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Monte Carlo + molecular dynamics modeling of radiation damages in Pu

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Abstract

The paper describes calculations on the evolution of damage cascades in self-irradiated unalloyed and gallium-alloyed δ-Pu. The fast stage of the evolution was simulated by the Monte Carlo (MC) method. When the energies of cascade particles became close to the displacement energy, the cascade configuration was transferred to a molecular dynamics (MD) code which tracked the further evolution of the system to ∼2 ns. The simulations showed that a cascade of damages from the U recoil nucleus caused a large energy release into a lattice subsystem within a local region about 10 nm in size where the material melted and then recrystallized. Preliminary estimates showed that the energy transferred to the lattice was enough to cause melting in a region whose characteristic size was ∼15 nm (∼200,000 atoms). MD simulations showed heat conductivity to reduce the characteristic size of the melting region to ∼8 nm (∼12,000 atoms) in a sample whose initial temperature was 300 K. The time of recrystallization was estimated to be ∼1 ns. It was shown that most point defects created during the fast stage of the cascade were recovered in melting and recrystallizing. A number of calculations were also done for polycrystalline samples.

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Keywords: δ-Pu; Radiation defects; Molecular dynamics; MEAM

1. Introduction

The ageing of actinides, i.e., the change of their properties with time due to self-irradiation, is caused by the accumulation of radiation defects and helium in the bulk of the material. Investigation into this complicated problem may be addressed to molecular dynamics (MD). The atomistic approach to material properties modeling is one of the rapidly developing directions of the theoretical material science. This approach gives detailed information on the structures and processes on the micro-level.

The radioactive decay of Pu generates high-energy particles of U (86 keV) and He (5 MeV) producing numerous damages when decelerating in the bulk of the material. In this paper we focuse on the evolution of damage cascades from uranium atoms. We consider the process to consist of fast and slow stages. The fast stage lasts while particle energies are several times greater than the assumed displacement energy. This stage is simulated in terms of the Monte Carlo (MC) technique. The slow stage is characterized by the absence of new displacements caused by high-energy atoms and the system evolves in the timescale of thermal relaxation. This stage is described by the MD technique. The Modified Embedded Atom Model (MEAM) developed for Pu and Pu–Ga alloys (see Refs. [1,2]) is used to describe the interatomic interaction. Calculations were carried out for pure δ-Pu and a Ga (1 wt.%) stabilized δ-Pu alloy.

The combination of MC and MD techniques allows us on the one hand, to take into account inelastic scattering and energy losses of high-energy particles and to eliminate shortcomings of the MEAM when interatomic distances are small and on the other hand, to track the cascade evolution in time. Using MC instead of the full MD treatment with the adequate short range potential makes it much easier to gather statistics and to obtain the averaged characteristics of the cascades.

2. Monte Carlo simulation

The MC technique applied is similar to MARLOWE [3]; it allows for the crystal structure and thermal vibrations of lattice atoms. According to Ref. [4], we assume Debye temperature $\theta_D = 116$ K and displacements of atoms from perfect

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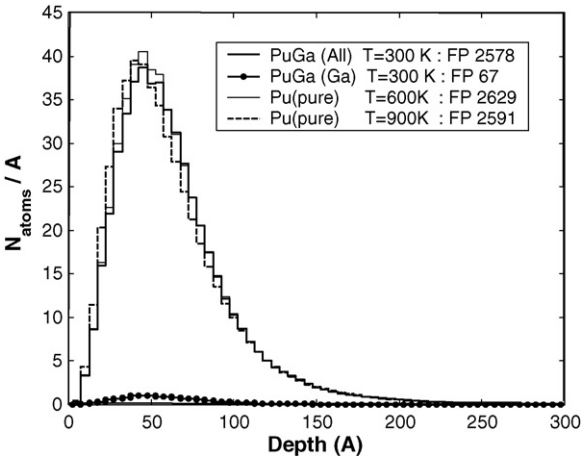

Fig. 1. The distribution of vacancies (interstitials) as a function of distance from the alpha-decay point; the Universal potential. FP is the number of Frenkel pairs. PuGa(All) is the distribution of all vacancies and PuGa(Ga) is the distribution of Ga vacancies.

lattice nodes are defined by the triangular distribution from Ref. [5].

The effect of the potential used was checked on two potentials: Universal and Moliere [6]. The result was found to be insensitive to the potential. The stopping power of inelastic scattering (the loss of energy due to ionization and excitation) was taken in Lindhard–Sharf form [6] ($k_L = 3$). The average displacement energy $E_d = 10$ eV was taken to be the same for pure Pu and Pu–Ga alloy. This value of E_d was evaluated in Ref. [7] and proved in this work in MD calculations.

MC calculations were carried out for pure δ-Pu at $T = 600$ and 900 K and for Pu–Ga alloy at $T = 300$ K. The distribution of Frenkel pairs as a function of distance from the decay point is shown in Fig. 1. The total number of Frenkel pairs agrees well with the estimate given in Ref. [10]. It is seen that the initial temperature is not a factor which significantly affects the result. The total number of Frenkel pairs slightly decreases as the temperature grows. The size of the damaged region is ~10 nm.

3. Molecular dynamics simulation

When the energies of cascade particles became close to the displacement energy (<15 eV), the cascade configuration resulted (coordinates + particle velocities) was transferred to a molecular dynamics code which tracked the further evolution of the system to ~1–2 ns. The size of the MD box was $100 \times 100 \times 100$ unit cells. Our MC calculations give the energy loss due to inelastic scattering to be about 40–50%. The rest energy is absorbed by the crystal lattice. Applying periodic boundary conditions would cause a temperature change $\Delta T \sim 150$ K in the whole system. To avoid this the MD box was adjoined to a thermostat kept at the initial temperature of the sample. Assuming specific heat of Pu lattice to be $C_P \sim 3R$ and melting heat to be $Q_{melt} = 2.88$ kJ/mole, one can evaluate that at the initial temperature $T = 600$ K, the energy transferred to the lattice is enough to cause melting in a region whose characteristic size reaches ~20 nm; the number of atoms in the region is ~350,000. At the initial temperature of 300 K, the number is 1.8 times smaller. The same is true for the Pu–Ga alloy so as for a Ga concentration of 1 wt.%, changes in melting temperature and melting heat are small (see Ref. [8]). In reality, heat conductivity must significantly reduce the size of the melted region. The cascade evolution in pure δ-Pu at the initial temperature 600 K is shown in Fig. 2. The time 0 ps corresponds to the beginning of the MD stage. It is seen from the figure that

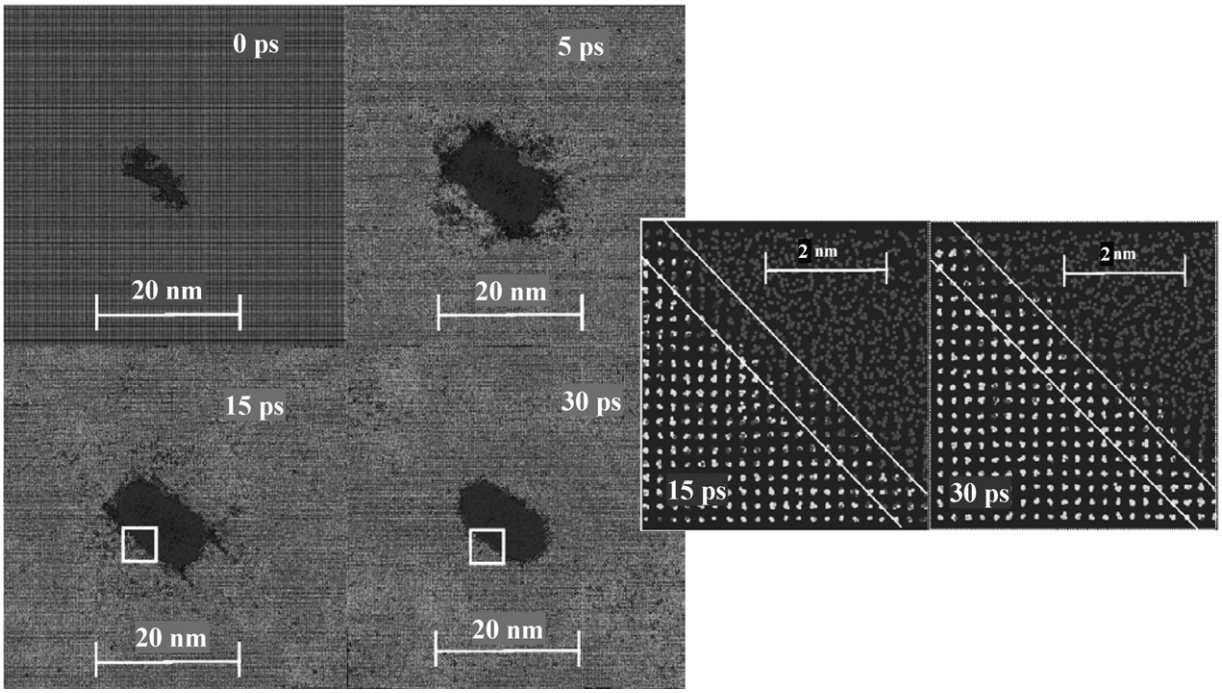

Fig. 2. Fragments of a monocrystalline sample of δ-Pu contaning the cascade at different times; initial temperature 600 K. Dark grey is for the disordered structure (melted region) and light grey is for fcc δ-Pu.