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Author(s): Steven M. Valone, Michael I. Baskes, Marius A. Stan, and Kurt E. Sickafus

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Production of point defects at low cascade energies in
fcc Pu metal at 300 K and constant volume

Steven M. Valone, Michael I. Baskes,
Marius A. Stan, and Kurt E. Sickafus

Materials Science and Technology Division, Structure-Property Relations Group, Los
Alamos National Laboratory, Los Alamos, NM, 87545 USA

‡ Corresponding author: smv@lanl.gov

Introduction

Modeling thermodynamic and defect production in plutonium (Pu) metal itself, say nothing of Pu
alloys, has proven to be singularly difficult. Recently, however, progress has been achieved in
the form of a modified embedded atom (MEAM) [1-2] potential for pure Pu [3]. The MEAM
potential is 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 monoclinic (α-phase) and fcc (β-phase) was
captured nearly quantitatively.

Here we use this potential to explore the formation of point defects due to self-irradiation in Pu
metal room temperature (RT). We find that Pu under these conditions has a very low minimum
displacement threshold energy compared to most other fcc materials, has less anisotropy in this
minimum, and, at low projectile energies, yields markedly greater damage than is predicted by
the Kinchin-Pease (KP) and Norgett-Robinson-Torrens (NRT) models of damage accumulation
[4-6].

The Pu lattice is not stable in its fcc arrangement at RT and zero pressure. Consequently, we
found it necessary to use constant volume conditions instead. Comparison between defect
formation at 600 K and constant pressure (P) vs 600 K and constant volume (V), where the fcc
lattice is stable under both conditions, showed good agreement. We take this as reasonable
assurance that our choice of boundary conditions has not unduly skewed our results.

Some of the peculiarities of the fcc-Pu lattice as predicted by this MEAM potential are illustrated
in Fig. 2. The lattice actually undergoes a tetragonal distortion in the <110> direction. The
structure is achieved by quenching a structure annealed at 600 K. At 600 K, the fcc-Pu lattice is
stable at 0 Pa. A space group of P42_1m is deduced from simulated X-Ray powder diffraction.
The distortion may be arising from the proximity of the α-Pu structure. The MEAM potential,
being a single, adiabatic surface, must mimic the fcc and monoclinic structures as local minima.
The presence of the other structures in other, near-by potential wells can then induce distortions.
It is significant that the distortions stay at about the same level, 0.25 Å, even down to 300 K.
Again, we mention that imposing symmetric, constant V boundary condition imparts
metastability to the fcc structure down to at least 300 K.

Besides the tetragonal distortion, the interstitial point defect is almost exclusively a structure
which is split along the (100) direction. Analogous dumbell interstitial defect clusters are also
Simulations of defect production in a cascade

The MEAM potential parameter values are given in Table 1. The values used to fit the potential are noted. Figures 1a and 1b show the near-quantitative reproduction of the enthalpy of formation and atomic volumes of the MEAM-Pu potential. The most difficult systems are γ (distorted diamond), for enthalpy, and the β (32 atoms/cell monoclinic), for atomic volume. Nevertheless, the differences in these quantities between the α- and δ-phases are very close to the best-available experimental values. These differences are seen as critical for self-irradiation damage simulations because of the possibility of radiation-induced phase transformation of the metastable δ-phase.

Simulations of defect production were carried out by selecting one atom in the simulation cell to act as the primary knockon atom (PKA). Conditions for the simulations were 300 K and constant V for 5 ps. The PKA energy was varied from 4.25 volts to 100 eV and directed in the <100>, <110>, <111>, and <121> directions. The size of the simulation cell was varied from 256 to 2048 atoms, depending on the PKA energy. Periodic boundary conditions were assigned in all three cartesian directions. A 1 fs timestep was used throughout. A Nordsieck integrator [7] was used with no timestep adjustment permitted.

Initial conditions were selected by repeatedly equilibrating the undamaged lattice at the desired temperature and volume, with a Nosé-Hoover-drag thermostat activated with a 0.1-ps time constant [8]. During a damage trajectory, atoms other than the PKA were mildly thermostatted with 10-ps time constant. The thermostatting was intended to mimic the thermal conduction of the electronic degrees of freedom, even though no quantitative basis was actually used in choosing the 10-ps time constant.

After a trajectory was run, the final configuration was subjected to a quenching processing using a conjugate-gradient method. This was done to eliminate very short-lived defects. This made identification of defects more definitive.

Besides the tetragonal distortion, Fig. 2 also shows a residual interstitial defect and a vacancy left by the PKA. The interstitial point defect is almost exclusively a structure which is split along the (100) direction. Analogous dumbell interstitial defect clusters are also found in Au. This defect will be discussed in the following. The production of these defects relative to the KP model is shown in Fig. 3. Based on these results, the minimum displacement threshold energy for fcc Pu at 300 K and constant V is estimated to be 10 eV, i.e. where the ratio is 1.

At much higher PKA energies, one would expect this ratio to drop to approximately 0.5 ± 0.2, based on other fcc metals. At these lower PKA energies, however, the effective temperature within the cascade apparently does not permit as much annealing as one would expect based on the models. On the one hand, this difference in behavior could be the result of a simple competition between cascade dynamics (i.e. how many atoms are displaced at a given energy) versus dissipation of the PKA energy. On the other hand, it could be an effect of the multiple phases available to Pu metal as the cascade cycles through its range temperatures.

In summary, we find that for fcc Pu, at 300 K and constant V, the minimum displacement threshold energy is 10 eV. The predominant point defect is a split interstitial oriented along the
principle axes of the fcc crystal. The defect production appears much less anisotropic compared to other fcc metals such as Au and Pb.

References


Table 1.
Source and values of MEAM parameters for plutonium. Where appropriate experimental data was not available, nominal values are chosen similar to the values used for other elements.

<table>
<thead>
<tr>
<th>parameter</th>
<th>source</th>
<th>value</th>
</tr>
</thead>
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<tr>
<td>$E_s$ (eV)</td>
<td>cohesive energy of the liquid (Wick, 1980)</td>
<td>3.80</td>
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<tr>
<td>$r_s$ (Å)</td>
<td>lattice constant of δ (Wick, 1980)</td>
<td>3.28</td>
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<td>$\alpha$</td>
<td>bulk modulus of Ga-stabilized δ (Ledbetter and Moment, 1976)</td>
<td>3.31</td>
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<tr>
<td>$\delta$</td>
<td>pressure derivative of bulk modulus of α (Roof, 1981)</td>
<td>0.46</td>
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<td>$A$</td>
<td>relative energy of δ and ε (Wick, 1980)</td>
<td>1.05</td>
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<tr>
<td>$\beta^{(0)}$</td>
<td>shear modulus of Ga-stabilized δ (Ledbetter and Moment, 1976)</td>
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<tr>
<td>$\beta^{(0)}$</td>
<td>nominal value</td>
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<tr>
<td>$\beta^{(0)}$</td>
<td>nominal value</td>
<td>6.0</td>
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<tr>
<td>$\beta^{(0)}$</td>
<td>volume per atom of α (Wick, 1980)</td>
<td>9.0</td>
</tr>
<tr>
<td>$r^{(0)}$</td>
<td>nominal value</td>
<td>1.0</td>
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<td>$r^{(0)}$</td>
<td>shear modulus of Ga-stabilized δ (Ledbetter and Moment, 1976)</td>
<td>4.64</td>
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<tr>
<td>$r^{(0)}$</td>
<td>relative energy of δ and α (Wick, 1980)</td>
<td>-0.8</td>
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</table>

Fig. 1. Energy (1a, left) and volume (1b, right) per atom as predicted by the MEAM potential for Pu constructed by Baskes [3]. The solid lines represent the best available experimental data and the symbols are the predictions and/or fits from the MEAM potential.

Fig. 2. Damage cascade simulation results for an 8.0 eV-projectile energy in δ-Pu at 300 K and constant volume.

- Distorted-δ Pu
- Vacancy marker
Fig. 3. Frenkel pair production vs projectile energy in δ-Pu, at 300 K and constant volume. The difference between damage energy and projectile energy stems from the effect of thermal conduction of the electrons in the metal.