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Breaking the power law: Multiscale simulations of self-ion irradiated tungsten

Miaomiao Jin [a], Cody Permann [b], Michael P. Short [a, *]

[a] Dept. of Nuclear Science and Engineering, Massachusetts Institute of Technology, 77 Massachusetts Ave., Cambridge, MA, 02139, USA
[b] EROB (Engineering Research Office Building) 2525 N. Fremont Ave., Idaho National Laboratory, Idaho Falls, ID, 83145, USA

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ABSTRACT

The initial stage of radiation defect creation has often been shown to follow a power law distribution at short time scales, recently so with tungsten, following many self-organizing patterns found in nature. The evolution of this damage, however, is dominated by interactions between defect clusters, as the coalescence of smaller defects into clusters depends on the balance between transport, absorption, and emission to/from existing clusters. The long-time evolution of radiation-induced defects in tungsten is studied with cluster dynamics parameterized with lower length scale simulations, and is shown to deviate from a power law size distribution. The effects of parameters such as dose rate and total dose, as parameters affecting the strength of the driving force for defect evolution, are also analyzed. Excellent agreement is achieved with regards to an experimentally measured defect size distribution at 30 K. This study provides another satisfactory explanation for experimental observations in addition to that of primary radiation damage, which should be reconciled with additional validation data.

1. Introduction

Tungsten has been chosen as one of the most suitable armor materials for fusion reactors, as plasma-facing materials will be subject to extreme conditions during operation. The divertor in particular will be exposed to simultaneous, heavy neutron irradiation and intense heat fluxes of up to 10 MW/m^2 [1]. Defects accumulated during the exposure to neutron irradiation are known to degrade key material properties, such as ductility and thermal conductivity [2]. Studies on radiation-induced defect cluster creation and evolution are key to providing fundamental explanations and predictions of thermomechanical property evolution, which will drastically impact the service lifetime of the components. It is therefore crucial to understand and be able to predict the long time evolution of microstructural change from the perspective of defect creation, interaction, and evolution.

Experimental research on radiation induced microstructural evolution is still limited, especially when fusion-relevant conditions are considered. Due to the extreme environment to be present in fusion reactors, fission neutrons and ion irradiation are commonly used as surrogates to study the radiation damage by fusion neutrons (typically at an energy of 14 MeV from deuterium-tritium fusion). However, due to the significant difference in the energy spectra of primary-knock-on atoms (PKA), the validity of using results from fission neutrons or ion beams to approximate fusion neutrons is still a matter of debate. Nevertheless, a number of previous studies have shed light on the mechanisms of radiation damage recovery. Residual electrical resistivity has been measured during the recovery process of radiation damage, to identify which defects were responsible and how quickly they moved [3]. The physical mechanisms governing damage recovery and evolution still remain controversial, as the microstructural changes responsible for damage cannot yet be fully characterized. Recent, direct observations of defect creation in tungsten in the transmission electron microscope (TEM) with automated image processing to detect cluster size distributions [4] have given indications of the longer-term evolution of defect clusters produced by radiation damage cascades. More recent experiments on annealing of polycrystalline and single-crystal tungsten present a better picture of defect evolution [5, 6] and links with material property changes [7].

In this work we combine multiple simulation methods including binary collision, molecular dynamics (MD), and rate theory (RT) in the form of cluster dynamics (CD) to model radiation damage in

* Corresponding author.
E-mail addresses: mmjin@mit.edu (M. Jin), cody.permann@inl.gov (C. Permann), hereiam@mit.edu (M.P. Short).

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tungsten. Our results show a deviation from previously predicted power law distributions of defect sizes. The result agrees well with the observations in a recent self-ion irradiation experiment [4], parameterized only by the types and sizes of mobile self-interstitial atom (SIA) clusters. This implies that the mobility of small SIA clusters in irradiated tungsten can be significant enough to explain experimental results, even at cryogenic temperatures. The influence of dose, dose rate, and PKA energy spectrum are also analyzed.

2. Background

Radiation damage continues to be quantified using displacements per atom (dpa) [8], which only describes the initial, ballistic stage of radiation damage. It was initially introduced to reconcile differences in observed radiation damage from reactors or irradiations with different energy spectra [9], and made great strides in discovering similarities between formerly disparate experiments. However, the residual radiation damage in the form of various microstructural defects, ultimately of interest in experiments and material property evolution predictions, is significantly affected by the types of incoming particles [10], the temperature [11], and the dose rate [12]. Also, the long timescale evolution of produced defect clusters over periods of seconds to decades can induce microstructural evolution [13], causing material property changes such as void swelling, hardening, and embrittlement [14]. Thus long timescale studies are important to quantify real damage to irradiated materials. Such lengthy experiments are often infeasible to conduct in realistic conditions, owing to the worldwide scarcity of steady-state neutron sources especially with fusion-relevant neutron energies. Therefore, heavy ion irradiations and simulations are often used as surrogates to neutron irradiation. Commonly employed simulation approaches include the binary collision approximation (BCA) of the initial stage of radiation damage, molecular dynamics (MD) simulations of radiation damage cascades, kinetic Monte Carlo (kMC) for longer timescale evolution of defects, and rate theory (RT) approaches for mean field approximations of defect concentrations. It is these final concentrations at long times, ranging from seconds to years, which are of interest to experimentalists and reactor designers.

The initial production of defects by irradiation can be obtained from BCA Monte-Carlo simulations. The Norgett-Robinson-Torrens (NRT) model can further save computational cost by calculating defect production in terms of dpa with simplified equations [15]. While the dpa cannot determine the defect populations produced by radiation damage, the resulting PKA spectrum turns out to be very useful in follow-on simulations of damage cascades, which allows for the precise description of the movement and formation of defect clusters. MD has been extensively used in this regard to generate a comprehensive database of surviving defects in bulk tungsten resulting from various PKA energies [16]. It was found that interstitial and vacancy cluster production on MD (nanosecond) timescales varies significantly with temperature, cascade energy, and interatomic potential. Meanwhile, the MD simulation time and length scales are on the order of nanoseconds and tens of nanometers respectively, preventing the accurate simulation of further annealing of residual damage from multiple damage cascades. Object Kinetic Monte Carlo (OKMC) picks up here, and can be applied to model the microstructural evolution of materials from the atomic to the mesoscopic scales [17, 18, 19]. It relies on pre-existing knowledge of all important interaction mechanisms of modeled entities, defect stability, and migration properties [20]. While low-barrier events may dominate the kinetics of OKMC simulations, focusing on the reactions of interest can allow one to leapfrog through time to approach experimental timescales. In addition, the number of simulated entities should be large enough to increase the statistical quality of the results. Finally, rate theory approaches such as cluster dynamics can simulate meso- and macroscale time and length evolution of microstructures due to radiation damage. Similar to OKMC, it also requires the parameterization of interaction coefficients and stability constants. Nevertheless, it approximates all defects and other simulated objects with mean field concentrations, thus there is no constraint on dose as encountered in OKMC due to timestep limitations from simulating discrete processes. One merit is that it can flexibly deal with spatial distributions and time evolution of defects during irradiation and annealing. A full picture of the multiscale evolution of radiation damage, culminating in the accurate prediction and understanding of long timescale phenomena, should combine these approaches to reach experimental timescales without sacrificing more detailed information.

The long timescale modeling of radiation damage has made many recent advances. Starting with initial CD studies of point defect cluster evolution [21], a number of improvements have been made. Multiple species have been modeled simultaneously, such as triple beam (H, He, and Fe) irradiation of ferritic alloys [22]. CD modeling of anisotropic materials, such as zirconium, has matched experimental predictions of growth [23]. Stoller et al. have discovered the conditions under which KMC and rate theory calculations overlap well [24]. More recently Jourdan et al. compared the effectiveness of different CD models and cluster size grouping methods [25]. Major advances have also been made in directly linking CD with experiments, such as direct observation of defect production and migration under *in situ* ion irradiation [26], and parameterizing CD simulations directly with experimental TEM observations [27]. Building on the lessons learned from these previous studies, the tungsten self-ion irradiation experiment of Yi et al. [4] is chosen as an ideal test case for our CD code, as its relatively low helium generation rate (from its (n,α) cross section) makes for a relatively simple validation case. In addition, these simulations can provide insights on the evolution processes of defects, and additional sensitivity studies would help identify the important factors that contribute to the final experimental observations.

3. Methods

The SRIM code [28] is widely used to study ion beam implantation, and to compute radiation damage exposure in dpa using the BCA. A recent study [29] recommends that the Kinchin-Pease (K–P) option of SRIM should be selected for accuracy. In this work, 150 keV and 400 keV tungsten self-irradiation were simulated in SRIM using Quick (K–P) mode with 100,000 ions, a displacement threshold energy of 80 eV [30], and a surface binding energy of 11.75 eV [31]. This resulted in PKA energy distributions for each ion irradiation condition, from which PKA energies were sampled in the subsequent MD simulations of damage cascades in tungsten. PKA energies were binned into energy groups of $[10^{1.9}, 10^{2.1}, \cdots, 10^{5.9}]$ eV.

For these simulations, we have tested two EAM potentials from [32] and [33], both of which are smoothly splined to the ZBL potential [34] for short range interactions. The system is first equilibrated for $\sim$ 20 ps at 30 K before introducing each PKA. Damage cascades are initiated by assigning a velocity to the center atom with a random direction. Each PKA energy studied consists of 10–20 independent simulations with different initial directions, with the full PKA energy range from 100 eV to 250 keV studied using the binning method described above. Note that this energy is a reduced form of particle kinetic energy, where electronic loss is excluded. Each cascade is simulated in the NVE ensemble, cooled to