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Computer Simulation of Helium Effects in Plutonium During the Aging Process of Self-Radiation Damage

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Abstract. Due to α radioactive decay Pu is vulnerable to aging. The behavior of He in Pu is the foundation for understanding Pu self-radiation damage aging. Molecular dynamics technique is performed to investigate the behavior of defects, the interaction between He and defects, the processes of initial nucleation and growth of He bubble and the dependence of He bubble on the macroscopical properties of Pu. Modified embedded atom method, Morse pair potential and the Lennard-Jones pair potential are used for describing the interactions of Pu-Pu, Pu-He and He-He, respectively. The main calculated results show that He atoms can combine with vacancies to form He-vacancy cluster (i.e., the precursor of He bubble) during the process of self-radiation as a result of high binding energy of an interstitial He atom to vacancy; He bubble's growth can be dominated by the mechanism of punching out of dislocation loop; the swelling induced by He bubble is very small; grain boundaries give rise to an energetically more favorable zone for the interstitial He atom and self-interstitial atom accumulation than for vacancy accumulation; the process of He release can be identified as the formation of release channel induced by the cracking of He bubble and surface structure.

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1 Introduction

Pu is perhaps the most complex metallic element known and has attracted extraordinary scientific interest since its discovery in 1941. It is of great technological importance due

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to its use in nuclear industry. However, modern day problems concerning Pu involve understanding and predicting the properties of Pu and its alloys for the safe handling, use and long-term storage of these important, but highly toxic materials which made experimental observations extremely difficult [1]. So, development of a predictive aging model for Pu is one of major goals of many researchers' works on Pu. Pu is vulnerable to aging because it is a radioactive element, decaying to U by emitting α particle. Although the widely used Pu-239 has a relatively long half-life of about 24,000 years, its decay rate is still sufficiently high to lead to a significant buildup of He and radiation damage within the metal after several decades [2]. As for the behavior of He atoms in Pu lattice, there may be a number of interactions occurring in the lattice. These interactions mainly include: 1) trapping and thermal detrapping of He atom in single vacancy, divacancies and vacancy clusters; 2) He atom trapping at dislocations and grain boundaries; 3) replacement of He atom bound to single vacancy by either interstitial Pu atoms or U atoms or other impurities; 4) He atom clustering into He-vacancy clusters and He bubble; 5) displacement of trapped He atoms by He-He or He-U collisions; and 6) diffusion of He atom as an interstitial [3]. Knowledge from Pu and other materials suffered from He and radiation damage has proved the degradation, or even worse, the invalidation of the material properties [4]. In fact, He effects in Pu are always associated with radiation damage effects and can be regarded as the focalization on probing the microstructural changes with ages.

Despite the central importance, there are still dearth of comprehensive studies in the literature concerning the complex He effects in Pu. Experimentally, transmission electron microscope (TEM), positron annihilation spectroscopy (PAS), X-ray diffraction (XRD), dilatometry, etc. have been used to probe into the He effects in Pu [5–7]. Although, these pioneering researches provide some valuable evidences on understanding the initial stage of He bubble nucleation and growth, many uncertainties still exist as a result of difficulties in experiments and lacking sufficient aged Pu samples. On the other hand, more and more theoretical modeling techniques have been performed to explore the atomistic mechanism of cascade damage effects during the last two decades [8, 9]. However, the atomistic behaviors of He effects have not been studied in detail with few exceptions, among them the classical works of Valone et al. and Dremov et al. who studied the stabilities of isolated He bubbles in δ Pu-Ga alloys by molecular dynamics simulation [10, 11]. They found that the stabilities of He bubble were strongly dependant on size, He-vacancy ratio and temperature. As for the macroscopical aspects of He bubble in Pu such as lattice swelling, material property changes, He bubble pressure and He release, there are few reports to the best of our knowledge.

In this article, we review our recent findings on He effects in Pu during the aging process of self-radiation damage by computer simulation technique, especially on investigating the microcosmic mechanism of initial nucleation and formation of He bubble. In addition, some macroscopical aspects of He bubble in Pu such as lattice swelling, He bubble pressure and He release process are also presented [12–14]. The remainder of the paper is organized as follows. In Section 2, the methods used are briefly described focus-