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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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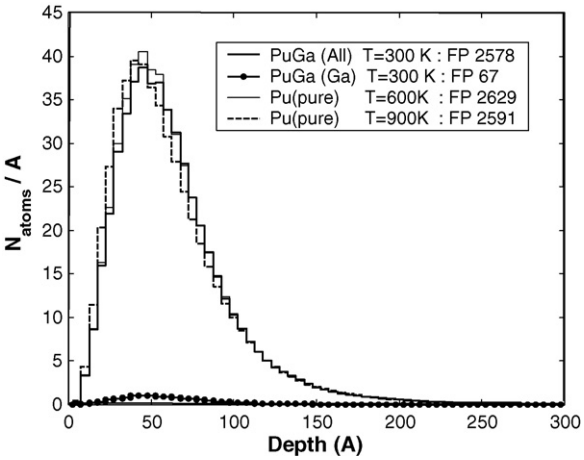

Fig. 1. The distribution of vacancies (interstitials) as a function of distance from the alpha-decay point; the Universal potential. FP is the number of Frenkel pairs. PuGa(All) is the distribution of all vacancies and PuGa(Ga) is the distribution of Ga vacancies.

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

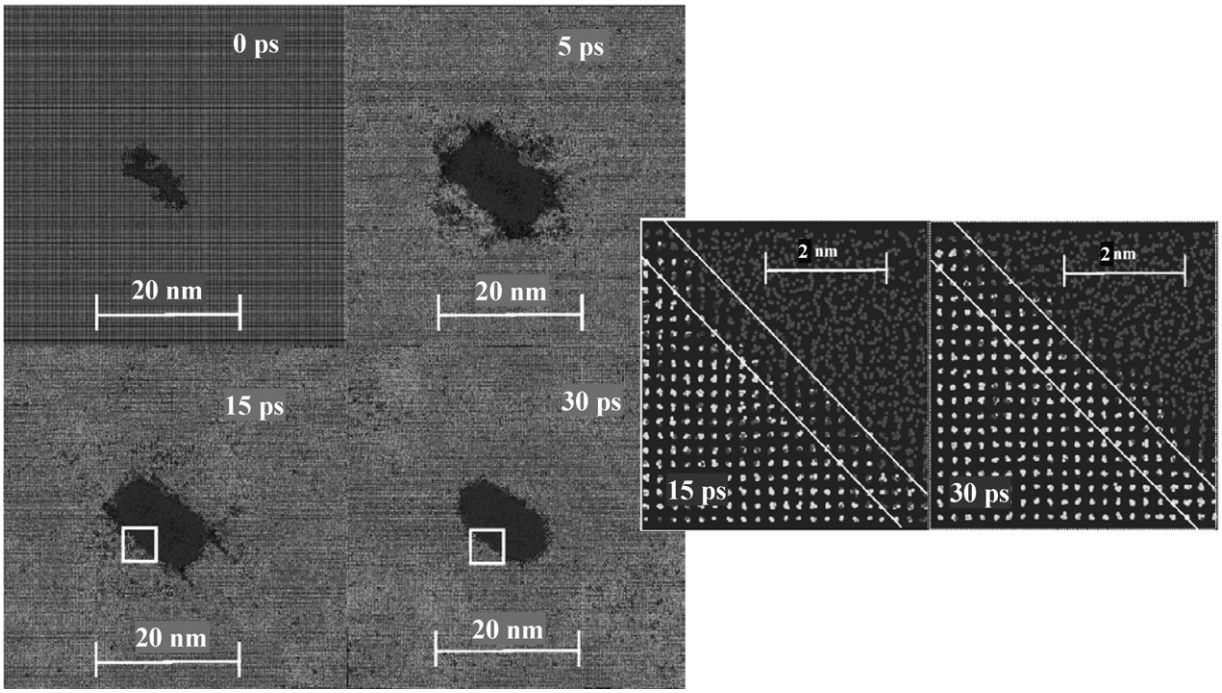

Fig. 2. Fragments of a monocrystalline sample of δ-Pu contaning the cascade at different times; initial temperature 600 K. Dark grey is for the disordered structure (melted region) and light grey is for fcc δ-Pu.

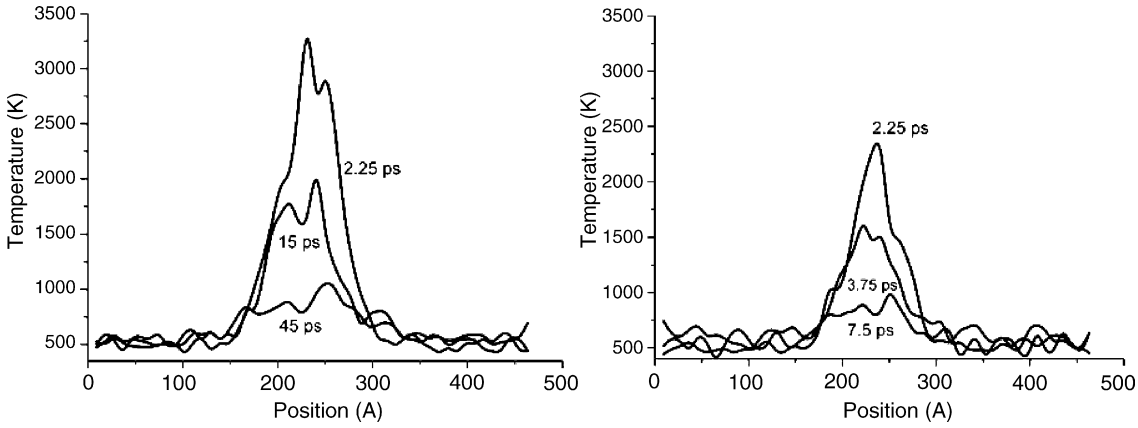

Fig. 3. Temperature profiles in a δ-Pu sample cross-section at different times; initial temperature 600 K. Left: without EPC and right: with EPC.

the hot cascade region heats the surrounding undisturbed region and causes its melting. Temperature profiles in the cross-section of the melted region are presented in Fig. 3(left). The temperature in the center is seen to reach 3500 K. At first the melted region rapidly expands but after approximately ∼10–12 ps the temperature in the heated region begins to decrease due to lattice heat conductivity and the process of recrystallization starts. Zoomed figures on the right side of Fig. 2 illustrate the process of recrystallization. After ∼50 ps, the temperature in the center drops below the melting point and after ∼80 ps, the damaged region is thermalized to the sample temperature. So the further annealing of the amorphous region takes place at this temperature.

The whole body of the melted material is shown in Fig. 4(left). The rapid growth and simultaneous thermal expansion of the melted region cause plastic deformation in the adjacent regions. High-rate straining induces stacking faults and partial dislocations (see Ref. [9]) shown in Fig. 4(left). The total number of atoms in the melted region at the moment of its maximum size is ∼25,000. After ∼1–1.5 ns, the amorphous region completely recrystallizes leaving point-like defects: vacancies and interstitials. Note that in pure Pu at $T_{\text{init}} = 600$ K, the residual amorphous region is not observed. In the PuGa alloy at $T_{\text{init}} = 300$ K, the process of annealing is slower and after 1 ns there is an amorphous region which makes 40% of its maximum size and gradually decreases with time. The number of residual point defects (250 vacancies + 250 interstitials) is an order of magnitude smaller than the number of Frenkel pairs created during the fast (MC) stage. At the moment corresponding to the end of the simulation, vacancies and interstitials have different spatial distributions shown in Fig. 4(left): vacancies form a compact cluster, whereas interstitials form an extended rarefied cloud due to their higher movability if compared to vacancies. The average time between interstitial jumps at $T = 600$ K is estimated to be ∼10–15 ps. In the region affected by the cascade, the temperature is much higher and the interstitial movability is higher as well. The further evolution of the defects is much slower than the nano-second time scale.

These calculations take into account only lattice heat conductivity. But plutonium is metal and its total heat conductivity at $T = 600$ K is, according to Ref. [11], 11 W/m K. Our calculations of lattice heat conductivity give 1 W/mK that is an order of magnitude smaller. So as about 60% of energy is absorbed by the lattice, the role of electron heat conductivity is defined

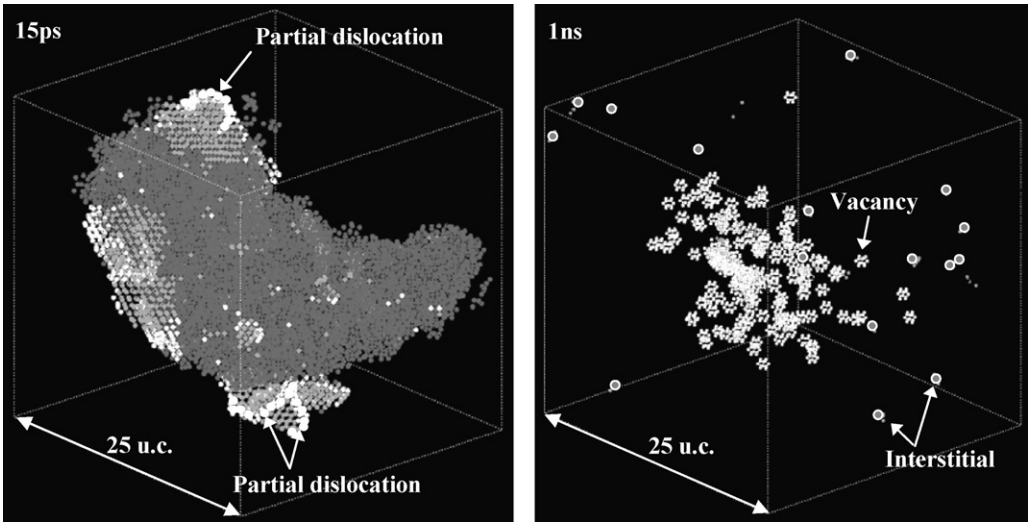

Fig. 4. (Left) Melted region and plastic deformation in δ-Pu. Initial temperature 600 K. Dark grey is for the disordered structure (melted region), light grey is for stacking faults, white is for partial dislocations. (Right) Residual point-like defects: white encircled spheres are interstitials and stars are vacancies.

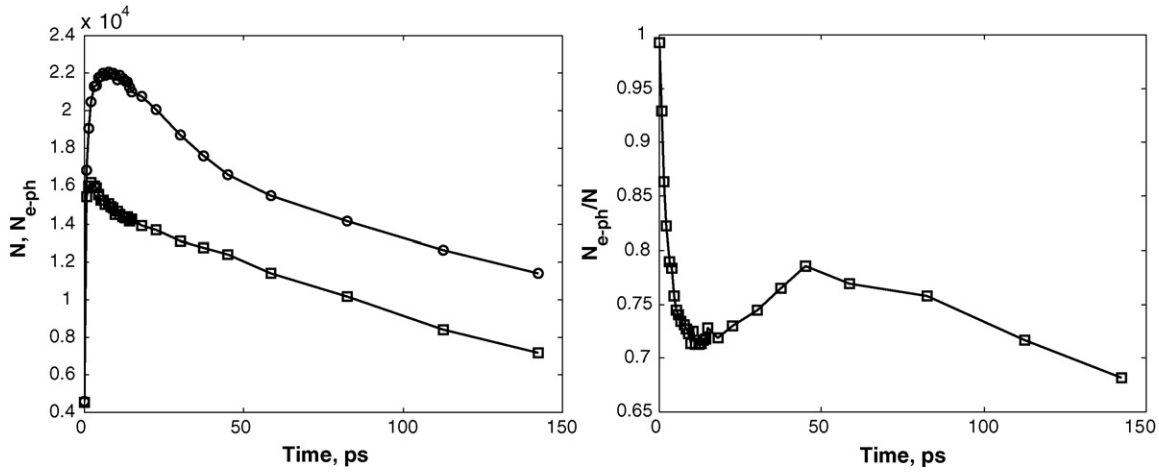

Fig. 5. (Left) Number of atoms in the melted (amorphous) region, the upper curve without EPC, the lower curve with EPC. (Right) Ratio of the number of atoms in the melted (amorphous) region for calculation with EPC to the number without EPC.

by electron–phonon coupling (EPC). According to Ref. [12], the EPC constant for Pu is $\lambda = 0.85$. Energy exchange between electronic and lattice subsystems is described by the equations [13]:

$$C_e \frac{dT_e}{dt} = -g(T_e - T_l); \qquad C_l \frac{dT_l}{dt} = g(T_e - T_l),$$

where C_e and C_l are electronic and lattice heat capacities, T_e and T_l are electronic and lattice temperatures, g is related to λ as $g = 3\hbar\gamma\lambda\langle\omega^2\rangle/\pi k_B$, here $\gamma = C_e/T$, and $\langle\omega^2\rangle$ is the second moment of the phonon spectrum. Taking γ and $\langle\omega^2\rangle$ from Ref. [12] gives $g = 4.5\times10^{17}$ W/K m^3. Since the characteristic time of the e–ph interaction is ~3 ps, taking EPC into account does not affect the fast MC stage. The equation of motion of atom i in MD is modified according to Ref. [14]:

$$M\frac{d^2\mathbf{r}_i}{dt^2} = -\nabla V(\{\mathbf{r}_j\}) - \frac{g}{c_a}\frac{T_a - T_e}{T_a} M\frac{d\mathbf{r}_i}{dt}$$

A new calculation of the cascade evolution was done in the assumption that $T_e = T_{init} = 600$ K, due to high electronic heat conductivity. It is seen from Fig. 3(right) that in this case the damaged region thermalizes in ~10 ps. The damaged region size as a function of time with and without EPC is plotted in Fig. 5(left). The size of the damaged region relative to that without EPC is presented in Fig. 5(right). The region grows during about 10–12 ps and 2–3 ps without and with EPC, respectively. After approximately 70 ps, the behavior in these two cases is quite similar. Further evolution occurs at 600 K.

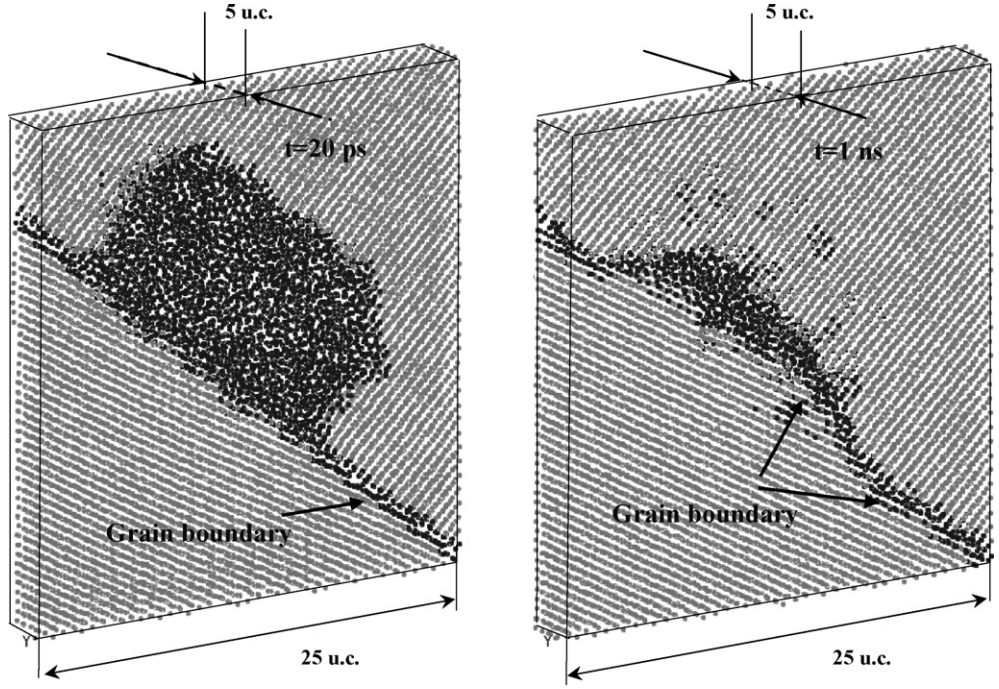

Fig. 6. Fragments of a polycrystalline sample containing grain boundary in the vicinity of the damaged region. Dark grey is for the disordered structure (grain boundary, melted region), light grey is for fcc δ-Pu.

So, the account for EPC gives a 30% reduction in the damaged region size. The finite value of electronic heat conductivity must slightly weaken the effect.

A number of calculations were also carried out for polycrystalline samples. If radiation melting occurs in a region containing a grain boundary, the recrystallization in the melted regions adjacent to different grains will differ. Such a process results in grain boundary sagging and broadening with time (see Fig. 6). The grain boundary also acts as a defect absorber. The number of interstitials dissolved in the grain boundary is higher than the corresponding number of vacancies. So, there is a disbalance between residual vacancies and interstitials.

4. Conclusion

The simulations have shown that the formation of the amorphous (melted) region is proper to the U cascade evolution in pure and alloyed Pu. Rapid recrystallization leaves point-like defects only. Dislocations created in high-rate straining also vanishes. It has been shown that accounting for only lattice heat conductivity leads to a melted region which measures ∼25,000 and ∼18,000 atoms for the initial temperatures of 600 and 300 K, respectively. If EPC is taken into account, the corresponding numbers are ∼17,000 and ∼12,000. Assuming that the melted region includes ∼12,000 atoms in the sample at the initial temperature of $T = 300$ K and the half-life period of Pu is 24,000 years, it is easy to estimate that it will take the whole sample about 2–3 years to completely melt and recrystalize.

Acknowledgment

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