Formation and Recovery of Self-Irradiation and Metallurgical Changes in Plutonium Alloys

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Summary: This paper describes efforts to better understand the volume expansion in the early stage of aging and the recovery of metallurgical properties from accumulated age-induced defects in plutonium alloys. This work reveals higher complexity of interplay between lattice damage and in-grown radiogenic helium that requires further work.

Introduction

The plutonium alpha-decay leads to the formation and accumulation of numerous defects and transmutations, which affect metallurgical properties [1]. During the past decade, a significant progress has been made in understanding the mechanisms of aging and its induced effects in metallurgical properties [2]. There remain, however, a number of outstanding questions regarding the fundamental aging process, particularly to better understand the fundamentals of the volume expansion in the early stage of aging and the recovery of metallurgical properties from accumulated age-induced defects. Here, two subjects will be discussed. First, discussion will be on evolving metallurgical properties of plutonium alloys with age using accelerated and naturally aged plutonium alloys. Experiments show increasing static strength, gradual swelling in volume, and decrease in density with aging. Second, an investigation to measure the effect of heat treatment on aged plutonium alloys will be discussed. By measuring formation and recovery of self-irradiation damage as a function of temperature and time, we can further quantify metallurgical changes induced by either the lattice damage or radiogenic helium in-growth in aged plutonium alloys. Upon heating above ~150C, results indicate some recovery from aging induced changes in metallurgical properties. These measurements indicate complexity of potential interplay between accumulated disorders and in-grown helium with age and annealing process.

Results and Discussion

Small volumetric expansion with aging has been detected in accelerated plutonium alloys by measurements of specimen length [3]. The initial volumetric expansion is followed by an extended linear region with a small positive slope. The initial amount of swelling is attributed to the number of Frenkel pairs that survive the radiation damage and subsequent self annealing processes. The progressive accumulation of survivor vacancies provides an increasing number of alternative sites for the capture of self-interstitial plutonium atoms. As the density of these alternative sites increases, the rate of swelling is thereby reduced. After the initial expansion, the extended linear expansion behavior is attributed mostly to a constant helium in-growth rate of ~41 appm (atomic parts per million) per year. After extended aging past 60 equivalent years, the volume expansion behavior deviates from the earlier linear trend and show near saturation-like behavior. This behavior indicates potential higher order complexity in disorder in damaged plutonium than the simple accumulation of Frenkel pair and radiogenic helium bubbles. There is also a hypothesis suggesting that small precipitates of the higher density ζ-Pu_3Ga phase form in the matrix [4]. Upon annealing above ~150C, aged plutonium alloys shows some recovery from age-induced changes but the amount of initial expansion is smaller and onset of the saturation-like behavior is much earlier (Figure 1). In addition, quasi-static mechanical tests shows drop in tensile strength when annealed indicating the annealing out of the accumulated lattice damage.

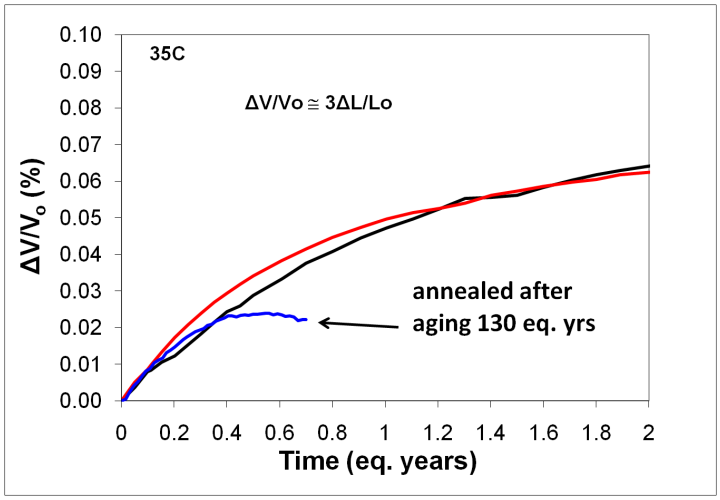

Figure 1. The relative volume change measured by dilatometry with the equivalent age of plutonium alloys. Annealing aged alloy leads to smaller initial expansion.

Conclusion

To summarize, both dilatometry and quasi-static strength measurements are used to measure age-induced changes in metallurgical properties of plutonium alloys. Results show that the simple accumulation of Frenkel-pair defects and radiogenic helium bubbles does not explain the observed behavior at extended aging. Annealing experiments indicate complexity of potential interplay between accumulated disorder and in-grown helium in aged plutonium alloys. This work reveals that we need better understanding of integrated contribution from several self-irradiation induced defects to plutonium alloys.

References

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