Radiation driven diffusion in γU-Mo

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Abstract

Under the United States High-Performance Research Reactor (HPRR) program, a number of research reactors are planned to undergo a conversion to a U-Mo monolithic fuel design. Both experimental and computational campaigns are underway to ensure the stable and predictable behavior of this fuel type under operation. The accurate prediction of fuel evolution under irradiation requires implementation of correct thermodynamic and kinetic properties into mesoscale and continuum level fuel performance modeling codes. One such property where there exists incomplete data is the diffusion of relevant species under irradiation. Fuel performance swelling predictions rely on an accurate representation of diffusion in order to determine the rate of fission gas swelling and the local microstructural evolution. In this work, we present molecular dynamics simulations combined with rate theory calculations to determine the radiation enhanced diffusion of U and Mo as a function of temperature and fission rate. In combination with previous studies on intrinsic diffusion and radiation driven diffusion in U-Mo alloys, this study completes the multi-component diffusional picture for the U-Mo system. Fuel performance simulations are conducted to illustrate the impact of such fundamental property collection.

1. Introduction

2. Computational Details

The rate of change of defect concentrations with time can be described by equation 1 and 2

$$\frac{dC_v}{dt} = \epsilon\dot{F} - K_{iv}C_iC_v - k_{vs}^2 D_v C_v \tag{1}$$

$$\frac{dC_i}{dt} = \epsilon\dot{F} - K_{iv}C_iC_v - k_{is}^2 D_i C_i \tag{2}$$

where $\dot{F}$ is the fission rate, ϵ is the defect production, K is the recombination constant of vacancies and interstitials, k^2 is the sink strength of grain boundaries, and D is the diffusion coefficient. The subscripts

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i, v, and s denote interstitial, vacancy, and sink, respectively. In this case, sinks are restricted to grain boundaries. The defect production is calculated from the arc-dpa model [**?**], which is a modification of the NRT model for calculating displacements that also includes recombination. The number of defects generated is described by the arc-dpa model as:

$$N_d = \frac{0.8T_d}{2E_d}\zeta(T_d) \tag{3}$$

where T_d is the damage energy, E_d is the displacement energy, and ζ is the arc-dpa efficiency function. The magnitude of E_d and ζ for γ-UMo is not well known, but can potentially be determined from molecular dynamics or from experiments. Given that such studies are beyond the scope of this work, reasonable approximations are made for the displacement energy of 60 eV, based upon molecular dynamics simulations in γU [**?**], and for the efficiency of 0.25, which is approximately the same as bcc Fe [**?**]. The damage energy is taken as the kinetic energy of the fission fragments produced from a fission reaction (approximately 170 MeV), and reduced to account for electronic energy losses. It is assumed that only ballistic effects are generating Frenkel pairs in this work. The electronic energy losses have been previously calculated by Beeler, et al. [**?**] to be 95%, thus the damage energy here is taken as 8.5 MeV. This yields a number of defects per fission in γ-UMo of approximately 14,000. Any bias towards interstitials or vacancies in the defect production process is neglected, assuming that an equal number of both types of defects are generated.

Two sets of molecular dynamics simulations are performed to calculate rate coefficients: a) point defect diffusion, and b) defect evolution in a bulk system. Molecular dynamics simulations are performed utilizing the LAMMPS [28] software package and a U-Mo-Xe embedded atom method (EAM) interatomic potential from Smirnova [29]. For short atomic distances, this interatomic potential was splined to a ZBL [30] via LAMMPS with spline cutoffs of 1 Å and 2 Å. This potential has been shown to reasonably predict a number of properties of γ-UMo, while also being the only ternary interatomic potential capable of describing the U-Mo-Xe system.

To determine diffusion coefficients of a single vacancy or interstitial, a system of 2000 atoms (10x10x10 unit cells) was generated in the body-centered cubic (bcc) structure with an atomic composition of 23% Mo, corresponding to U-10Mo (weight percent). A single defect is randomly generated and the system is equilibrated at a given temperature in an NPT ensemble for 50 ps. Subsequently, the ensemble is switched to an NVT and the system is allowed to evolve for another 100 ns, over which the mean-squared displacement (msd) as a function of time is analyzed. The Nose-Hoover thermostat and barostat as implemented in LAMMPS is utilized, with damping parameters each of 0.1. The msd as a function of time is fit to a linear function and utilized in the Einstein equation (D=msd/6t) in order to calculated the diffusion coefficient for a given temperature. Data for temperatures from 600 K up to 1200 K are obtained, and the entire dataset is fit to an Arrhenius equation, allowing for the determination of the diffusion coefficient prefactor and migration energy for a given defect type.

In order to determine the rate coefficient of recombination (K_{iv}), a supercell of 128000 atoms (40x40x40

unit cells) was generated in the body-centered cubic (bcc) structure with an atomic composition of 23% Mo at the equilibrium lattice constant for a given temperature. Fifty vacancies are removed and fifty interstitials are inserted, ensuring that the distance between individual defects is at least 4×a0, where a0 is the lattice constant. The system is equilibrated for 20000 timesteps with a variable timestep such that a maximum distance for an atom to move in one timestep is 5 fm, in order to perform a constrained relaxation of the defects. Subsequently the system is evolved for 10 ns with a timestep of 1 fs, tracking the number of Frenkel pairs as a function of time via the Voronoi occupation methodology within LAMMPS. Without production and GB absorption, solving equations 1 and 2 leads to $C = C_0/(C_0 K_{iv} t + 1)$ for vacancies and interstitials, where C_0 is the initial concentration. For a given temperature, the number of defects as a function of time can be fit to this relationship.

In classical rate theory, the GBs are constant sinks and their strength k^2 is estimated as $15/L^2$ (L is the grain size, in unit of nm here) for GBs with a regular pattern, and is identical for both interstitials and vacancies. This assumption is utilized here as a first approximation, and completes the parametrization of the rate theory equations.

Given a steady-state concentration of defects under irradiation, the radiation enhanced diffusion coefficient can be expressed as:

$$D_{red} = D_i^{th} C_i^{irr} + D_v^{th} C_v^{irr} \tag{4}$$

where D_{th} is the thermal (intrinsic) diffusion coefficient, C_{irr} is the equilibrium concentration of defects under irradiation, and subscripts i and textitv denote interstitials and vacancies. Utilizing both experimental diffusional investigations and previously computational studies, the total diffusivity can then be taken as the summation of the intrinsic diffusion, D_1, the radiation driven diffusion, D_3, and the radiation enhanced diffusion, D_2.

3. Results

4. Conclusions

In this work, molecular dynamics simulations were performed to determine the radiation driven diffusion of U, Mo and Xe in U-Mo nuclear fuels. Diffusion coefficients for each species were determined and their variance as a function of composition and temperature was analyzed. Updated diffusion coefficients were presented that are applicable under irradiation that incorporate both intrinsic and radiation driven diffusion. This work demonstrates that at temperatures relevant to U-Mo research reactors, it is critical to account for radiation driven diffusion, as this is the dominant mode of diffusion when compared to intrinsic diffusion. The data generated in this manuscript can directly be incorporated into mesoscale and continuum fuel evolution and fuel performance models that describe fission gas and point defect behavior in U-Mo fuels. Finally, this

work points to the possibility of an important role of radiation enhanced diffusion in U-Mo-Xe systems, and as such this will be the topic of future investigation.

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