstructure and the neutron-induced macroscopic property changes using ion irradiation. Because these microstructures are very complex, the task of verifying that the ion irradiation microstructures are similar to that of a reactor irradiation is correspondingly complex. This task is best addressed using a combination of state of the art experimental interrogation techniques closely coupled to modeling, which can yield mechanistic understanding of the defect development process. The next section considers the approach for verifying and modeling the irradiation induced microstructure and the structure – property relationship using experiment and modeling.

2.2.3 Best Practices

Performing ion irradiations requires the selection and control of many experimental parameters such as temperature and local dose rate. It is well known that the resulting microstructure is sensitive to these parameters as well as the manner in which the irradiation is being conducted. While several ASTM standards have addressed aspects of the issue [32, 33, 34, 35], what is missing is a set of best practices that those conducting ion irradiation should follow. As summarized in the following, a group of experienced ion beam practitioners has proposed a set of best practices for the use of ion irradiation to study radiation effects, including sample temperature control, temperature monitoring, beam dosimetry, irradiation mode, vacuum control, and method of determining displacement damage.

Beyond the study of radiation effects, ion irradiation is being explored for the capability to emulate the irradiated microstructure resulting from reactor irradiations. This application brings additional requirements, including the selection of ion type, ion energy, damage rate difference, the effects of injected interstitials, the damage profile, and modes of injecting He into the sample. The best practices described herein are aimed at addressing the use of ion irradiation to achieve microstructures comparable to those in reactor. These two sets of best practice recommendations are provided below.

Best Practices for Conducting Ion Irradiation to Study Radiation Damage in Materials

This set of best practices aims to provide guidance for conducting ion irradiation in a way that results in high quality, reproducible results. The following parameters/conditions are specified to achieve this goal.

Irradiation Mode: Only mass-analyzed beams should be used.

Temperature Control: Ion irradiations should utilize backstage heating and cooling (to achieve rapid response time) plus a well-controlled sample-stage interface (e.g., soft metals with high conductivity, high conductivity gas, liquid metal, and uniform physical contact).

Pastes, tapes and any organic (carbon-containing) materials should be avoided. Vacuum compatible, high temperature paste may be acceptable if it is known that it will not contaminate the vacuum.

Temperature Monitoring: The use of two independent methods of temperature monitoring with at least one *on* the irradiated surface is recommended, for example in situ infrared pyrometer of the sample surface and thermocouples attached to the irradiated surface. Ideally, both should be measuring the in-beam irradiated surface temperature but having one out of beam is acceptable.

Dosimetry: Measurement of the current deposited on the sample (with electron sputtering suppressed or correcting for it) is optimal. The stage must be electrically isolated (except for the monitoring circuit) from the chamber. This method is only practical for irradiations that do not produce much electron sputtering (such as protons).

The next best method is to use a Faraday cup – slit system placed close to the sample stage. The first Faraday cup will measure the total current down the beam line, the slits will record the current hitting them, and the second Faraday cup will determine the current passing through to the stage and to the target/sample. Even if the current on the slits is not electron-suppressed, the magnitude can be tracked to determine changes in current with time. A second Faraday cup should be inserted periodically (and momentarily) to check the current incident on the target.

Beam uniformity should be verified using methods such as a multi-pin Faraday cup, a moveable slit system, or other techniques that provide quantitative information on beam current as a function of position. Because of the nonlinear response of beam-induced florescence of materials such as alumina or quartz, this method is not recommended to ensure beam uniformity.

Vacuum control: Irradiations should be conducted with a pressure in the irradiation chamber below 1×10^{-7} torr. The sample chamber should be equipped with a high vacuum pump. The sample chamber should be baked periodically or as needed. If possible, samples should be heated prior to irradiation to outgas the samples and stage without altering the microstructure. The vacuum near the sample should be monitored continuously throughout an irradiation and, if possible, a cold trap should be used.

Determination of dpa: Use the Stopping and Range of Ions in Matter (SRIM)[2] in the quick K-P mode [36] for determination of dpa depth profile as well as the value at the depth(s) where post irradiation measurements are being made. The full cascades mode in SRIM should not be used due to errors in the calculated dpa levels [36]. It is recommended to show the damage profile in publications. The displacement energy should be taken from [32] and always listed in the publication.

The following section provides best practices for conducting ion irradiation with the aim of emulating reactor irradiation conditions.

Best Practices for Conducting Ion Irradiation to Emulate Reactor Irradiation Conditions

Irradiation mode: It is known that raster-scanning affects swelling differently than does a defocused beam [37, 38]. The effect of raster-scanning on other microstructure features is less well known. For experiments aimed at examining steady state radiation damage phenomena, a defocused beam (or defocused with a minor amount of wobble) should be used. Raster-scanning may be used in cases in which it is known to capture the key features of reactor irradiation, such as proton irradiation relevant to LWR core conditions.

Damage rates: Invariance theory for point defect evolution should be used as a guide to determining the temperature change required to account for the damage rate difference between ion and neutron irradiation [39]. Given the limitations of the theory and the dependence of the temperature shift on the specific microstructure feature, invariance theory calculations should only be used as an approximation and must be accompanied by experimental confirmation. The goal should be to determine an irradiation temperature at which the critical microstructure elements emulate those in reactor.

Selection of ion type: For bulk irradiations (ions remain in sample), ion species should usually be the same as one of the major alloying elements so as to minimize the change in composition in the sample.

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[2] See www.srim.org.

For both bulk and in situ irradiations in the TEM, the selected ion/energy combination should be made so as to produce a primary knock-on atom energy spectrum that is as close as possible to that produced by neutrons in reactor.

Penetration depth (ion energy): The deeper the penetration of the ion the better as a larger depth means a larger region for analysis that is far (>300 nm) from the surface and also from the damage peak and injected interstitial. Penetration depths of at least 1 µm are recommended. Very high dose irradiations (>200 dpa) at elevated temperatures should generally use ions with ranges ≥1.5 µm due to diffusional broadening of the implanted ion profile.

Injected interstitial: Analysis of the damage in the region of the injected interstitial should be avoided due to potential suppression of void swelling levels and other microstructural effects. Attention must be paid to the growth of the injected interstitial tail into the “valid” analysis region at very high dose.

Techniques to flatten the damage profile: It should be recognized that techniques such as use of multiple ion energies, energy degrader foils, or rocking the sample relative to the incident ion beam brings the effects associated with the damage peak and the injected interstitial into the analysis region, and therefore are not recommended.

Injection of He: Possible modes of He injection are pre-injection cold, pre-injection hot, sequential, and co-irradiation/implantation. Co-irradiation/implantation to match the He/dpa ratio of the reactor irradiation is the most representative way to emulate irradiation conditions in reactor.

3 Challenges in Nuclear Energy Development

Understanding and ultimately predicting the response of materials in irradiation environments is crucial for making the safety case for nuclear energy systems, for extending the life of current reactors, and for designing new materials for future application. However, actual in situ reactor irradiations are costly and challenging to assess. While not a replacement for neutron irradiations, ion beams offer a complementary route to interrogate material response to extreme environments and, as shown in Figure 5, can aid in all stages of material development and deployment. Further, ion beams can produce a large amount of data on well-controlled samples that prove invaluable for modeling verification and validation. Here, we describe how ion beams can assist in the development and deployment of nuclear materials referenced to the steps provided in Figure 5.

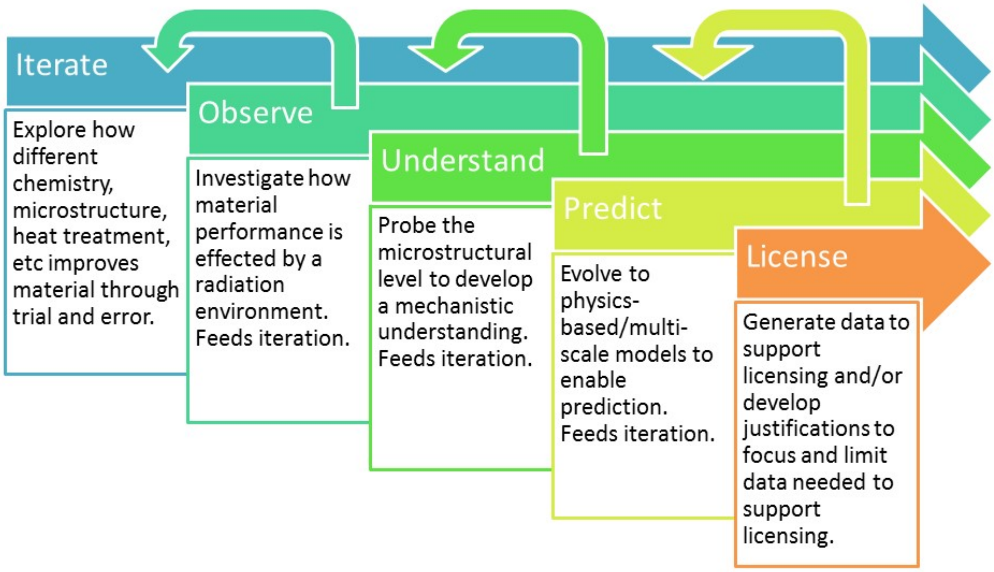


Figure 5. The use of ion beams to assist the development and deployment of nuclear materials through the various steps of iteration, observation, understanding, prediction, and licensing.

3.1 Developing and Supporting Methods for Rapid and Cost Effective Materials Selection and Development

The Problem: *Materials development has historically been a very cost and time intensive endeavor.*

Many of the hoped for advancements in nuclear energy will require new materials that can withstand increasingly aggressive environments including intense high-energy neutron fluxes, high temperatures and corrosive environments (e.g., super critical water, molten salt, liquid metals). Relatively minor changes in chemistry, microstructure and phase structure in a material can have major impacts on its performance. The phase space (chemistry, microstructure, phase structure, etc.) is wide and material developers need ways to explore, with fine resolution, the optimal parameters. To support the needs of the nuclear community, methods for rapid and cost effective material selection and development are needed.

Fuels (nuclear fuel and cladding) and structural materials (e.g., grid plates, ducts and other core components) in current or proposed (e.g., Gen IV) nuclear reactors experience some of the most extreme environments consisting of a high neutron flux at temperatures from 250°C to >1,000°C while under stress and in contact with different coolants or the nuclear fuel itself. Testing in this challenging environment is very difficult, because of the extreme nature of the environment. Thus, once a material is qualified for such an application, it is very difficult and expensive to qualify another material to replace it. In fact, there are very few places in the world where materials can be tested in nuclear reactors under these extreme environments.

Ion beams offer the ability to test materials under a high particle flux, under stress and even in contact with a coolant. In addition, through selecting the correct energy of the particles, one can avoid activating the sample which makes the post irradiation examination much easier and less expensive. A few drawbacks with using ion beams are that the depth of penetration of the ion is very small, the damage can vary significantly within that short depth of penetration, and the rate of damage accumulation is much higher than in a reactor which is an advantage and a curse at the same time. The development of small scale mechanical testing along with ion irradiation greatly expands the mechanical properties that can be measured on specimen during or after irradiation. Although there are many advantages with using ions to irradiate materials, significant research is needed to accurately correlate ion irradiation data to that measured under actual conditions in a nuclear reactor.

3.2 Characterizing Fundamental Material Response under Irradiation (Observe)

The Problem: *Mechanical properties of irradiated materials are controlled by the microstructure, and evolution of the irradiation-induced microstructure must be understood to develop a fundamental description of material properties in operation so that designers can accommodate or minimize the changes. These changes may arise from radiation-induced defect accumulation, segregation, precipitation, etc. and are sensitive to the starting microstructure and irradiation conditions.*

Irradiation drives materials into highly non-equilibrium states that are essentially impossible to predict from equilibrium considerations such as phase diagrams. Even the simplest materials exhibit complex behavior under irradiation; consider the formation of stacking fault tetrahedra in face-centered cubic metals. As material complexity increases, the evolution of the material under irradiation becomes increasingly complex. The possible responses become even more complex and myriad when materials are either in contact with other materials or the environment. For example, recent experiments on model metal/oxide ceramics [40] have found that irradiation drives chemical mixing between the metal alloy and the oxide, enhancing the amorphization rate of the oxide. To have any predictive capability for such radiation-induced phenomena, one must first have some understanding of what kind of changes can occur in order to include them in models. While atomistic modeling can provide direct insight without any preconceived notion of the possible response, higher-level models can only simulate behavior that is directly input into the model. At the same time, atomistic modeling is limited to relatively small length scales and short time scales that prohibit detailed study of complex radiation-induced behavior over experimentally relevant conditions. Thus, there is a great need to observe the fundamental response of materials to irradiation to have any possibility of developing predictive models.

Because of the accident at Fukushima, there is a surge in interest in accident tolerant fuels. The proposed fuels are wide ranging and include novel uranium compounds such as silicide and nitride. In principle, these compounds have a higher fissile density and higher thermal conductivity and thus offer advantages as compared to uranium dioxide (UO_2). However, a number of silicide and nitride phases are possible and their basic structural integrity under irradiation must be assessed in order to select promising compounds for further study and development.

Next-generation nuclear energy systems, including concepts such as the traveling wave reactor advocated by TerraPower, Inc., require clad and structural materials that can withstand extremely high doses, in some cases, greater than 500 dpa. Consequently, the research and development community is examining a wide-range of new materials in search of one that can tolerate these environments. These range from established materials such as oxide-dispersion-strengthened steels to so-called high entropy alloys. One thing all of these materials have in common is their chemical complexity. While stable as synthesized, an open question often exists whether new alloys will retain that stability under. Will high entropy alloys begin to phase separate under the driving forces of irradiation? Are the oxide particles in oxide-dispersion-strengthened steels stable against irradiation and are other chemistries likely to be more stable? How such materials will evolve under extremely high doses, even qualitatively, needs to be examined before their behavior can be predicted.

In the quest to identify potential waste form materials that could be used to encapsulate radioactive species for long term disposal, complex oxides have become a leading candidate. The drivers are two-fold. First, natural analogs exist in which crystalline minerals are observed to retain their crystallinity while bearing radioactive actinides. Second, the artificial mineral composite SynRoc contains a number of crystalline oxide phases. However, the natural analogs often contain complex oxide phases that include titanium as a key constituent. These compounds, such as pyrochlore ($A_2B_2O_7$), have a wide range of possible chemical composition, offering the possibility that chemistries other than B=Ti might prove even better hosts for radionuclides. Indeed, zirconium and hafnium bearing pyrochlores have been found (using ion beams) to be much more radiation tolerant than the titanium compounds.

Observing even the qualitative response of complex materials to irradiation is critical for knowing what kinds of phenomena need to be included in predictive models. However, in-pile testing is simply too expensive and too time consuming to provide answers in a timely manner for the large suite of possible materials. Ion beams can play a crucial role in understanding basic irradiation-induced phenomena. Because of the extremely high flux, the damage induced by ion beams might be considered a bounding scenario of what can potentially happen to the material in-pile. That said, ion beams provide a window into the possible phenomena that can occur that, once observed, can be understood and modeled to provide a predictive capability applicable to other conditions.

3.3 Developing a Mechanistic Understanding of Microstructure Evolution under Irradiation

The Problem: *Today many of the effects of irradiation are only quantified empirically, and at a macroscopic level. To better control unwanted irradiation effects and design better materials, the microscopic evolution must be understood. This advance requires fundamental models of the physical changes that occur under irradiation.*

Experimental observation is limited to a small subset of the operation conditions encountered in nuclear reactors. The limitations include time, dose, dose rate, and the primary knock on atom energy spectrum. To predict material behavior outside of the experimentally observed conditions, a solid mechanistic understanding of the physical processes occurring under irradiation and knowledge of the state variables, such as microstructural features, that govern material property evolution is required.

Phenomenological interpolation of experimental data suffers from the difficulty of experimentally reproducing reactor irradiation conditions. While the Eason, Odette, Nanstad, and Yamamoto [41] model for reactor pressure vessel embrittlement has widely been considered successful, it is an interpolation-based model based on actual reactor irradiation data from coupons. Data availability hinges on the availability of irradiated coupons, which are irradiated in real time in power-producing reactors and are unavailable for operating periods exceeding the current reactor fleet. Data extrapolation bears the risk of omitting new phenomena, not present in the data set, such as late blooming phases.

A mechanistic understanding of a complex process entails successively breaking the process down into smaller sub-processes and mapping their interactions. For example, the fission gas life cycle can be broken down into generation, intra-granular diffusion and trapping, intra-granular precipitation, re-solution, grain boundary precipitation, percolation, and venting. Models for all sub-processes do exist, however the fine details of each sub-process are debated. Sub-process level experiments and validation could help improve the existing models. One example is the re-solution process, for which two mechanisms are proposed (heterogeneous and homogeneous re-solutions).

Irradiation conditions in reactors are characterized by broad spectra of primary knock-on energies, distributions of temperature, and mechanical loading. For model building and validation purposes, systematic experiments with mono-energetic ions, and well-defined thermal and mechanical boundary conditions, would be preferable. These experimental conditions can be provided by ion beam facilities.

In practice, the microstructure, which is basically defined by the arrangement of atoms is represented in a homogenized manner using a list of field/state variables such as concentrations of defects and impurities, grain size, etc. While the fuel behaviors are resolved at the macroscopic scale (millimeter and days), much finer resolutions in both time and space are needed to accurately describe the underlying physics. For example, fuel swelling can be measured at the pellet level, but the cause, i.e., the production of lattice defects and fission products, occurs at the atomic scale in space (Angstrom or 10^{-10} m) and the time scale of atomic jumps (picosecond or 10^{-12} s). Therefore, the microstructure-based approach is necessarily multiscale in time and space.

In the Nuclear Energy Advanced Modeling and Simulation (NEAMS) mesoscale fuel modeling approach the internal state of the fuel and its microstructure are described using continuous field variables. The time evolution of these field variables is governed by the partial differential equations of the phase field method. Discrete atomistic phenomena can be modeled by coupling to lower length scale methods, such as density functional theory, molecular dynamics or Monte Carlo techniques.

To develop a mechanistic understanding for both fuels and structural materials it is strongly desired that the experiments be done with detailed three-dimensional characterization of initial and final microstructure, and ideally in situ measurement when transient behavior is concerned. Due to the harsh reactor environment (combining irradiation, fission, high temperature/temperature gradient and stress for fuels, and irradiation, corrosive medium, high temperature and stress for structural materials), real-time monitoring and measurement are essentially prohibited. Even for post-irradiation experiments, the cost and the time needed are significant for materials irradiated in reactors due to the radioactive hazards. Meanwhile, many of the phenomena are irradiation condition dependent, meaning that long experimental time is needed to observe the effects that develop over the long service time of fuels (~years) and materials (~years to decades) in reactors.

Compared to the reactor environment for fuels, ion beam experiments are usually done without the fissile events and the large temperature gradient. The primary damage state and the flux of ion beam irradiation can also be very different from those of neutron irradiation in reactors. Therefore, it might not be possible to use ion beam experiments to replace reactor tests. However, ion beam experiments are still of tremendous value in advancing fundamental and mechanistic understanding, and in developing and validating predictive models. By coupling predictive modeling and ion beam experiments, it is possible to reduce or accelerate the reactor tests, thus reducing the time and cost for materials design and development. For the relatively uncomplicated environment and the absence of radioactive hazards, ion beam experiments sometimes serve better the purpose of validating predictive models. Without radioactive hazards, detailed characterization of the initial and final microstructures, and at the same time property measurements, to help establishing and validating the microstructure-property correlations are possible. In situ experiments become possible for better modeling and experiment comparison. Due to the relatively lower cost compared to reactor tests, more systematic investigations of the effects of controlling factors such as temperature and flux are possible. To better elucidate this, the role of ion beam experiments in validating predictive models is elaborated by taking the multiscale, microstructure based fuel performance models being developed under NEAMS as an example.

3.4 Development and Validation of Robust Predictive Models for Microstructure Evolution under Irradiation

The Problem: *Many of the physics-based/multi-scale models cannot be validated with large-scale experiments because the instrumentation does not measure sufficient detail. If we are going to advance to physics-based/multi-scale models, we need to progress to a whole new resolution of measurement. This requires well-defined initial conditions, well-controlled irradiation conditions, and high-resolution characterization. Further, reactor experiments convolute a large number of effects at once and separating phenomena can significantly complicate validation efforts. Finally, typical reactor experiments are, by nature, ex situ and post mortem, limiting the data available for validation.*

One way forward from mechanistic understanding is to develop predictive models to predict transient materials behaviors in reactors. Such predictive models are highly desired for various DOE programs (e.g., to predict fuel performance for NEAMS and Consortium for Advanced Simulation of Light Water Reactors and to predict reactor pressure vessel embrittlement for LWR sustainability). Ideally, these models need to capture the physics occurring in materials in reactors. Predictivity is assured by plugging in correct materials parameters and the model can be extrapolated to different operation conditions and materials by switching the materials parameters, provided the same physics still operate. However, it is extremely challenging to develop such predictive models due to the lack of sufficient mechanistic understanding, particularly for fuels, and due to the significant time and cost to achieve that experimentally.

For these reasons, historically most fuel performance models were established using experimental data in the form of empirical correlations between fuel properties and burnup, a measure of the amount of energy extracted from fuels. The applicability of models developed this way is thus limited to fuels and operation conditions where experimental data are obtained to establish the correlations, limiting its usage for fuel and reactor development and design. To overcome this drawback, a new approach, so-called microstructure based fuel performance modeling [42], has been established by the NEAMS program. The same philosophy has been borrowed by the GRIZZLY (a code for modeling degradation of nuclear power plant systems, structures, and components due to exposure to normal operating conditions) project under LWR sustainability for reactor pressure vessel embrittlement [43]. This new approach centers on the microstructure (here microstructure refers to the phases and defects and chemistry as well as interfaces and dislocation structure), which governs the properties of materials. The major tasks are to develop materials models that predict microstructure evolution and the microstructure-property correlations.

The experimental demands posed above for developing a mechanistic understanding of the fuel and structural material processes leading to property evolution and degradation also apply to the important step of model validation. The computational combination of multiple sub-processes will result in a prediction for both the microstructural and effective property evolution in a material sample. Validation is necessary to ensure that the set of simulated sub-processes encompasses all relevant phenomena and the coupling is implemented correctly.

During fuel operation, a number of phenomena are occurring at the same time. In the NEAMS approach, this means the entire list of field variables are evolved concurrently: for each individual variable multiple mechanisms may operate at the same time. The overall fuel behaviors are predicted by coupling models for individual variables, with each model possibly involving coupling individual mechanisms. Along with these models are the materials parameters that define the material system. Therefore, validations can be done with different levels of complexity using well-designed ion beam experiments. A few examples are elaborated below:

1. Measurement and derivation of key materials parameters, such as diffusivities of lattice defects and chemical species
2. Individual mechanism concerning a certain microstructure feature (field variable), such as irradiation induced segregation in binary alloys
3. Competing mechanisms concerning a certain microstructure feature, such as grain growth under thermal aging and irradiation
4. Coevolution of multiple microstructure features: gas ion implantation and the consequent bubble evolution and swelling
5. Structure-property correlation, such as irradiation induced degradation in thermal and mechanical properties.

In Tonks et al. [44], a sophisticated thermal conductivity model has been developed for UO_2 considering the effects of grain size, lattice defects, fission products, and bubbles. Ion beam irradiation can be used to create various microstructures followed by measurement of the corresponding thermal conductivity. This allows for the validation of the contribution of each microstructure: grain size (before irradiation), lattice defects, bubbles, etc.

The list can be expanded with more examples. Although many of the above phenomena are not the same under ion irradiation and neutron irradiation in reactors, they are useful to explore and validate the underlying physics, and therefore are valuable for nuclear materials development. Combined with the development of predictive models, ion beam experiments may be used to reduce reactor tests needed for materials qualification and licensing.

3.5 Developing Methods for Scoping, Focusing, and Limiting In-pile Tests Needed for Material Licensing Basis

The Problem: *In-pile testing is expensive and time consuming, so it is desirable to have a very focused test matrix that addresses the most important challenges and understanding areas of greatest uncertainty with the material.*

Ion beam irradiations can assist in scoping, focusing and limiting neutron irradiations needed to develop a material's licensing basis. Regulatory requirements often specify that data collected in-pile is necessary to build and validate material property models and correlations. However, in-pile testing of materials is expensive and time consuming. For in-pile testing, it is desirable to have a refined test matrix that addresses the most important challenges and focuses on the areas of greatest uncertainty. Research areas of interest for ion beam irradiation studies to inform subsequent in-pile test programs include, but are not limited to, evaluating the performance of current LWR internals during long term operation, and investigation of irradiation-induced property changes of new candidate materials for reactor internal and accident tolerant fuel applications.

LWR internals will experience high irradiation doses at certain locations during long-term operation. In-pile and test reactor irradiation is characterized by a relatively low rate of damage accumulation and there is very limited data on the neutron irradiation-induced microstructural evolution of reactor internals at the doses that reactor internals will experience after their first and second license renewals. Many of the models used to assess the implications of microstructural evolution are highly empirical and thus regulatory requirements often do not permit extrapolation beyond the range fully validated with experimental and in-service observations. As an increasing number of first and second license renewals are pursued, it is desirable to understand potential license renewal safety questions before in-service observations are available. Consequently, there is a need to establish a rapid and cost effective approach to reach elevated doses and provide insight into the neutron irradiation-induced microstructural evolution occurring at elevated doses. Ion irradiation studies have the potential to provide such insight, since very high doses can be achieved in a short timeframe. In addition, ion irradiation can be used to increase the level of irradiation damage of samples fabricated from in-service reactor components to elevated doses. Finally, ion irradiations offer a highly cost-effective alternative to in-service or test reactor irradiation because the experimental specimens can often be handled, examined, and transported by normal means. One continual challenge for both test reactor and ion beam irradiations is to develop improved mechanistic understanding of the effect of dose rate on microstructural evolution.

Many have proposed the development and use of physics-based or mechanistic models to provide greater insight into the implications of microstructural evolution at higher doses than available from existing experimental observations. Physics-based or mechanistic models require extensive validation to increase their predictive capabilities for high dose microstructure evolution. Mechanistic understanding of microstructure evolution is one area where ion beam irradiation provides advantages relative to in-pile testing. In addition to the cost and time advantages discussed above, ion beams may be installed together with in situ measurement and imaging capabilities. This combination is invaluable in the pursuit of a mechanistic understanding of microstructure evolution because they allow for the capture of a data continuum rather than just the initial and end state.

Furthermore, ion irradiation can help identify candidate materials for structural applications in LWR reactor internals that have increased resistance to irradiation-induced degradation compared to currently used materials. Ion irradiation can be used in screening studies to down select from a wide variety of candidate materials for a more cost effective and efficient in-pile irradiation campaign. Similarly, ion irradiation can be used to evaluate various accident tolerant fuel concepts and improve the mechanistic understanding of irradiation-induced microstructural evolution of these materials under specific irradiation conditions.

4 Application to the Advancement and Implementation of Nuclear Energy Technologies: Gaps and Needs

4.1 Timelines for Deployment of Advanced Nuclear Concepts and the Critical Role of Ion Beams

Along with worldwide federal government investments, a set of privately funded companies is working to commercialize advanced nuclear concepts [45]. In the publicly available plans for some of these companies, they are looking to commercialize their concepts as early as the 2020s. A common thread through all of their plans is the importance of materials of construction. Some companies that need to demonstrate advanced materials will be adequate to commercialize their design while others are purposefully engineering around materials challenges to get a first generation product to market quickly. In any case, materials decisions are foundational to their plans and the ability of the research and development community to deliver advances in materials performance quickly is important.

Enabling innovation in nuclear products, specifically moving from concept to commercial product, requires answering key technical questions to prove viability of an idea (sometimes referred to as the “first valley of death”) and then turning a full scale demonstration into a commercial product (sometimes known as the “second valley of death”). Because innovative nuclear products often require an advance in material performance, ensuring adequate response of a material to irradiation could be critical to both of these phases. Ion beams can play an important role on the path to a deployed product.

In many instances, proving a material is acceptable to a licensing authority is a critical step in a commercial deployment. Simply stated, the licensing authority needs proof that a material’s performance will be adequate to maintain the required safety functions. This could come from extensive testing or from a validated model in which the regulator has confidence.

Ion beams can provide critical data in developing validated models. This validation can be in the form of proving the model’s predictive capability across a wide range of conditions or by providing a cost effective way to generate large data sets to understand material’s variability of performance under irradiation.

Ultimately, these validated models of material’s performance rely to some extent on prototypic data from in-reactor testing. These tests are expensive and need to be thoughtfully planned. Ion beam testing can provide guidance to allow for the best use of expensive in-reactor testing. This might include extrapolating known material’s performance to help justify extended operation while waiting for further in-reactor irradiation data.

Supporting commercial deployment is important for companies with an existing product, but maintaining a vibrant nuclear industry also requires a portion of the research portfolio be dedicated to new ideas, some of which will become the next generation of commercial products. These research programs are the foundations for future development. To create new materials that have superior performance under irradiation, requires a means to cost effectively test them.

Ion beams can be critical to these foundational development programs in providing low cost screening of materials, validating separate effects models, and creating the knowledge needed to design for radiation resistance.

4.2 Value of Ion Beams in Meeting Specific Program Needs

Key nuclear components are developed through a progressive series of technology readiness levels [46] or maturation phases, and ion beam irradiation can play an important role in advancing materials from proof-of-concept through proof-of-principle to proof-of-performance. In the proof-of-concept phase, a materials designer will have a basic idea that needs clarity, for instance “is a particular precipitate uniquely stable under irradiation?” for a structural component whose mechanical properties rely on maintaining a certain distribution of precipitates. Ion irradiations can be used to rapidly screen these specific engineered microstructures (including evaluation of spectral and dose rate dependencies) before costly development work is performed. These screening tests provide information that can be used to optimize the engineered microstructure so that eventual neutron irradiation testing is performed on only the subset of materials that are predicted to have superior performance.

The proof-of-principle phase involves compiling basic properties and establishing a design basis for materials. Small-scale testing and ion irradiation are useful for measuring properties and can establish an envelope of conditions that define how a material can be used. By comparing microstructures, it will be possible to establish the conditions where certain phenomena dominate the material performance. For example, it is known that irradiation-induced precipitates form in steels at certain temperatures. Ion irradiation could be used to predict the temperature, flux and fluence conditions for which the precipitates will form for specific alloy compositions [47]. These techniques can be used to estimate material properties for preliminary designs and will support design iterations while neutron testing is performed. In addition, these estimates can be used to support determination of the test conditions required for neutron irradiations.

In the proof-of-performance phase, ion irradiation can be used for validation of material performance codes. The ion irradiation technique can be used to measure certain fundamental properties that assist with validation and can be used as input to the codes. Ion irradiation can also be used to characterize the microstructure after exposure to off-nominal conditions that cannot be assessed during standard reactor testing. High-temperature transients are predicted during accidents that will cause rapid recovery of material properties. Ion irradiation can be used to generate many samples with similar microstructures to neutron-irradiated materials. These samples will have lower activity and can be more easily handled for additional testing. Subsequent annealing studies can be performed at extremely high heating rates that can be used to map out a region of design space where little data exists. Combined with the ability to reach extremely high dpa, this will permit better prediction of material performance during high-temperature transients and reduce operational conditions that require extrapolation of material properties. The high-throughput provided by ion irradiation will provide more data for better statistics and reduce the required margins due to uncertainty in the material property measurements. Performing fewer tests on selected materials under more closely defined conditions will also reduce the costs associated with the reactor testing required to demonstrate proof-of-performance.

Example 1: Designing and Developing Radiation-Resistant Materials

Designing, developing and qualifying new reactor materials and fuel systems is traditionally resource and time intensive. New approaches making maximum use of ion irradiation and computational tools with subsequent confirmatory in-reactor testing of a small number of the most promising options can not only reduce the time and cost of initial development, but also process scale-up and material qualification.

Advanced reactor materials, fuels and waste forms are complex, often consisting of multi-components, multi-phases, and hierarchical microstructures. It is possible that subtle differences in microstructures from variations in processing parameters could lead to significantly different irradiation performance. The multitude of interactions between material constituents and irradiation-induced defects and their nonlinear behavior introduces enormous complexity in the discovery process. The extensive parameter space to be explored prohibits the traditional trial and error approach and a large campaign of expensive reactor irradiation testing. Ion irradiation can be an effective way for systematic exploration of the parameter space, significantly reducing the cost and development time. For example, a comprehensive survey of model and commercial alloys can be conducted using ion irradiation techniques to determine the dependence of radiation resistance on major and minor alloying element contents and thermo-mechanical treatments. These data can provide critical input and direction to the selection of candidate materials for specific applications. In concert with integrated computational materials engineering design of new high performance structural alloys, this screening process will significantly shorten the material development cycle.

The acceptance of a new material in a new reactor concept, particularly in a high-dose environment, can be a taunting task. The behavior of materials at these high doses is unknown and difficult or impossible to explore in a timely fashion. The high dose rates achievable through ion irradiation can shorten the time of high-dose irradiation from years in a reactor to days. A viable approach is to take high dose data from accelerated ion irradiation experiments and scale them with computer models to predict material responses in the high-dose regime of reactor environment. The reliable extrapolation of low-dose neutron irradiation data and accurate prediction of high dose behavior requires a thorough, quantitative understanding of effects of both irradiation and metallurgical conditions on time evolution of complex defect structures, which again can benefit from detailed mechanism studies using ion irradiation techniques. There is a distinct advantage by combining ex situ and in situ ion irradiation, particularly for a fundamental understanding of high-dose behavior of a material. Ex situ ion irradiation of a specimen with prior to or subsequent in situ ion irradiation of the same specimen in a microscope allows detailed and systematic studies of defect evolution and interactions at different stage of irradiation, and reaches the high-dose state within a reasonable time.

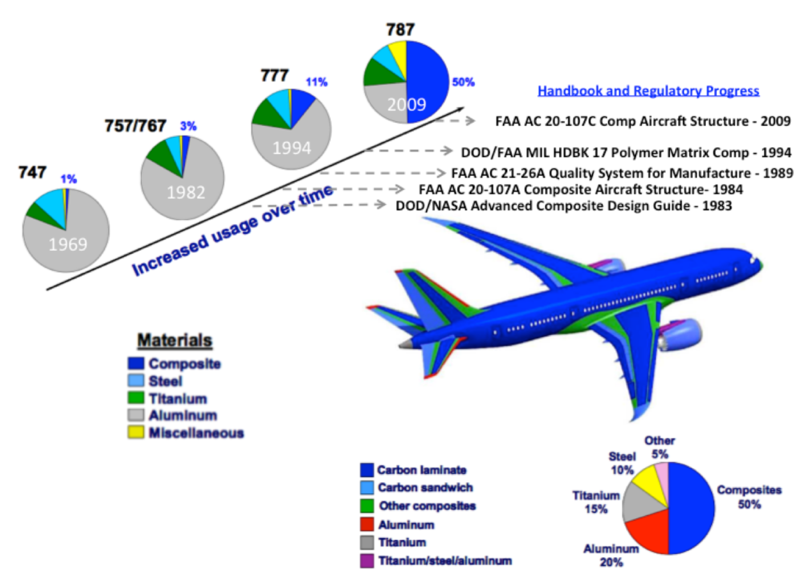
Developing next-generation materials with superior radiation resistance requires new material design concepts and processing methods. A fundamental understanding of radiation resistance provided by a particular concept is essential in the early stage of development. Ion irradiation, when combined with in situ high-resolution characterization techniques such as electron or X-ray beams is invaluable for understanding the underlying science of radiation damage production and defect evolution. For example, recent in situ ion irradiation work has shown that deliberate introduction of nanovoids in conjunction with nanotwins improves radiation damage tolerance in metallic materials [48]. This result and similar innovative strategies for manipulating nanoscale structures suggest multiple pathways are available for designing highly radiation-resistant materials. For example, mapping of the temperature-dependent critical dose for amorphization through well-controlled in situ ion irradiation experiments has led to optimization of chemistry and structure of radiation-resistant waste storage materials [49].

Example 2: Modeling and Predicting Materials Performance in Reactor Environments

Computer simulations of physical processes have been increasingly used in the development of new materials and the safety analysis and design of fuel systems and components in nuclear energy systems. Because of the impact modeling and simulation can have, accurate, credible computer models are needed for both safe operations of current nuclear power plants and future growth of nuclear energy. One of the most important roles of ion beams is to provide validation data to computer models and thereby test model predictions. The seamless integration of modeling and simulations with interactive ion and neutron irradiation experiments will enable

MODERN MATERIALS DEVELOPMENT

Nuclear power has yet to realize the opportunities available through the application of modern materials and methodologies of modern manufacturing and design (S. J. Zinkle, K. A. Terrani, L. L. Snead, 2016, Current Opinions in Solid State & Materials Science, Vol. 20, pp. 401-410). In comparison to other advanced energy or transportation industries, nuclear has been very slow to adopt both the tools and materials for a new generation of safer, higher-performance systems. As example of a safety-conscious approach to maturing the technology readiness of materials and systems, a study of the introduction and deep penetration of composite technology into commercial aviation is instructive. In this case, despite intense regulatory oversight and high attention to public safety, commercial aircraft have evolved from being constructed largely from metallic alloys (75% aluminum, 1% composite) in the 1960s to a majority composite in the current Boeing 787 (50% composite), with significant performance improvements (W. G. Roeseler, B. Sarh, M. U. Kismarton, 2007, “Composite Structures: the first 100 years, in: K. Kageyama, T. Ishikawa, N. Takeda, M. Hojo, S. Sugimoto, T. Ogawawara (Eds.) 16th International Conference on Composite Materials. Kyoto, Japan). These improvements were made possible by a sustained effort of materials development strongly coupled with a wide range of modeling, codes and standards development.



accelerated material development, validation and qualification and ensure safe, prolonged operation of aging nuclear reactors.

Highly controlled ion irradiation experiments can provide high-fidelity data to benchmark computer models of materials' microstructure and performance. Measurements at a series of irradiation temperature, dose and dose rate and of materials with a wide range of metallurgical variables are critical to the calibration of model parameters and testing of their internal consistency to changes to both irradiation and metallurgical conditions.

Ion irradiation with in situ characterization techniques allows the access to the full history of the kinetic development of microstructure under irradiation, providing critical input to models at relevant length and time scales. For example, in situ TEM of heavy ion irradiation damage at the liquid helium temperature can reveal "quenched-in" cascade damage that occurs over picosecond time scales, providing valuable data for models of cascade phenomena, such as single cascade events, cascade – cascade or cascade – sub-cascade interactions, cascade defect production and annihilation. As defects move, dissolve, appear, coalesce and annihilate at elevated temperatures, a complete picture of the actual fate of defects can be formed, and critical physical parameters (e.g., migration energies, trapping for various types and sizes of defect clusters) can be determined to effectively constrain the development of models of microstructural evolution. Accurate, reliable prediction of neutron damage in a nuclear reactor can be made by a coordinated approach of computer modeling and in situ ion irradiation experiments with TEM.

The damage processes in real reactor environments revealed by computer models often involve combined effects of irradiation, temperature, stress and corrosive media. Coupling in situ ion irradiation with in situ stages for heating, cooling, straining and environmental cells enables studies of synergistic effects of irradiation, temperature, stress and corrosion in relevant reactor environments that cannot be achieved through traditional post-irradiation examinations. This is important for

STRUCTURAL MATERIALS DEVELOPMENT

For the sodium-cooled fast reactors (SFRs), alloy NF-709 was on the priority list of austenitic stainless steels and its optimized version A709 alloy was down selected for further assessment. As the core support structural material, A709 alloy is expected to receive a dose of 20 dpa or more. However, to date, there is only one published neutron irradiation data set from the as-annealed NF709 irradiated to 3 dpa at 500°C in the Advanced Test Reactor, and researchers at Oak Ridge National Laboratory are currently examining samples irradiated to 6 dpa. Unfortunately, there are no other neutron-irradiated samples in existence or currently under irradiation. For timely material qualification and to be cost effective, the evaluation of irradiation performance to high dose for A709 alloy has to be supported through ion irradiations. The microstructural changes over the low dose ion condition can be benchmarked against the 3 and 6 dpa neutron irradiation results. Together with the newly developed simulation and modeling tools and extrapolated microstructural evolution expected at high irradiation doses, the material performance of NF709 or A709 at anticipated SFR related conditions can be accurately predicted. One of the major concerns for the A709 alloy is the irradiation stability of specially tailored precipitates. The ion irradiation study does not require a large irradiated volume of material and the precipitate stability can easily be evaluated using ion beam irradiations over a wide range of temperature and dose conditions. Modeling dose rate effects on precipitate stability and evolution (incorporating ballistic dissolution, radiation enhanced diffusion, radiation induced solute segregation effects) will be a crucial step in order to predict the corresponding precipitate stability behavior under representative SFR dose rate verifying the physical phenomena predicted by computer simulations. The influence of transmutation products, particularly production of inert gases such as helium, can be studied by conducting irradiations under He-free, He pre-injected or continuous production of He test conditions during irradiation in a dual-beam facility to understand separate and combined effects of cascade damage production and He implantation.

4.3 Existing and Needed Ion Beam Capabilities

There are numerous ion-beam irradiation facilities at government, university, and industry laboratories around the world (see Section 2 of this report) [2, 50, 51, 52, 53, 54]. Most of them can provide single ion beams, while some include dual and triple beam capabilities [51, 52] that allow simultaneous irradiation with several species to simulate (1) a combination of atomic displacements, helium and hydrogen production in reactor environment, or (2) multi-radionuclide decay effects (such as the α particle and the α recoil nucleus that are produced during α decay). These multibeam facilities also allow in situ ion beam characterization (channeling, Rutherford backscattering, elastic recoil detection analysis, nuclear reaction analysis, particle-induced x-ray emission, etc.) or electron beam characterization (TEM) [54]. In situ corrosion and mechanical testing capabilities are also integrated in some ion beam facilities [52] to combine environment, property, and radiation conditions.

There are currently 13 facilities that combine transmission electron microscopy with in situ ion irradiation in operation around the world. Two currently operate in the U.S. The IVEM-TUF at the ANL features high-quality TEM interfaced to an ion accelerator with high incident angle of the ion beam permitting continuous observations and data recording at the time resolution of 5 ms under most sample tilting conditions, which is extremely important for dynamic studies of defect evolution. The cooling, heating and straining stages allow samples to be held at the controlled temperature between -253°C (20 K) to 1,300°C and under load up to 400°C. A dual beam capability is being added for studies of synergistic effects of heavy ion induced cascade damage and helium implantation. The Ion Beam Laboratory at Sandia National Laboratories offers seven ion accelerators for multitude of irradiation/implantation experiments. This includes the in-situ ion irradiation TEM (i3TEM) facility that permits

FUEL MATERIALS DEVELOPMENT

Another application example that highlights the value of ion beam irradiation is the development of a mechanistic understanding of nuclear fuel behavior under irradiation, specifically, using dual beams of MeV Kr and swift heavy ion irradiations to address the high burn up structure (HBS) formation mechanism. There have been several explanations for the onset of HBS, including grain refinement due to the irradiation-induced dislocation self-reorganizations, gas bubble-induced sub-grain formation, and new structure formation resulting from the fission fragments inducing non-uniform stress fields. However, there is no consensus on which mechanism is responsible for HBS formation. The experimental investigation of HBS in commercial LWR fuel is challenging due to the very limited restructured fuel occurring at typical operational burn-up levels and the related complexity in working on high burnup fuels. The dual-beam irradiation offers all three effects including irradiation-induced dislocation, fission gas and void swelling, and radiation from energetic fission fragments. The different combinations of those Kr and swift heavy ion irradiations can help to map out the HBS formation mechanisms, and the related physical models can be built. The experimental microstructural data of irradiated fuels and related physical models can then be used for improving the multi-scale modeling (MARMOT) and validations.

direct real-time observation of triple beam studies (helium, deuterium, and heavy ions) over a range of tilts and environments (including 15 TEM stages).

Future enhancements beyond those available at the Argonne and Sandia facilities may include analytical tools for analysis of changes in local elemental concentrations in complex alloys as reflected in precipitation, dissolution, segregation and second phase formation processes under irradiation. For instance, energy-filtering TEM will not only allow imaging chemically distinct phases and two-dimensional spatial mapping of elemental composition and distribution in the sample, but also improve contrast/resolution for conventional TEM imaging and diffraction of thicker foils, and accurate determination of the local foil thickness of the sample. Integrated with

WASTE FORM DEVELOPMENT

High radiation stability is one of the requirements for nuclear waste form development. The science challenges of radiation damage associated with nuclear waste materials are somewhat similar to those of nuclear fuel and structure materials research. However, there are several key differences. The primary differences are (1) the extraordinary time scale for nuclear waste storage: not years or decades, as for nuclear fuel and core internal structure lifetimes, but thousands of years; (2) various radiation sources in nuclear waste. The principal sources of radiation in high-level wastes (HLW) are β decay of the fission products (e.g., ^{137}Cs [Caesium] and ^{90}Sr [Strontium]) and α decay of the actinide elements (e.g., U, Np, Pu, Am, and Cm). β decay produces energetic β particles (~0.5 MeV), low-energy recoil nuclei, and γ-rays; whereas α decay produces energetic α particles (4.5–5.5 MeV), energetic recoil nuclei (70–100 keV), and some γ-rays. In general, β decay of the short-lived fission products is the primary source of radiation (and heat generation) from HLW during the first 600 years of storage. Because of the long half-lives of the actinides and their daughter products, α decay is dominant at longer times. Radiation effects include ionization and electronic excitation, ballistic processes/atomic displacement, and transmutations and gas production.

The effects of radiation from the decay of radionuclides will accumulate over very long periods; consequently, a broad range of accelerated irradiation techniques must be utilized to study radiation damage effects in nuclear wastes. These irradiation techniques and procedures include: (1) short-lived actinides-incorporation, (2) actinides in natural minerals, (3) gamma irradiation utilizing ^{60}Co (Cobalt) or ^{137}Cs sources, (4) neutron irradiation, and (5) charged-particle irradiation using electrons, protons, α-particles, and heavy ions. Experimental facilities that can accelerate the radiation processes involved and allow observation of the complex evolution of the waste are very desirable. Generally, ion beam irradiation using particle accelerators are a more cost-effective alternative to study radiation damage in materials in a rather short period, allowing researchers to gain fundamental insights into the damage processes and to estimate the property changes due to irradiation. α particle irradiation using particle accelerators, is an effective tool for understanding α particle effects; similarly, heavy-ion (e.g., Xe [Xenon]) irradiation is an effective method to study α recoil effects. Finally, electron irradiation can be used to study the effects of ionization and electronic excitations from β particles and γ rays on nuclear waste materials. Advanced ion beam irradiation experiments performed at multiple damage rates coupled with start-of-art characterization techniques will be important in all the areas outlined for nuclear waste research: phase instability due to transmutation, helium accumulation and bubble formation, volume expansion, increase in chemical reactivity and decrease in durability, phase separation associated with recoil cascades, increased diffusivity and transport of minority species and precipitates, accumulation of stored energy, and radiation-induced amorphization.

diffraction-contrast defect imaging and in situ ion irradiation, evolution of irradiation-induced defects, phases and chemical segregation can be monitored in real time during irradiation. Three-dimensional tomographic imaging of irradiation-induced structural changes and defects enabled by TEM tomography stages will add extreme values to study spatial distributions of defects and roles of interfaces in the thin foil sample. Efforts have also been made to develop in situ nano-indentation and corrosion capabilities. Next-generation in situ ion irradiation capabilities may involve ultrafast, high-resolution TEM interfaced with ion accelerators for studies of cascades at the time resolution of pico-seconds, and point defect and interface interactions and nucleation processes at the time and spatial resolutions not currently achievable.

Another future enhancement will be the combination of synchrotron X-ray beams with ion accelerators. This combination will open up enormous new opportunities for probing irradiation-induced nano- and meso-scale defects, phase transformation and stability, quantitative measurements of the local stress-strain response, and the role of internal and external stresses in the evolution of microstructure under irradiation, permitting direct correlation between microstructure and property at the microstructural and mesoscale scales. Well-controlled specimen environment (temperature, stress, gas or liquid) and simultaneous measurements with multiple probes are also of great interest for future in situ ion irradiation studies. Computational modelers are frequently required to decipher complicated and path-dependent material processes from very few data points for macroscopic materials properties at the end of long irradiation exposures. In situ observation and continuous monitoring of radiation effects on materials will provide a basis for code validation. Synchrotron techniques, including diffraction, spectroscopy and imaging will provide unprecedented detail for observing crystallographic and mesoscale microstructural changes that occur in materials due to irradiation damage. The irradiations can be combined with additional in situ techniques, such as mechanical straining or temperature to capture the influence of competing mechanisms on microstructural changes such as thermal and irradiation creep. X-rays are inherently highly penetrating and synchrotron techniques can measure multigrain samples representative of bulk materials. The interactions of defects with surfaces and grain boundaries will also be more representative of bulk materials than is possible with current in situ ion and electron beam characterization techniques.

Finally, an expanded use of available higher energy ion beams and the exploitation of new compact cyclotron technology to produce intermediate-to-high energy ion beams is suggested from which engineering data can be obtained from ion studies and used to validate modern modeling tools.

5 Path Forward

5.1 Development and Deployment Scenarios

The objective of ion irradiation is to serve as a useful, and perhaps, necessary tool to provide predictive data to industry for operating plants, and to designers for future reactor concepts, as well as ongoing material screening and fundamental radiation effects studies in order to develop improved radiation effects models for materials.

5.1.1 Perspectives on Prospects for Development and Deployment

The demonstrated capability of ion irradiation to produce microstructures, mechanical properties, stress corrosion cracking behavior and corrosion response that are very similar to those experienced in-reactor establishes the technique as a viable method for predicting the behavior of materials under irradiation in a reactor environment. The necessary accelerator technology to obtain precisely controlled and reproducible irradiation conditions is mature and widely available at multiple research institutions. While continued development is needed to fully identify the regimes in which ion irradiation can be profitably exploited, deployment of the technique to problems on which it has been extensively benchmarked may begin immediately. These include the prediction of radiation induced segregation, irradiated microstructure, irradiation hardening, and IASCC initiation in austenitic alloys exposed to boiling water reactor or pressurized water reactor core conditions. These are all essentially "low temperature, low dpa" processes in that they evolve quickly and tend to saturate above a damage level of 5–10 dpa. Hence, they can be probed using proton irradiation, for which the larger penetration depth is useful to achieve measures of mechanical properties and stress corrosion cracking behavior. Useful data from proton or heavy ion irradiation can also be acquired to predict the likelihood of failure of key core components such as baffle former bolts, instrument tubes, springs, etc. that are exposed to higher dose levels, and to predict the potential impact of operational changes such as power uprates (slightly higher operating temperatures and irradiation fluxes) on material performance.

Given the capability of today's accelerators and ion sources, the application of ion irradiation to the case of high dpa in LWRs or for core components in fast reactors requires irradiation with heavy or self-ions to reach the >100 dpa regime in reasonable times. This technique results in much higher damage rates and so the acceleration factor over the reactor case is greater, requiring more adjustments to the irradiation conditions to mimic the irradiated microstructure. The impact of changes in damage rate on radiation-induced property changes associated with neutron and ion irradiation has been reported to range from minor to very significant, depending on the specific material and irradiation conditions. By virtue of the much shallower penetration depth, mechanical property assessment is more difficult for heavy ion irradiations, though not impossible. While microstructure characterization in the irradiated region is easily achievable, advances need to be made in extracting strength information from a roughly 1 μm layer that can be used to accurately predict tensile data from a neutron irradiated tensile sample.

To further enhance the value of ion irradiations for understanding and quantifying neutron irradiation effects in materials, it would be beneficial to expand the number of facilities with in situ capabilities to apply reactor-relevant operating environment conditions such as applied stress and contact with flowing coolants during ion irradiation. Anticipated continued development of nanoscale property measurement tools (mechanical properties, thermal properties, etc.) would also enhance the quantitative predictive value of ion irradiation studies in the future.

5.1.2 Cost-Benefit Analysis

A detailed cost-benefit analysis is beyond the scope of this report; however, the allure of ion irradiation includes its speed to achieve desired irradiation doses and avoidance of induced radioactivity, both of which are responsible for a truly enormous cost savings. As an example, a self-ion irradiation of a sample to a damage level of 200 dpa requires roughly 4–5 days of irradiation at a cost of about $10,000 in current state of the art ion irradiation facilities. Several samples can be irradiated simultaneously and the irradiated volume is sufficient to provide enough material for a complete microstructure characterization as well as nano-mechanical property experiments. The cost of full characterization is approximately $10,000/sample, not including staff time. So for $20,000, a full characterization of microstructure and nano-mechanical properties can be conducted. The entire iteration consisting of sample preparation, ion irradiation, and characterization can be comfortably completed in 2–3 months.

For comparison, consider irradiation of the same material in a fast test reactor. A damage level of 200 dpa will require a minimum of 10 years of irradiation. The cost for this irradiation will be in excess of $1M, although several hundred TEM specimens and dozens of mechanical property specimens could typically be accommodated in a single irradiation capsule. Due to residual radioactivity, a cooling time of approximately one year will be required. Characterization will require a hot lab for sample preparation and will require ~$100,000 just for unpacking and cataloguing samples from a single capsule. Characterization will cost another $20,000 per sample. Additional expenses associated with shipping, authorization, etc. are likely to cost several $100K. So the comparison is ~$20,000 for ion irradiated samples for which the data is available within 2–3 months after the irradiation vs. >$200,000/sample for which the data is available 11–12 years from the start of the irradiation. In 12 years, ~50 iterations using ion irradiation could have been conducted to systematically explore effects of dose, temperature, dose rate, etc., including the possibility to rapidly pivot toward investigating a different alloy system if unfavorable radiation effects behavior is discovered during the initial irradiation experiments. In summary, for ion irradiations the cost advantage per microstructural sample is on the order of a factor of >10, the time advantage is on the order of 50 and the rate of progress in moving the field forward is immeasurable.

5.2 Barriers to Implementation

Several barriers currently exist that inhibit the full implementation of ion irradiation research to accelerate the development of nuclear energy materials and technologies.

Delineation of the conditions under which ion irradiation can be used to reliably predict reactor irradiation behavior of materials for providing useful data to industry and designers is clearly a hurdle for the technique. In particular, improved understanding of dose rate effects, as a function of temperature, will require improvements in theory and modeling and simulation (going beyond simplistic Frenkel pair rate theory calculations) in concert with analysis of neutron and ion irradiation experimental data. Ion irradiation has well-recognized limitations, and identifying the regimes in which the technique is valid and useful for reliable quantitative prediction of radiation effects in a reactor environment is a challenge. For example, it may be that a specific ion irradiation condition (dose rate and temperature) is identified that accurately captures dislocation loop and void evolution in reactor, but not radiation induced segregation. That is, the complete reactor-relevant microstructure may not be reproducible under ion irradiation. However, if the processes are only weakly dependent on each other, then ion irradiation can be used to understand and predict loop and void evolution in reactor with confidence. Other irradiation conditions can then be used to capture the evolution of radiation induced segregation.

A long-range goal is to rigorously establish science-based linkages between microstructural data and material properties for both bulk neutron and ion irradiation conditions. This will require development of improved modeling and simulation tools to predict the material properties for a given microstructure, with extensive experimental validation over a wide range of relevant materials and operating conditions. In the near term, utilization of ion irradiation results and advanced modeling and simulation to predict incremental future behavior of reactor-irradiated materials would be a valuable first step to establishing the quantitative accuracy of microstructure-based irradiated material property models. Such successful demonstrations would facilitate the future acceptance of microstructure-based model predictions by regulatory authorities rather than continued sole reliance on empirical bulk neutron irradiation test data and correlations.

Ion irradiation facility upgrades to support broader capabilities for in situ testing and characterization are another barrier (financial) to full development of ion irradiation as a predictive tool for nuclear materials and technology development. Since the value of in situ testing at reactor-relevant operating conditions (applied stress, flowing coolant, etc.) has already been demonstrated in a few limited cases, it would be beneficial to expand this capability to additional facilities and/or other important unique in situ test configurations.

In order to most efficiently harness the current ion irradiation facility capabilities, it will be important for the ion irradiation user community to adopt and embrace operational "best practices" guidelines and, where relevant, round-robin experimental campaigns such as those recently organized by the International Atomic Energy Agency on specific materials. Such guideline adoption and user community activities will foster increased demonstrated confidence in the reliability and reproducibility of data generated from individual ion beam facilities, and should accelerate the acceptance of ion irradiation data in nuclear reactor regulation, design and licensing processes as well as providing potentially improved in efficiencies fundamental radiation effects research and screening of candidate reactor materials.

5.3 Regulatory Challenges

Acceptance of ion irradiation data for reactor licensing purposes is perhaps the biggest challenge. To be a truly useful technique, the data must be accepted by the regulator for use in predicting behavior of materials in reactor. Ion irradiation can be an excellent tool for separate effects testing to isolate behavior that can be convoluted by multiple simultaneous effects within a reactor. In fact, the limited capability to control individual irradiation parameters in reactors make these irradiations less than ideal for determining the effects of such parameters on the irradiated microstructure and properties. However, regulatory requirements often specify that data collected in-pile is necessary to build and validate material property models and correlations. This requirement ensures that material property models and correlations account for the effects of neutron irradiation and other associated environmental conditions within the reactor. In order to utilize ion irradiation data in a regulatory application, a technical basis would need to demonstrate the correlation between ion irradiation data and in-reactor material performance for the particular property or phenomenon of interest. Additionally, for safety-related applications, extrapolation of data beyond the range where representative experiments and observations are available requires an accepted technical basis. Therefore, this technical basis would likely need some form of in-reactor data to support the entire range of applicability for the model or correlation.

Staff from the NRC has recently presented an evaluation of strategies for obtaining high-fluence materials data, including consideration of ion irradiation techniques [55]. The authors cited some technical considerations that arise when applying ion irradiation data to predict neutron irradiation behavior in reactor applications. As examples, they write, “selecting the proper irradiation (temperature to accurately represent temperature sensitive mechanisms such as void swelling) may be complicated by differences between ion and neutron irradiation” and that “neutron irradiation damage depths are larger by at least three orders of magnitude in comparison to ion irradiation.” This second point refers to the challenge of extracting mechanical property information from a thin ion irradiated layer that agrees with the performance of the bulk material. These are two examples of issues that would require an articulated technical basis to demonstrate the correlation between ion irradiation data and neutron irradiation and in-reactor performance.

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