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METAL

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Atomistic models of point defects in plutonium metal

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The aging properties of plutonium (Pu) metal and alloys are driven by a combination of materials composition, processing history, and self-irradiation effects. Understanding these driving forces requires a knowledge of both thermodynamic and defect properties of the material. The multiplicity of phases and the small changes in temperature, pressure, and/or stress that can induce phase changes lie at the heart of these properties. In terms of radiation damage, Pu metal represents a unique situation because of the large volume changes that accompany the phase changes. The most workable form of the metal is the fcc (δ-) phase, which in practice is stabilized by addition of alloying elements such as Ga or Al. The thermodynamically stable phase at ambient conditions is the monoclinic (α-) phase, which, however, is 20 % lower in volume than the δ phase. In stabilized Pu metal, there is an interplay between the natural swelling tendencies of fcc metals and the volume-contraction tendency of the underlying thermodynamically stable phase. This study explores the point defect properties that are necessary to model the long-term outcome of this interplay.

Point-defect properties are atomistic in nature. To study point defect production and migration, it is necessary to construct an atomistic model of the interactions among Pu atoms. Recently progress has been achieved in the form of a modified embedded atom (MEAM)[1,2] potential for pure Pu.[3] The MEAM potential was able to capture the most salient features of atomic volume and enthalpy of Pu metal and liquid metal as a function of temperature at zero pressure (Figs. 1a and 1b). Most significantly the atomic volume difference between the α- and δ-phases was captured nearly quantitatively.

Here we use this potential to simulate the point defects that form as the result of a self-irradiation cascade in Pu metal at room temperature (RT) and ambient pressure. The first property of interest is the minimum displacement threshold energy (DTE). The
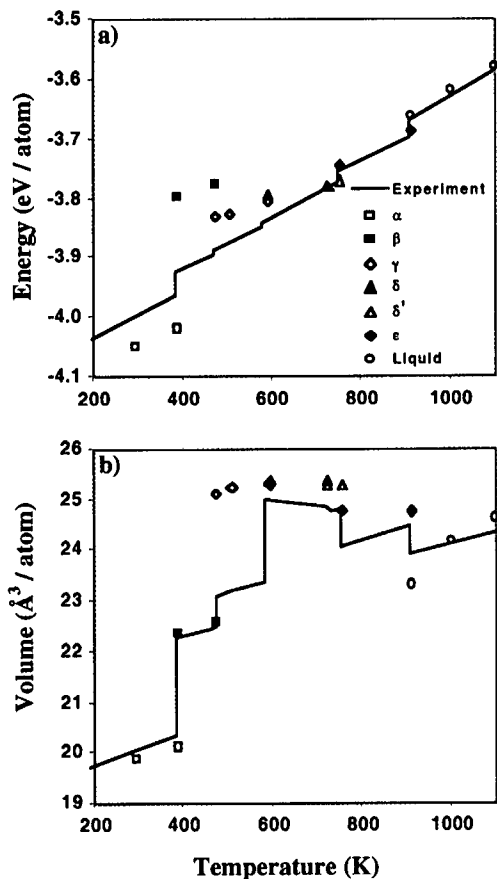
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Figure 1. MEAM potential model of Pu metal at low pressures as a function of temperature. The differences in atomic energy and volume between α and δ Pu are quite close to the experimental values.

DTE represents the minimum energy in a cascade required to cause the formation of a Frenkel pair. The potential represented in Fig. 1 (model I) has a very low DTE, 10 eV, compared to other fcc metals.[4] See Table 1. For this reason, we consider a second set of parameters (model II) in which the fcc phase is stable all the way down to 0 K. The DTE for model II is 18 eV.

The reason for the low DTE in model I can be directly related to the low-temperature properties of the model. Below 100 K, the fcc lattice of model I undergoes a rhombohedral distortion.[4] In real Pu metal, the fcc lattice is unstable as the temperature is decreased (Fig. 1a). So model I is realistic in this sense. The fcc lattice in model II has no such low-temperature distortions. Consequently, it behaves more fcc-like, as one might expect for Ga-stabilized

Table 1. Comparison of DTEs (E_d) for the two models of Pu and some fcc metals. The experimental values are for polycrystalline samples using electron beam irradiation, except for Pu.

Metal	Pu	Ag	Al	Au	Cu	Ni	Pt	Pb
Experiment E_d (eV)	33[a] 14[b]	25[e]	16[e]	36[e]	19[e]	23[e]	24[e]	12.5-15[f]
MEAM E_d (eV)	10[c] 18[d]			20.9[c]	17.7[c]	23.7[c]	24.1[c]	15[c]

[a]Reference 6 value based on sublimation energies in fcc metals.
[b]Reference 7 value based on melting temperature of metals.
[c]Reference 4.
[d]Reference 4 (Model II).
[e]Reference 5.
[f]Reference 8.

Pu. Currently, there is no way to validate either model.

Next we are concerned with interstitial defects. Each cascade produced by the fission of a Pu atom produces about 2500 Frenkel pairs. The structure of the interstitial for model I is a (100) split dumbbell, whereas the interstitial for model II remains in an octahedral site. The migration barrier for the interstitial in model I is 0.055 eV.[4] This value is typical for fcc metals.

Vacancies migrate much more slowing, leading to an imbalance between interstitials and vacancies. In model I, the barrier is at most 1 eV, while the value for model II is 1.5 eV. These values appear to be high relative to the experimental value of 0.68 eV measured by isochronal annealing.[9] Parallel replica dynamics simulations[10] shows that mono-vacancy migration proceeds by two-atom hops as often as by one-atom hops.

Finally, the bias volume, resulting from the difference between interstitial and vacancy volumes, provides a simple measure of the driving force for void swelling. Model I has no measurable bias volume. Figure 2 shows bias volume for Model II Pu as well as for Ni and Pb. The Ni and Pb values are close to the experimental values.
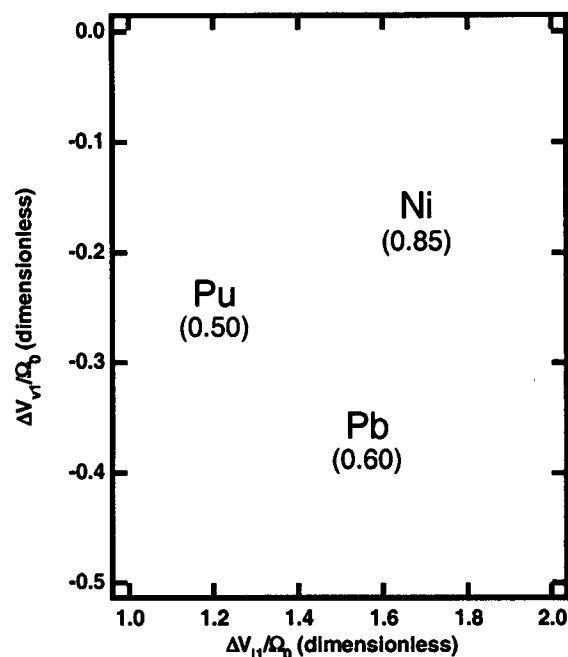

Figure 2. Bias volumes from MEAM potentials. Pu value is for model II.

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