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LLNL-CONF-435984

Evidence of Abrupt Lattice Expansion in delta-Plutonium due to Self-Irradiation during the Aging Process

C. K. Saw, M. A. Wall, B. W. Chung

June 16, 2010

Plutonium Futures
Keystone, CO, United States
September 19, 2010 through September 23, 2010

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Cheng K. Saw, Mark A. Wall, and Brandon W. Chung

Lawrence Livermore National Laboratory
P.O. Box 808, Livermore, CA, 94551
saw1@llnl.gov

ABSTRACT

To better understand the plutonium material behavior caused by self-irradiation during the aging process, we examined the lattice behavior by an x-ray diffraction technique from a sample spiked with 7.5 weight percent of Pu-238 into Pu-239. Pu-238 is highly active component and this addition accelerates the aging process. The lattice damage comes predominantly from the uranium recoil resulting in Frenkel-type defects consisting of vacancies and self-interstitial atoms. We examine the changes in the crystallographic structure during the aging process by using an in-house transmission x-ray diffraction (XRD) technique over a period of 60 days, which is equivalent to 2.8 years for aging Pu-239. The sample was polished and etched to as small as 2 μm in thickness. We observed an abrupt lattice expansion of about 0.5% at roughly 1.1 equivalent years follow by atomic redistribution at the lattice sites. A model for the atomic process is proposed.

INTRODUCTION

We focus our investigation on understanding the structural behavior of material cause by self-irradiation. Pu-239 atoms transmutes by the α-decay process into uranium atom of 85 keV and α-particle of 5 MeV [1.2]. The recoil of the uranium atoms result in the lattice damage as Frenkel-type defect pairs made up of vacancies and self-interstitial atoms. It has also been reported that significant number of the interstitial atoms can also diffuse back into the lattice, a process known as "self-healing" however there is no information on the kinetics of this process.

We have setup an in-situ transmission XRD experiment to monitor structural changes from a very small and thinly prepared spiked Pu sample, similar to that used for TEM examination. The experimental setup has already been described earlier. Briefly, the CuK_{α} (~8 KeV) incident x-ray beam is defined to 0.2 mm size and a curved Position Sensitive Detector is used for data accumulation. There is no motor movement, thus avoiding the uncertainties in the scattering angle determination. Transmission technique is selected to probe the bulk of the sample rather than the surface, which can often be contaminated with oxides. By spiking with 7.5 wt% of the active isotope Pu-238 into the Pu-239 lattice, we accelerated the aging process by about 17 times the normal rate.

RESULTS AND DISCUSSION

At the start of the experiment, the sample is annealed at 300°C for an hour in order to remove any residue defects and lattice strains. We reported earlier [3] the observation of an abrupt lattice expansion of about 0.5 % in 22 days or 1.1 equivalent years. In this report, the peak width and intensity for the (111) reflection used to determined the lattice parameter is plotted in Figure 1 and Figure 2. Change in peak intensity is an indication of site occupancy and the change in peak width estimates the level of lattice distortion. There is no change in the peak width over the period of 60 days suggesting that the lattice ordering or distortion is not disturbed during the decay because the defects created at the lattice sites are not necessarily co-operative. It is possible that self healing occurs, however the number of atoms knocked off the lattice sites is greater than the atoms relaxing back as evidenced by the continuing reduction in x-ray peak intensity with time.

At roughly 22 days, or roughly 1.1 year in the normal sample, an abrupt expansion of 0.5% of the lattice can be observed. Interestingly, an increase in peak intensity can also be observed. These observations lead us to conclude that there is a threshold number of vacancies and interstitials that can be contained in the structure. Above this threshold free energy, there is an abrupt rearrangement of the interstitial atoms

into the vacancies along with an increase in lattice parameter. An increase in peak intensity is related to the self healing, where atoms from the interstitial collapse into the lattice defect sites, thus changing the occupancy of the sites. On further aging, the intensity continues to decrease, however the rate of this decrease is much lower than that before the expansion. This observation also suggests that rate of creating defects and interstitials is lower in this larger cell than the smaller cell.

1. W.G. WOLFER, *Los Alamos Sci.* 26 (2000) 274.
2. S.S. HECKER, J.C. MARTZ, *Los Alamos Sci.* 26 (2000) 238.
3. C.K. SAW, M.A. WALL, B.W. CHUNG, *MRS meeting* (Nov-2006)

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory in part under Contract W-7405-Eng-48 and in part under Contract DE-AC52-07NA27344.

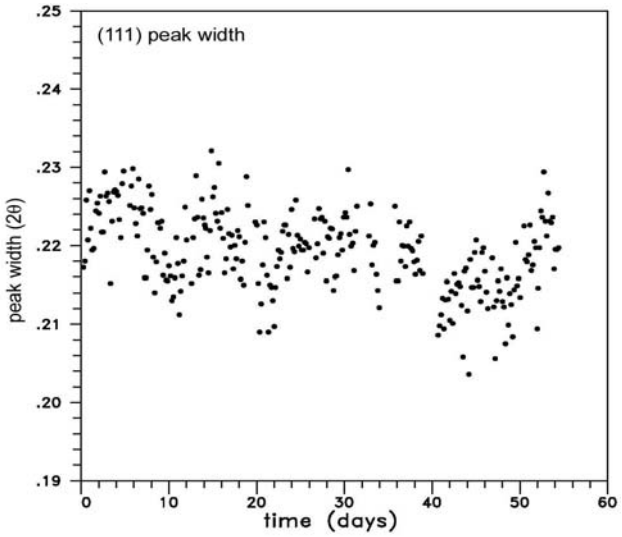

Figure 1: Peak width of (111) δ-Pu over 2.8 equivalent years

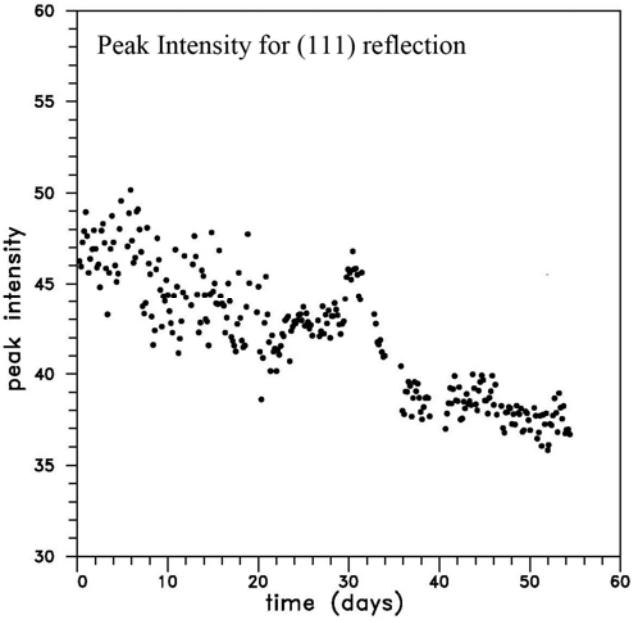

Figure 2: Changes in the (111) peak intensity over a period of 60 days