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Computer simulation study of self irradiation in plutonium

Gérald Jomard [a,*], Lilian Berlu [b], Gaëlle Rosa [b], Philippe Faure [b], Jérôme Nadal [b], Nathalie Baclet [b]

[a] *CEA DIF/DPTA, F-91680 Bruyeres le Châtel, France*
[b] *CEA VA/DRMN, F-21120 Is sur Tille, France*

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Abstract

There is clear experimental evidence that plutonium based materials exhibit density changes with time. By comparison to what is known for nuclear fuel cell aging, it is believed that this phenomenom could be linked to the radioactive α-decay of plutonium. Schwartz et al. have identified three possible age-related phenomena due to self irradiation in Pu alloys that would cause dimensional changes: the initial transient, helium accumulation and void swelling [A.J. Schwartz, M.A. Wall, T.G. Zocco, W.G. Wolfer, Philos. Mag. 85 (2005) 479]. Even if the later phenomenon has not yet been observed in naturally aged Pu alloys, the aim of this work is to examine its possible occurrence by means of a multi-scale modelling approach. We coupled classical molecular dynamics simulations (MD) to mesoscopic Monte Carlo ones (MMC) in order to predict the long-term evolution of point defects created by self irradiation in plutonium. In this article, we focus on the results obtained for the MD radiation damage simulations. We show that plutonium does not seem to behave like other metals under ion irradiation. The annealing process of the defects produced by a recoil nucleus is indeed very long compared to what is known for various other metals. An MD parametric study of displacement cascade simulations combining temperature and cascade energy will be exposed. At the end, we will present results of preliminary MMC simulations based on our MD data which show that the spatial correlation of the stable defects populations created by the cascades seems to have a great influence on the predicted swelling.

Keywords: Pu; Radiation damage; Annealing; Molecular dynamics; Monte Carlo

1. Introduction

In all plutonium based materials, the α-decay of plutonium nuclei is responsible for the creation of numerous point defects in the crystal. The way these primary defects evolve may give rise to important changes in the structural properties of the material. As an example, swelling of plutonium alloys has been experimentally observed with various techniques (dilatometry and X-ray diffraction) [1–4]. But it is not clearly established yet if this phenomenon is linked to the radioactive property of plutonium. One reason is that the radioactivity and toxicity of plutonium are limiting drawbacks to extensive experimental measurements. Predicting the long-term effects of self irradiation in plutonium based materials is thus a real challenge for simulation. To reach this goal, we have developed a multi-scale approach based on classical MD simulations coupled to event-based Monte Carlo calculations. Obviously, there is a strong interplay between experiments and the theoretical approach in order to parametrize and validate the various models we used. This work is an extension of a previous study of damage production in Pu [12]. We had compared the development of 2 keV collision cascades in plutonium predicted by both the Embedded Atom Method (EAM) and the Modified EAM (MEAM) methods [11,6]. Since the MEAM formalism is the more reliable one to capture the unusual physical properties of Pu, we continued this study in this framework. In this paper, we will show that the predicted void swelling in plutonium is sensitive to the spatial configuration of the defects populations obtained by MD. Once again, we should mention that this part of our work is purely prospective since at this point, there is no experimental evidence that void swelling occurs in Pu-based materials.

2. Computational details

The MEAM formalism of Baskes has been implemented in our MD code STAMP which is designed for use on massively

* Corresponding author.
E-mail address: gerald.jomard@cea.fr (G. Jomard).

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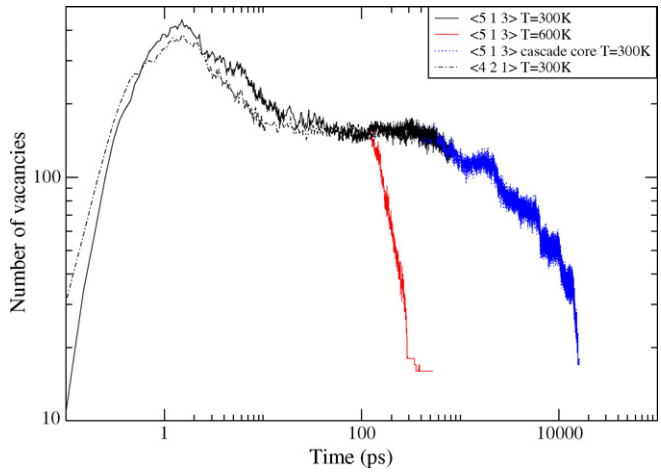

Fig. 1. This figure represents the number of vacancies created by the cascades vs time for a 2 keV PKA.

parallel machines [6]. An efficient parallelization based on MPI with a 3-D spatial decomposition scheme allows one to treat systems of millions of atoms. In order to simulate high energy cascades, we have modified the repulsive part of the original potential from Baskes to provide a better treatment of atomic interactions for small distances. For that purpose, we used the universal Kr–C potential of Wilson and coworkers [9], which is smoothly joined to the MEAM potential with a polynomial exponential of order 3 between 1 and 2 Å. More details on this procedure can be found elsewhere [10]. To model the defect production, we follow the usual procedure. An fcc crystal of plutonium is first equilibrated at $T = 300$ K in the statistical microcanonic thermodynamical ensemble (NVE). Then an atom located near the center of the simulation cell is chosen to play the role of the primary knock-on atom (PKA). A kinetic energy E_{PKA} is given in a desired crystallographic direction to this atom to model the emission of a recoil nucleus that initiates a collision cascade.

3. Defect production results

In order to provide reliable data to the mesoscopic models, we need a correct statistics on the predicted damage production. We have thus varied the PKA energy and its initial directions in our simulations. The $\langle 4, 2, 1\rangle$ and $\langle 5, 1, 3\rangle$ directions have been explored for 2 keV cascades, and the $\langle 4, 2, 1\rangle$, $\langle 5, 1, 3\rangle$, $\langle 10, 5, 2\rangle$ and $\langle 10, 2, 5\rangle$ ones have been studied for 10 keV cascades. Since the observed trends are very similar for all these simulations we will focus on the 2 keV cascades results. The evolution of the computed number of vacancies versus time is plotted in a log–log scale in Fig. 1.

We can see that the initial direction of the PKA has almost no effect on the defects production. The maximum number of Frenkel pairs created is more or less the same and this value is reached in both cases after 1.5 ps. For both directions, the development of the cascade leads to the formation of an amorphous zone in the crystal. It is surprisingly stable for many nanoseconds, the cooling stage is thus extremly long compared to what is known from other fcc metals (in Cu, Au and Pb for example, the cooling stage lasts only a few tens of picoseconds [13,14]). This is far from usual for a fcc metal and is more likely observed for semi-conductors materials [5]. As suggested by Valone et al., this phenomenon may be explained by the fact that the MEAM potential exhibits a lot of minima because of its ability to describe all the crystal structures of plutonium. Atoms can thus be frozen temporarily in amorphous-like atomic arrangements. This assumption should be tested by doing MD simulations with

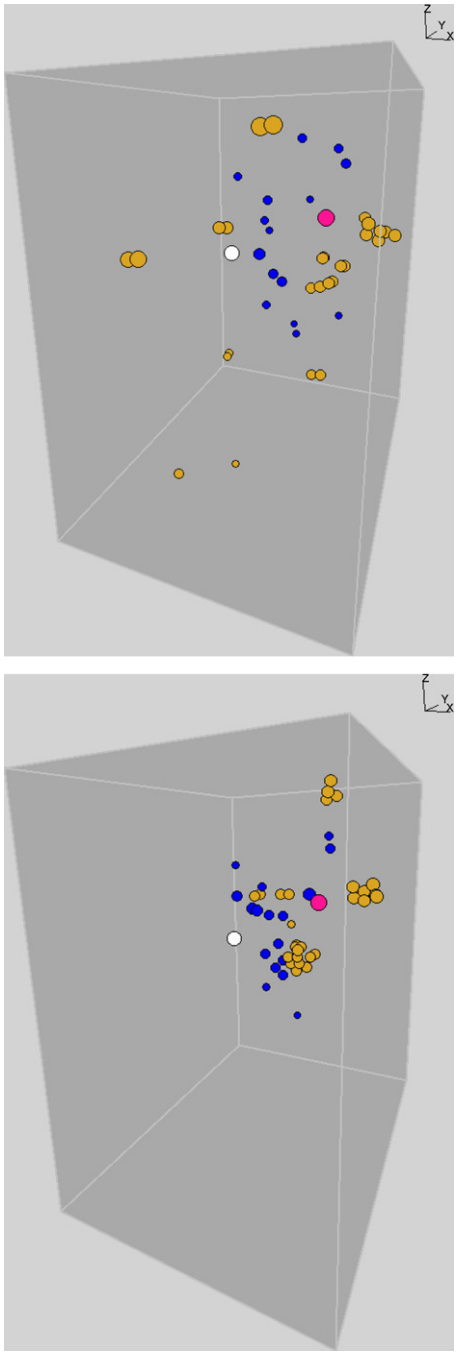

Fig. 2. Stable defect population for a 2 keV cascade simulated with the MEAM potential in the $\langle 5, 1, 3\rangle$ initial direction for the PKA. (Top) picture corresponds to $t = 300$ ps of the simulation where the cell is heated at $T = 600$ K. (Bottom) picture corresponds to $t = 15$ ns of the simulation at $T = 300$ K. Only vacancies (in blue), single insterstitials and dumbells (in orange) are represented.