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Evolution of Static Physical Properties in Plutonium by Self-irradiation Damage

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ABSTRACT

The alpha-decay of plutonium leads to the age-related change in physical properties. This paper presents updated results of age-related effects on enriched and reference alloys measured from immersion density, dilatometry, and mechanical tests. After nearly 100 equivalent years of aging, both the immersion density and dilatometry show that the enriched alloys are decreasing in density by less than 0.002% per year and now exhibit a near linear density decrease, without void swelling. The tensile tests show that the aging process increases the strength of plutonium alloys, followed by possible saturation past 70 equivalent years of age. The ultimate goal of this work is to develop capabilities to predict physical properties changed by aging effects.

INTRODUCTION

Plutonium exhibits notoriously complicated metallurgical behaviors, depending sensitively on phase as well as on chemical content and microstructure [1, 2]. Current studies in plutonium metallurgy are motivated by the need to better understand the influence of the metallurgical phenomena on the physical properties for stockpile stewardship, nonproliferation, environmental issues, and nuclear power. One of the key areas of research is developing capabilities to predict physical properties changed by the radioactive decay of plutonium that incessantly creates lattice damage and in-growth of radiogenic helium. Because these integrated aging effects would normally require decades to measure, studies are underway to assess the effects of extended aging on the physical and static mechanical properties of plutonium alloys by incorporating roughly 7.3 atomic % of highly specific activity isotope ^{238}Pu into the ^{239}Pu metal to accelerate the aging process. By monitoring the properties of the ^{238}Pu enriched alloy and naturally aged plutonium alloys, the aging properties of plutonium from the self-irradiation damage can be predicted.

EXPERIMENT

Radiation damage from alpha decay in plutonium occurs at a rate of ~0.1 dpa (displacement per atom) per year. Because the effects of interest occur over decades, our approach is to accelerate the effects of radiation damage in plutonium metal by incorporating 7.3 atomic % of the higher specific activity isotope ^{238}Pu into the ^{239}Pu lattice. The rate of alpha-decay of ^{238}Pu is nearly 300 times that of ^{239}Pu so the rate of radiation damage accumulation can be increased. Using this method, the radiation damage in plutonium equivalent to sixty years of natural aging can be simulated in only a few years. Additional details of sample preparation are presented elsewhere [3]. In addition, naturally aged plutonium alloys of various ages are characterized to validate the accelerated aging approach.

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Details of operation of the dilatometer system, immersion density, and static tensile test techniques are presented elsewhere [3], so only a brief description is provided here. Specifically designed dilatometers, immersion density equipment, and a static tensile tester were set up inside a nitrogen atmosphere glovebox. The dilatometer is designed to monitor long-term growth resulting from the lattice damage and helium in-growth in plutonium alloys. The immersion density equipment closely matches a design used by Bowman et al. [4] and uses about 200 ml of Fluorinert Electronic Liquid FC-43 as the immersion fluid. The tensile tester has a specially designed fixture for testing plutonium samples. With a 6.1 mm GL extensiometer for the strain measurement, typical testing was performed at crosshead speed of 1.27 mm/min, so that the ultimate strain rate was about 3.5 x 10^{-3}/sec.

DISCUSSION

Three principal aging mechanisms have been identified as a result of the self-irradiation of plutonium that would cause changes in physical properties: the initial transient; accumulation of radiogenic helium and actinide daughter products; and void swelling. The initial transient saturates after a short time and results mainly from the lattice damage. The second contributor to the change is the build-up of helium and actinide daughters from the radioactive decay of plutonium. The void swelling is another phenomenon, but has not yet been observed in aged plutonium alloys.

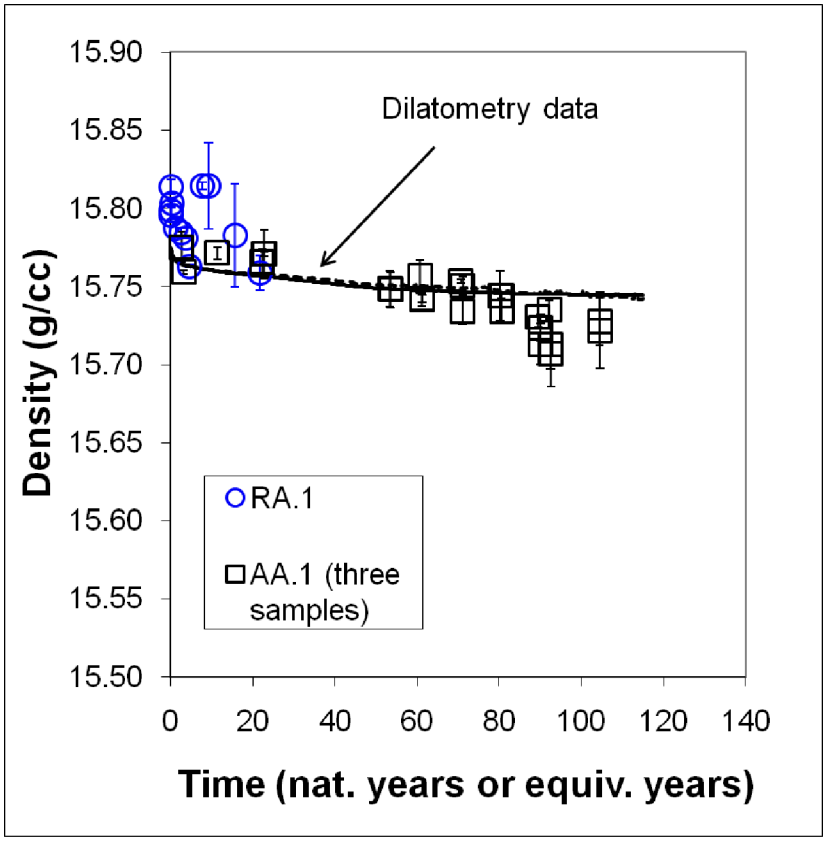

Figure 1. Comparison of density changes between enriched alloys from the dilatometry to naturally aged and enriched alloys from the immersion density. Both dotted and solid lines are dilatometry data converted to equivalent density data. Circles and squares are immersion density data measured from naturally aged (RA.1) and enriched (AA.1) alloys, respectively. Both dilatometry and immersion density measurements show decrease in the density of plutonium alloys by the aging process.

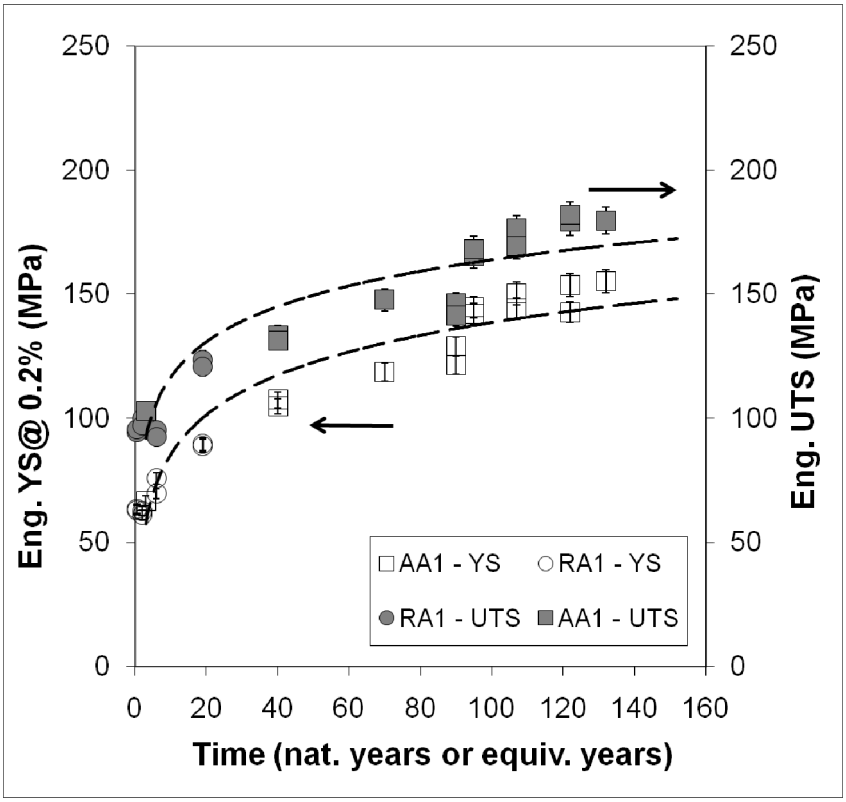

Figure 2. Engineering yield strength (YS) and ultimate tensile strength (UTS) of plutonium alloys from aging. Circles are naturally aged alloys (RA1) and squares are enriched alloys (AA1).

Results from dilatometry, immersion density and tensile measurements show effects from the first two mechanisms on plutonium alloys with ~2 atomic % Ga (see Figures 1 and 2). These techniques are well suited in measuring small property changes produced by aging mechanisms. Results indicate that these plutonium alloys undergo small changes in properties with time, without any signs of void swelling.

Our measurements show initial decrease in the density and increase in the tensile strength in plutonium alloys by aging. These initial changes in properties saturate in a few years and mostly due to initial increase in the number of Frenkel pairs that quickly attain stationary values. After this initial transient stage, the rate of change in each property becomes reduced. The continuous build-up of helium in-growth in the form of bubble and actinide daughter products leads to continued changes in both density and tensile properties.

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Aged plutonium alloys exhibit a drop in tensile strength when annealed to 300°C indicating the annealing out of the accumulated lattice damage. Current annealing experiments show reduction in the engineering yield strength by ~30 MPa from ~177 MPa on enriched alloys doped with ~3 atomic % Ga (and aged to ~90 equivalent years). This reduction appears to be related to the annealing out the accumulated lattice damage from aged plutonium alloys. We estimate the in-growth of helium contributes ~70 MPa for this alloy aged to ~90 equivalent years.

CONCLUSIONS

We have developed analytical techniques to measure small changes in plutonium properties by aging. Results of measurements show evolving physical properties of plutonium alloys from incessant self-irradiation damage. So far, however, void swelling has not been observed. Annealing recovery experiments are also under way to better understand aging mechanisms responsible of evolving physical properties caused by self-irradiation damage.

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