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Point-defect production and migration in Pu metal at ambient conditions

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Modeling thermodynamics and defect production in plutonium (Pu) metal and its alloys, has proven to be singularly difficult. The multiplicity of phases and the small changes in temperature, pressure, and/or stress that can induce phase changes lie at the heart of this difficulty. 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 the δ phase is stabilized by addition of alloying elements such as Ga or Al. The thermodynamically stable phase at ambient conditions is the between monoclinic (α-) phase, which, however, is approximately 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 phase transformation to the thermodynamically stable phase. This study explores the point defect production and migration properties that are necessary to eventually 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 formation of point defects due to self-irradiation in Pu metal at room temperature (RT) and ambient

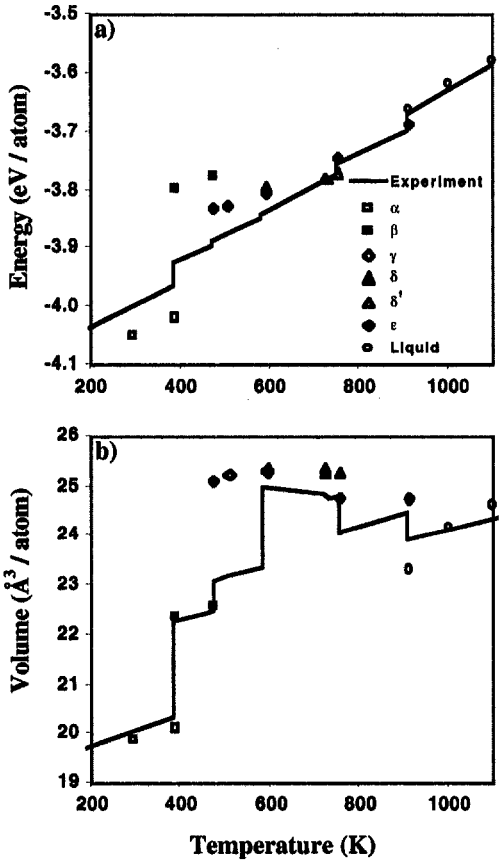

Figure 1. MEAM potential model of Pu metal at low pressures as a function of temperature. The differences in atomic energy and volume between α and δ Pu are quite close to the experimental values.

pressure. The simulations are performed by molecular dynamics trajectory calculations of damage cascades events. A cell of several hundred or more atoms of Pu are arranged in an fcc lattice. The cell is periodic in all three dimensions. One atom is selected to be the primary knock-on atom (PKA) and is assigned the desired cascade energy. We are concerned with the point defect damage that typically occurs at the end of a cascade. Therefore, the energy given to the PKA is much lower than that

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of a fission product or radiative particle from a reactor environment. The cascade phase of the simulations lasts in the range of 0.1 to 0.5 ps, depending on the PKA energy. PKA energies from 5 to 300 eV have been simulated.

Because of the phase instability of δ Pu, it is necessary to perform the cascade simulations at constant volume rather than at constant pressure. In the MEAM model of Pu, allowing the cell boundaries to relax would lead to a volume collapse of the cell and subsequent transformation of the atomic arrangement to the α phase. Two facts suggest that constant-volume conditions are not too severe of an approximation to normal environmental conditions. One is the small thermal expansion coefficient of δ-Pu. Two is a comparison between defect formation at 600 K and constant pressure (P) vs. 600 K and constant volume (V) showed good agreement. The fcc lattice is stable under both sets of conditions. We take these as reasonable assurance that our choice of boundary conditions has not unduly skewed our results.

In its α phase, the Pu lattice is stable under ambient conditions. Molecular dynamics simulations of the damage cascades can be performed with constant pressure boundary conditions.

We find that δ-Pu under these conditions has a very low minimum displacement threshold energy compared to most other fcc materials, has less crystallographic anisotropy in this minimum.[4] At low projectile energies, the models suggests markedly greater damage than is predicted by the Kinchin-Pease (KP) and Norgett-Robinson-Torrens (NRT) models of damage accumulation[5-7]. On the other hand, simple monoclinic (α phase) Pu has a damage threshold energy minimum consistent with that predicted by sublimation energy arguments.

Some of the peculiarities of the Pu lattice as predicted by this MEAM potential are illustrated in Figs. 2a and 2b. The lattices undergo a number of distortions which all lie closely in energy to their respective parent perfect lattices. The dominate distortions in the fcc lattice in the <110> direction. The structure is achieved by quenching a structure annealed at 600 K. At 600 K, δ-Pu lattice is stable at 0 Pa. One possibility for the low damage threshold may be the presence of low-energy nearby configurations of the lattice in this model potential.

For the damage annealing phase, the cell was allowed to gradually thermalize under the influence

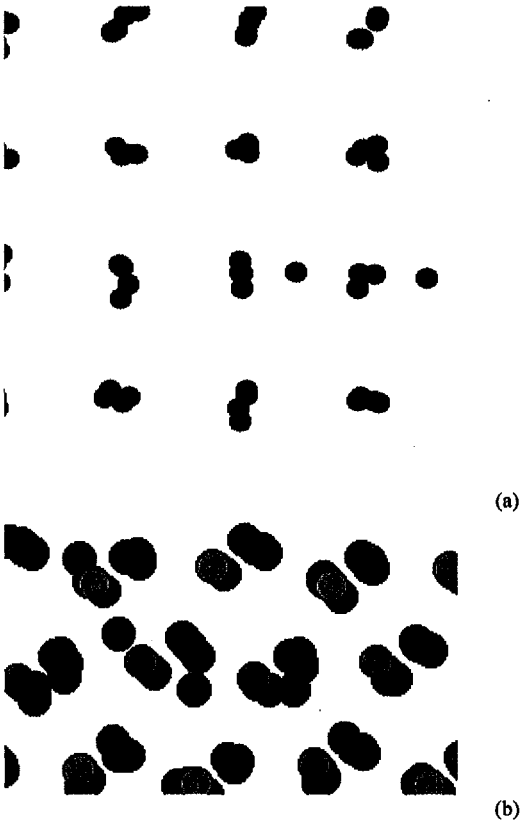

Figure 2. Examples of lattice distortions in the MEAM model for Pu metal and point defects generated by molecular dynamics simulations of self-irradiation damage at low energies. (a) The fcc lattice with a split interstitial shown by the two darker atoms. (b) The simple monoclinic lattice with at least one split interstitial in medium gray to the left of center.

of a weak Nosé-Hoover thermostat[8] with a time constant of 10 ps. The majority of the initial displacements are removed during this time, with only a few point defects surviving. The annealing phase lasts from 5 to 10 ps depending on the PKA energy.

The remaining defects in the fcc lattice are typically split interstitials, oriented along a <100> direction. Analogous dumbbell interstitial defect clusters are also found in Au. Based on molecular dynamics simulations, the minimum displacement threshold energy for fcc Pu at 300 K and constant V is estimated to be 10 eV. The major point defects in α-Pu also appears to be a split interstitial. Distortions in the α lattice make a clear determination of its orientation somewhat ambiguous. The minimum damage threshold energy is estimated at 30 eV.

Finally, we simulated interstitial and vacancy migration under constant-volume/constant-temperature conditions. The migration barrier for the

interstitial is sufficiently small that its migration can be simulated directly. By calculating a mean-square displacement of the interstitial as a function of time for a range of temperatures, an migration barrier of 0.055 eV is found.[4] This value is typical for fcc metals. The mono-vacancy migration is much higher. Simulations at 900 K for 1 ns produced no migration events. This places a lower bound on the vacancy migration barrier of 0.8 eV.[4] This value appears to be high relative to the experimental value of 0.68 eV measured by isochronal annealing.[9] Given that no defect migration information was used in fitting the MEAM Pu potential, it is not too surprising to find a disagreement at the 0.12-eV level in a migration barrier.

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