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Radiation Damage from Atomic to Meso-Scales in Extreme Environments

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Abstract. Overcoming radiation damage degradation is a key rate-controlling step in fusion materials development. New science, approaches, and facilities are needed at multiple scales. Understanding at the atomic scale the behavior of materials subject to extreme radiation doses and mechanical stress in order to synthesize new materials that can tolerate such conditions is needed. Current computational and experimental research into radiation-tolerant materials will be described. Plans for future irradiation facilities and science are also described. The Matter Radiation Interactions in Extremes (MaRIE) concept is a National User Facility to realize the vision of 21st century materials research and development. The Fission and Fusion Materials Facility (F^3) segment of MaRIE proposes to use the present proton linac at Los Alamos with a power upgrade to drive a spallation neutron source that can provide the required radiation environment. Importantly, F^3 would also provide the capability for *in-situ* measurements of transient radiation damage, using unique x-ray and charged particle radiography diagnostics. Coupled with integrated synthesis and characterization capability, these would reveal not only the atomic scale origins of radiation effects but also their meso-scale consequences. The mission of the facility would be to study material science in radiation extreme environments to provide predictive capability of material performance to allow certification of new nuclear systems. It is not intended for large-volume component testing. The radiation environment possible will be described.

1. Introduction

A foreboding materials challenge is to be able to withstand the 10–15 MW-year/m^2 neutron and heat fluence expected in the first wall and blanket structural materials of a fusion reactor.[1] Overcoming radiation damage degradation is a key rate-controlling step in fusion materials development. New science, approaches, and facilities are needed at multiple scales.

The Grand Challenge of materials science, as identified by a series of United States Office of Basic Energy Sciences workshops, is to transition from the present era of observation of material properties to an era of control of the functionality of materials by design.[2] This paper describes both current efforts at observation of radiation-tolerant materials and what makes them that way, through advanced modeling that identifies new mechanisms for radiation tolerance that can be used, to plans for future irradiation science facilities and experiments that can lead to the necessary control of materials needed to make fusion power a reality.

2. Radiation damage science from observation to control

Ferritic-martensitic steels provide attractive alternatives to the use of austenitic stainless steels for fission, fusion, spallation and fast reactor applications. This is in part a consequence of their generally greater resistance to neutron irradiation-induced swelling and creep and their reasonably acceptable high temperature strength[3], although these attributes are somewhat sensitive to alloy composition and processing history.

The ferritic-martensitic alloy HT9 has been investigated in the USA for application to fast, fusion and spallation neutron driven devices. This alloy has shown great promise, especially with respect to its resistance to swelling and excellent compatibility with sodium. To date, however, all reported swelling and creep data on this alloy have been derived from very simple, free-standing experimental specimens irradiated under rather well-defined conditions in material test assemblies[4]. Components of long-lived fuel assemblies are more complex in shape and will experience progressive changes in environmental conditions.

It is known that the swelling of austenitic stainless steels is sensitive to "history effects" such that materials subjected to reactor-relevant changes in neutron flux, temperature and stress state often reach much higher swelling levels than is reached in identical material irradiated under constant irradiation conditions[5]. To explore whether the resistance of HT9 to void swelling is maintained under more realistic operating conditions, the radiation-induced microstructure of an HT9 ferritic/martensitic hexagonal duct was examined following a six-year irradiation of a fuel assembly in the Fast Flux Test Reactor Facility (FFTF).[6] The calculated irradiation exposure and average operating temperature of the duct at the location examined were ~155 dpa at ~443°C. It was found that dislocation networks were composed of predominantly a/2<111> Burgers vectors. Surprisingly, for such a large irradiation dose, type a<100> interstitial loops were observed. Additionally, a high density of precipitation occurred. These two microstructural characteristics may have contributed to the rather low swelling level of 0.3%.[6] Based on this study using post-irradiation examination only, it can not be determined whether the transient regime of swelling is still in progress or the steady-state swelling rate is being approached.

The grand challenge of materials science is to transform from the era of observation (that HT9 is radiation resistant) to the era of control of functionality where we can design the material functions that are needed [2]. Designing nuclear materials that can sustain extreme amounts of damage is an important challenge for future nuclear reactors. During service, radiation-induced point defects (interstitials and vacancies) are created [7], which can aggregate to form interstitial clusters, stacking fault tetrahedra, and voids [8]. Eventually, this can lead to swelling, hardening, amorphization, and embrittlement, which are primary causes of material failure [9]. It has been speculated that grain boundaries (GBs) and interfaces may serve as effective sinks for defects. Although grain boundaries can serve as effective sinks for radiation-induced defects such as interstitials and vacancies, the atomistic mechanisms leading to this enhanced tolerance are still not well understood. Recently, several nanocrystalline (NC) materials containing a large fraction of GBs have been shown to have improved radiation resistance compared with their polycrystalline counterparts. These experimental results are consistent with the expectation that GBs absorb defects, but how defects interact with GBs at the atomic scale is far from understood. In particular, the temperatures in some irradiation experiments are so low (for instance, 100 °K [10]) that vacancies are essentially immobile [a vacancy in gold with a barrier of 0.6 eV moves one nearest neighbor distance (2.9 Å) on a time scale of 10^{19} s at 100 °K], but their annihilation is still somehow enhanced by GBs [10,11]. Thus, there must be an alternative mechanism assisting the annihilation of vacancies within grain interiors.

One of several new Energy Frontier Research Centers funded by the DOE's Office of Basic Energy Sciences is one Los Alamos leads named the "Center for Materials at Irradiation and Mechanical Extremes." Its objective is to understand, at the atomic scale, the behavior of materials subject to extreme radiation doses and mechanical stress in order to synthesize new materials that can tolerate such conditions. The understanding of fundamental unit mechanisms responsible for processes such as Frenkel pair recombination, defect clustering and precipitate nucleation, point defect interactions with interfaces, and dislocation core spreading are critical to the prediction and control needed for the bottom-up design of materials with unprecedented tolerance to extreme irradiation environments. The importance of interface structure in influencing these unit mechanisms operating at very small spatial scales cannot be over emphasized. Early work studying the interaction between point defects and grain boundaries (homophase interfaces) [12] confirm qualitatively that the response of different grain boundaries to irradiation and impurity concentrations depends on interface structure [13] and the presence or absence of coherency. However, because experimental and modeling methods were limited, the connections between modeling and experiments were necessarily qualitative in nature. The dependence of interface response to atomistic and collective processes occurring at extreme doses, dose rates, stress and temperatures were not investigated and remain poorly understood.

Improvements in computational resources and the advent of accurate interatomic potentials have allowed atomistic modeling to provide greater insight to the connection between interface structure and response to irradiation. When exposed to >25 eV neutron flux, a conventional material incurs displacement damage. Atoms knocked out of place (called self-interstitial atoms) can collect to form clusters causing hardening, leaving behind vacancies that collect to form voids causing swelling. But the same material made of nanocrystals behaves differently. Each tiny crystal, or grain, in the material has a surrounding boundary. These grain boundaries temporarily trap the interstitial atoms, holding them until vacancies are near enough to fill. Thus the material is able to heal itself after radiation damage. Traditional structural materials degrade and can embrittle under intense irradiation, but certain nanocomposites contain high volume fractions of "super-sink" interfaces that allow these materials to self-heal. Understanding how radiation damage is trapped and removed at such interfaces will help in designing a new class of radiation-tolerant materials that would make future nuclear reactors maximally safe, sustainable, and efficient.

Radiation damage spans from the atomic (nanometer, picosecond) to the macroscopic (meter, year) length and time scales [14]. Critical processes involving individual point defect migration are difficult to observe experimentally [15]. Molecular dynamics (MD) simulations are widely used for simulating defect production [16]. Because of time-scale limitations, only information on the picosecond to nanosecond time scale can be obtained, as longer time scales involving defect annihilation and accumulation are inaccessible. Some high-level modeling methods, including kinetic Monte Carlo [15] and dislocation dynamics [14], are useful for extending time

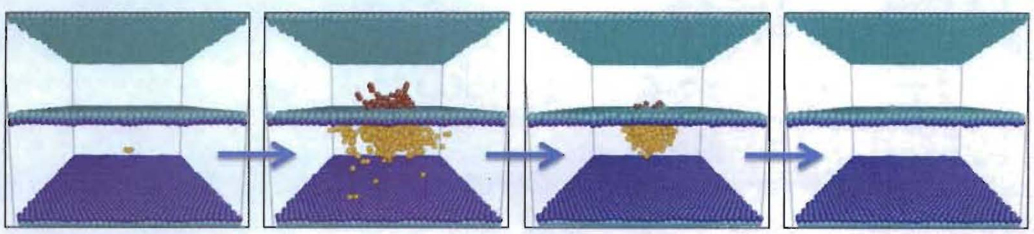

Figure 1: The time sequence (left to right) of radiation damage evolution near a super-sink interface formed by joining copper and niobium in a molecular dynamics simulation. Unlike in pure crystalline materials, the radiation-induced damage is completely absorbed by the interface. Similar effects are seen in post-irradiation examination of irradiated materials. Blue are interface Cu, gray interface Nb, yellow high-energy Cu, and red high-energy Nb in the CuNb nanocomposite material. All perfect fcc and bcc atom environments removed for clarity.

and length scales but require prior knowledge such as defect migration mechanisms and barriers. In complex systems, many key events are non-intuitive, and neglecting them could result in inaccurate predictions. With the use of three atomistic simulation methods, we investigated defect– grain boundary interaction mechanisms in copper, a widely used model system for face-centered–cubic (fcc) metals in radiation damage research, from picosecond to microsecond time scales [17]. Specifically, we use classical MD for simulating defect production at the picosecond time scale, temperature accelerated dynamics (TAD) [18] to bridge the time-scale gap between defect production and aggregation/ annihilation near a GB at the microsecond time scale, and molecular statics to obtain thermodynamic properties of defects near the GB. We found that grain boundaries have a surprising "loading-unloading" effect. ***Figure 1*** illustrates an example. Upon irradiation, interstitials are loaded into the boundary, which then acts as a source, emitting interstitials to annihilate vacancies in the bulk. This unexpected recombination mechanism has a much lower energy barrier than conventional vacancy diffusion and is efficient for annihilating immobile vacancies in the nearby bulk, resulting in self-healing of the radiation-induced damage. The "loading-unloading" mechanism of interstitial emission may help explain the experimental observations that NC materials have better or worse radiation tolerance than polycrystalline (PC) materials, depending on the conditions[10]. Assuming other types of GBs behave similarly to the two GBs studied here, our results explain recent irradiation experiments.

3. The Matter-Radiation Interactions in Extremes (MaRIE) Concept and Future Facilities for Studying Radiation Damage

The Matter Radiation Interactions in Extremes (MaRIE) concept is a National User Facility to realize the vision of 21st century materials research and development. It will provide scientists and engineers with new capabilities for modeling, synthesizing, examining, and testing materials of the future that will enhance the USA's energy security and national security. A key extreme environment for future energy security is intense radiation. In the area of fusion power, the development of new structural alloys with better tolerance to the harsh radiation environments expected in fusion reactors will lead to improved safety and lower operating costs. Evaluation of radiation effects in a fusion environment requires simultaneous displacement damage (~200 dpa) and He generation (~2000 appm He) on polycrystalline samples with properties of bulk material. Data without high fusion relevant dpa and He/dpa are of limited value. Evaluation of mechanical properties for a given material at a given temperature requires a minimum volume of ~10 cm^3 with flux gradients < 20%/cm.

The Fission and Fusion Materials Facility (F^3) segment of MaRIE proposes to use the present proton linac at Los Alamos with a power upgrade to drive a spallation neutron source that can provide the required radiation environment. The calculated radiation damage conditions within the F^3 match, in many respects, that of a fusion reactor first wall, making it well suited for testing fusion materials. Importantly, F^3 would also provide the capability for *in-situ* measurements of transient radiation damage, using unique x-ray and charged particle radiography diagnostics. The mission of the facility would be to study material science in radiation extreme environments to provide predictive capability of material performance to allow certification of new nuclear systems. It is *not* intended for large-volume component testing. Scientific issues such as corrosion, strength and structural integrity, phase stability, thermal transport, and swelling in extreme radiation environments would be addressed. Coupled with integrated synthesis and characterization capability, the *in- situ* measurements with F^3 would reveal not only the atomic scale origins of these effects but also their meso-scale consequences. The radiation environment possible with the current beam current of 16.5 mA and duty factor 7.75% or slight upgrades is shown in ***Figure 2***.

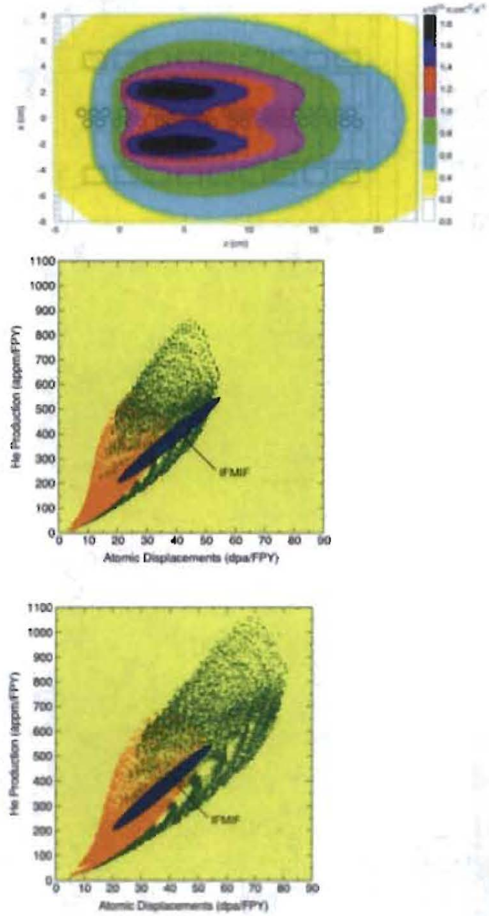

Figure 2: Radiation Environment for the proposed Fission and Fusion Materials Facility. a) Plan of the spallation neutron source, showing neutron flux contours at 1 MW level. b) He production vs atomic displacements at 1.8 MW, with each point representing a volume element in the source. The proposed IFMIF design space is shown for comparison. c) Same as b) but at 3.6 MW.

Building upon a billion-dollar site credit at LANSCE, a linear-accelerator power upgrade is envisioned that, when combined with the DOE-NE LANSCE Materials Test Station Project and the ongoing NNSA LANSCE Linac Risk-Mitigation Project, could provide the U.S. with a cost-effective, unsurpassed testing capability that meets fusion program requirements well before the end of this decade. The E.U. and U.S. fusion communities have recently concluded that a fusion-relevant, high-flux neutron source to enable the accelerated characterization of the effects of radiation damage to materials is a top priority for the next decade [1]. Data from this source will be needed to validate designs for the multi-$B next-generation fusion facilities such as the CTF, ETF, and DEMO, that are envisioned to follow ITER and NIF.

As early as the 1980s, spallation sources were proposed as one option for meeting the fusion materials irradiation mission. To focus the international effort to develop a neutron source for fusion materials studies, the International Energy Agency (IEA) convened a Neutron Source Working Group that evaluated the three leading alternatives: spallation sources, d-Li stripping sources, and D-T sources. The Working Group concluded that the differences in damage parameters were not great enough to permit a selection of one of the alternatives on the basis of displacement rate, primary recoil spectrum, and important gaseous and solid transmutations [19]. That is, all three alternatives met the requirements for producing a neutron source whose damage parameters were sufficiently close to those experienced in the first wall of a fusion reactor. With appropriate upgrades, LANSCE is capable of adequate neutron fluxes for fusion materials studies. In contrast, the IFMIF will likely cost significantly more than a LANSCE power upgrade, be more costly to operate, and take years longer to construct and commission. Similarly, D-T fusion-based neutron source concepts are estimated to cost an order-of-magnitude more than a LANSCE power upgrade, and currently have a considerably lower technology-readiness level. In addition, the use of a spallation source for fusion materials testing offers distinct advantages over other options. For example, thermal reactors cannot produce prototypical helium concentrations, and ion beam sources cannot achieve macroscopic-scale

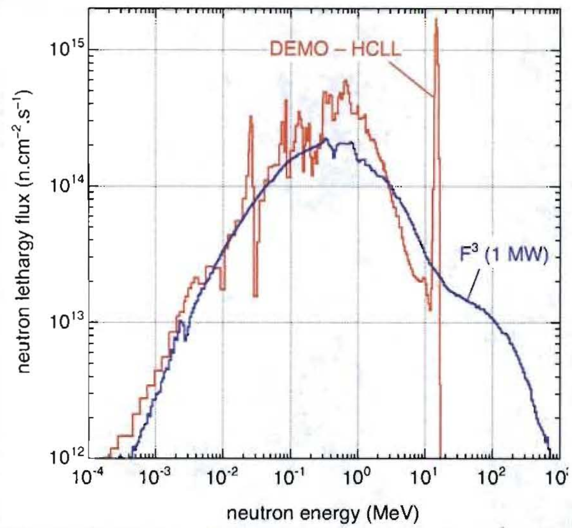

Figure 3: Neutron flux lethargy spectra for F^3 and the DEMO HCLL.

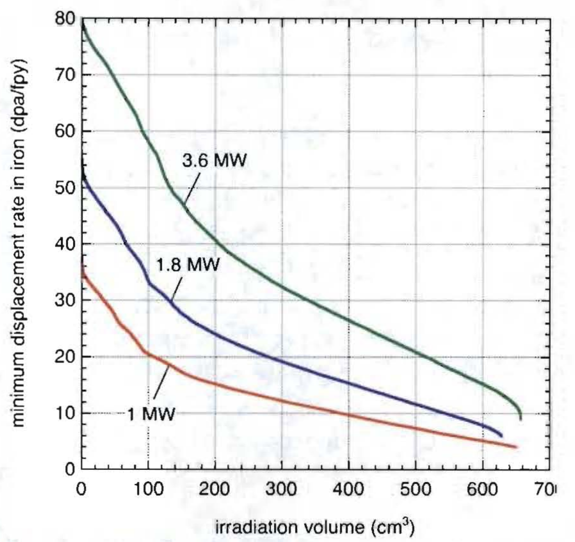

Figure 4: Displacement rate in iron as a function of volume for three different beam powers. The tallied volume includes only that for which the He/dpa ratio ranges from 8 to 13 appm/dpa.

testing. Therefore, today, upgrading the LANSCE linear accelerator offers the lowest technical-risk and the lowest-cost alternative, while also providing important technological advantages, to the DOE and fusion-materials researchers.

There are two important characteristics of the radiation environment with relevancy to radiation damage: the primary knock-on atom spectrum and the impact of the pulse structure of the proton beam on temporal characteristics of the atomic displacement rate. With respect to both of these, analyses show the F^3 has conditions that are consistent with those of a steady-state fusion reactor first wall (see ***Figure 3***).[20]

The F^3 pillar of MaRIE is predicated on the successful deployment of LANL's proposed Materials Test Station (MTS) [21]. This spallation source facility uses the powerful 1-MW proton beam available at the Los Alamos Neutron Science Center to produce 10^{17} neutrons/s. The MTS is sponsored by the US Department of Energy's Office of Nuclear Energy, with the mission of testing fission reactor fuels and structural materials in a fast neutron spectrum. The current schedule calls for the MTS to begin operation in 2016. An assessment of the MTS irradiation environment[22] shows that, with respect to displacement rate, helium production, and production of impurities via transmutation, the MTS has characteristics that match well those expected in the first wall of a fusion reactor.

As part of MaRIE, LANL proposes to augment the MTS with unprecedented in-situ diagnostic capability and an increase in beam power over the MTS level of 1 MW. With these augmentations, the MTS becomes the F^3. The amount of beam power that will drive F^3 is under study, but the goal is to provide researchers with a peak displacement rate of 50 dpa/fpy (full power year), equivalent to an instantaneous rate of 1.58×10^{-6} dpa/s.

The peak displacement rate in the irradiation regions of F^3 is roughly proportional to (beam power)$^{0.8}$. Assuming a beam power of 1.8 MW driving the F^3, the range of displacement and He production rates in iron accessible in the irradiation regions is shown in ***Figure 2***. Each pixel plotted represents a 40-mm^3 volume element in either the fuel (green pixels) or material (orange pixels) irradiation regions. The F^3 offers a broad range of He-to-dpa ratios, from 5 to 33 appm/dpa. This allows scientists to measure materials properties as a function of this important parameter. Increasing the power level of the linac can increase the volume available at ever-higher displacement rates (see ***Figure 4***).

These irradiation regimes are then planned to include dynamic *in-situ* measurements of the effects of radiation as it happens. A suitably brilliant x-ray light source for diffraction and spectroscopy measurements, coupled with radiography such as possible with energetic charged particles, will provide understanding needed to lead to predictive capability for materials in radiation environments. It will also lead to new materials with their functionality controlled to meet the needs of future advanced energy systems. It is an important step towards successful implementation of fusion energy technology.

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