

Simulations of vacancy cluster behavior in δ -Pu

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Abstract

Using rates for vacancy diffusion in plutonium (Pu) found with parallel-replica dynamics, we develop a kinetic Monte Carlo (KMC) model of void growth and mobility. We compare and contrast the behavior of voids in Pu as predicted using vacancy mobilities from two different modified embedded atom method (MEAM) descriptions of Pu. We find that void behavior depends sensitively on the values used for vacancy mobility. In particular, we find that voids are very mobile in one model of Pu, but are essentially immobile in another, leading to very different void structures over time. This second model also predicts lifetimes for voids that are extremely long, and seemingly unphysical, suggesting that the first model is more representative of real Pu.

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1. Introduction

Plutonium (Pu) is a key component of many important technologies, from the nuclear weapons program to fission reactors. To maximize its potential, maintain safety, and enhance reliability in these applications, its properties must be thoroughly understood. One important property is the structural evolution during self-irradiation, or aging. As Pu atoms decay, the recoils due to the decay create damage in the lattice. This damage, initially consisting of interstitials and vacancies, evolves into larger scale features such as voids and bubbles (when He is present). These larger defects, in turn, govern changes in macroscopic properties such as volumetric expansion and structural stability.

The goal of this work is to examine some evolutionary properties of voids in Pu. We begin by characterizing, using parallel-replica dynamics [1], the mobility of small vacancy defects containing just one or two vacancies using two different parameterizations of Pu within the modified embedded atom method [2,3] description. We then develop a kinetic Monte Carlo model that uses the atomistic results

on vacancy diffusion to examine the evolution and mobility of large voids. We find that the predicted behavior of voids depends on which MEAM description we use for Pu. In particular, long-time pore-size distributions differ for the two models. Such results impact continuum level models that track radiation damage evolution [4,5]. Pore-size distribution can affect other properties such as swelling rates, thermal conductivity, and material strength. Finally we observe unexpected transformations of voids into stacking fault tetrahedra (SFTs).

2. Methodology

Our examination of vacancy behavior in Pu relies on two simulation methodologies: parallel-replica dynamics and kinetic Monte Carlo (KMC). Parallel-replica dynamics involves a straight-forward parallelization of time simulated on multiple processors. Each processor evolves an independent replica of the entire system until a transition to a new state is detected on any of the processors. If the rare events that govern dynamics from state-to-state in the material are first order processes, then parallel-replica dynamics is exact, even describing correlated events correctly. Using M processors in parallel, one can reach a

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maximum speedup compared to standard molecular dynamics (MD) of M times. Reaching this theoretical maximum efficiency depends on how infrequent the events are, as there is some computational overhead involved each time an event is observed. Other details on our use of parallel-replica dynamics as it pertains to studying Pu can be found in Ref. [6]. The events of interest to us are the hopping events associated with vacancy migration and the equilibrium structure of vacancy clusters.

KMC [7], on the other hand, estimates the solution of a set of simultaneously occurring rate processes via a stochastic method. The rates for known mechanisms must be obtained from experimental measurements or, as here, another model or simulation such as parallel-replica MD. The solution method allows for the simulation of much longer times and larger length scales than possible with any type of MD. Given all possible atomic processes and their rates, KMC results in exact dynamics for the system. The disadvantage of this method is that it is typically not possible to know all possible atomic events *a priori*, causing a reduction in the fidelity of the simulation. However, in some cases, even though it is certain that events are missing in the KMC simulation, important insight into the physical system can still be obtained.

3. Plutonium

Pu is an extremely complicated material, exhibiting more allotropes at ambient pressure (6) than any other element. Correctly describing this behavior theoretically has proven challenging, and even the best density functional theory methods have problems accurately describing the δ -phase. In the following work, we use the modified embedded atom method (MEAM) [2,3] to describe the Pu–Pu interaction. This potential is remarkable in that it correctly predicts the relative volume changes of each of the six allotropes of Pu.

We have discussed the particulars of applying parallel-replica dynamics to this system before [6]. Here, we note that the simulations are performed under constant volume conditions and that the temperature of some of the parallel-replica simulations are higher than the melting temperature of Pu (about 913 K). For more details about the simulations, the reader is referred to Ref. [6].

4. Mono- and di-vacancy diffusion in Pu

Past experimental work has yielded a consistent picture of self-diffusion in δ -Pu via vacancies that included tracer diffusion and creep experiments (see Ref. [8] and references therein). As summarized by Fluss et al. [9], the measured activation energy for the vacancy component of self-diffusion is 1.3 ± 0.3 eV. This activation energy is the sum of the vacancy formation energy and the energy barrier for vacancy migration. KMC simulations, in an effort to reproduce experimental results, find the migration energy of vacancies to be about 0.55 eV [9]. However, earlier experi-

Table 1

Prefactor and migration barrier for vacancy mobility in Pu as calculated using the Pu₄ and Pu_X parameterizations of MEAM

	Mono-vacancy		Di-vacancy	
	$v_{\text{mono}} \text{ (s}^{-1}\text{)}$	$E_{\text{mono}} \text{ (eV)}$	$v_{\text{di}} \text{ (s}^{-1}\text{)}$	$E_{\text{di}} \text{ (eV)}$
Pu ₄	5.0×10^{12}	1.06	6.4×10^{13}	1.00
Pu _X	5.3×10^{13}	1.06	2.4×10^{12}	0.48

The principle difference between the two is in the migration barrier for di-vacancy diffusion.

ments found a slightly higher value for the migration energy of 1.1 ± 0.3 eV [10].

We have calculated mono- and di-vacancy diffusion in Pu for two different parameterizations of MEAM using parallel-replica dynamics. These two parameterizations are referred to here as Pu₄ and Pu_X. The parameterizations for these two models can be found in Ref. [11] and [12], respectively. In summary, the difference between the two parameterizations is a small change in phase stability between δ and α . Pu₄ has the α -phase more stable relative to δ than Pu_X. This is accomplished in two ways. One is by altering the parameter that controls whether inversion symmetry in the local crystal structure increases or decreases the cohesive energy relative to the δ -phase, and by how much. The other has to do with the mechanism of ‘screening’ whereby the pair interactions are smoothly transitioned between 1st and 2nd nearest neighbors values.

Vacancy mobilities predicted by Pu₄ have been described elsewhere [6]. We performed a similar study for vacancy behavior in Pu_X. The vacancy mobilities in Pu_X are compared with those found in Pu₄ in Table 1.

The main difference between the mobilities of vacancies predicted by Pu₄ and Pu_X is in the migration barrier of the di-vacancy. The migration barrier for the mono-vacancy is essentially identical in both descriptions. While the rate prefactors for both the mono- and di-vacancy differ in each description by an order of magnitude (with Pu₄ predicting a smaller prefactor for mono-vacancy diffusion and a larger one for di-vacancy diffusion compared to Pu_X), these are relatively small differences, dwarfed, at most temperatures, by the difference in the di-vacancy migration barrier. Pu_X predicts a di-vacancy migration barrier that is about half of that predicted from Pu₄. This means that, even taking into account the higher rate prefactor predicted by Pu₄, the rate of di-vacancy diffusion is predicted by Pu_X to be 10^4 times greater at $T = 500$ K than as predicted by Pu₄. Thus, the evolution of vacancies and vacancy clusters predicted by the two models will be very different.

5. KMC model of vacancy clusters

In order to understand the consequences of the difference in di-vacancy mobility on void behavior, we have constructed a simple KMC model that incorporates the atomic-scale mobilities found via parallel-replica dynamics. The model has two components: the stability of vacancy

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