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Evolution of defects in Pu during isochronal annealing and self irradiation

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We report on the evolution of defects in Pu during isochronal annealing and self irradiation using positron annihilation spectroscopy. Positron annihilation spectroscopy is a sensitive probe (part per million level) for atomic-scale defects. The spectroscopic tools available at LLNL allow the determination of size, concentration, and chemical surroundings of defects in aged Pu samples.

Positron lifetime analysis was performed on eight samples aged 7 months to 42 years. All samples except the 7-month old sample contained a high concentration of positron trapping centers. The dominant component yielded a lifetime value of ~ 182 ps. In aged samples, a second longer lifetime component was observed that appears to increase in strength with the age of the sample. The observed lifetime values and their relative strengths are shown in figure 1. The top panel corresponds to the lifetime values and the bottom panel corresponds to the intensity of the long lifetime component.

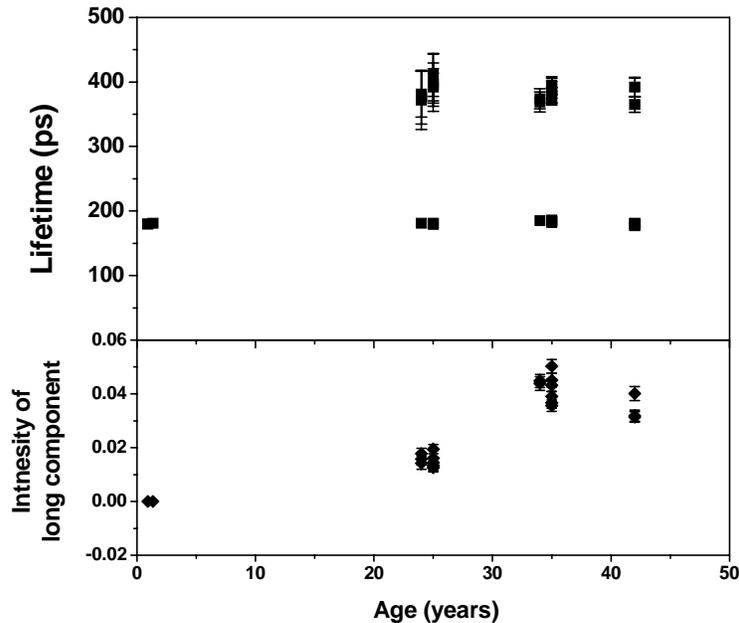


Figure 1. Positron lifetime values and relative intensity of the long lifetime component in aged Pu material. The intensity of the lifetime components sums to unity. The data at ~ 1 year is from melted and recast material.

Positron lifetime values are determined by the bubble size and He content. When He is added to a bubble, the positron lifetime is shortened due to the increased electron density. When the size of the bubble is known from an independent measurement, the observed positron lifetime values and the

associated first principle calculations can be used to estimate the He content of the bubble. Our positron results can be combined with the recent transmission electron microscopy measurements of Schwartz *et al* to estimate the He content of the bubbles. The short lifetime component corresponds to 2-3 He atoms per vacancy.

A 35-year old sample was annealed to 440 °C in several steps to examine the temperature stability of the two lifetime features. The isochronal anneals were performed for 30 minutes, increasing the temperature with each subsequent step. This measurement complements a previous annealing study on recast, 7-month old material. The primary objective of these studies is to establish the appropriate limits on the storage temperature of the accelerated aging samples. Figure 2 shows the intensity of the long lifetime component as a function of the annealing temperature. Also shown is the corresponding He evolution from the capsule measured using a standard He leak detector. The intensity of the long lifetime component starts to increase at about 150 °C and reaches a maximum at around 250 °C. A similar behavior is seen in He evolution. The storage temperature of the accelerated aging sample should be below the onset of these annealing effects.

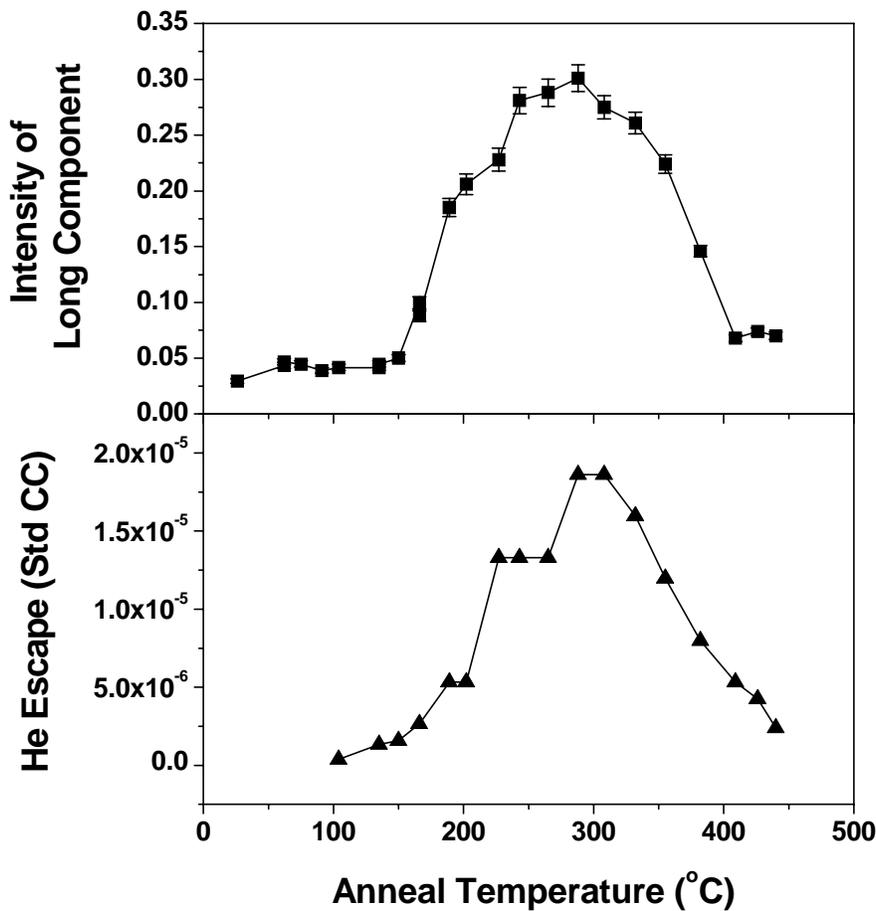


Figure 2. Intensity of the long lifetime component and He evolution of a 35-year old sample as a function of the isochronal annealing temperature. Each anneal lasted for 30 minutes and the measurements are performed after cooling the sample to room temperature.

We also report results from a new spectroscopic method that examines chemical surroundings of the bubbles observed in aged Pu material. The new spectroscopy determines the momentum distribution of the electrons participating in the annihilation process. At high momentum values these distributions are dominated by electrons bound to atoms and is therefore element specific. No significant variations are observed in aged samples, suggesting that the chemical surroundings of voids and bubbles are not evolving with age.

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