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Theory of Positron Annihilation in Helium-filled Bubbles in Plutonium

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Theory of positron annihilation in helium-filled bubbles in plutonium

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Positron annihilation lifetime spectroscopy is a sensitive probe of vacancies and voids in materials[1]. This non-destructive measurement technique can identify the presence of specific defects in materials at the part-per-million level. Recent experiments by Asoka-Kumar et al.[2] have identified two lifetime components in aged plutonium samples – a dominant lifetime component of around 182 ps and a longer lifetime component of around 350-400ps. This second component appears to increase with the age of the sample, and accounts for only about 5 percent of the total intensity in 35 year-old plutonium samples.

First-principles calculations of positron lifetimes are now used extensively to guide the interpretation of positron lifetime data[3]. At Livermore, we have developed a first-principles finite-element-based method for calculating positron lifetimes for defects in metals[4]. This method is capable of treating system cell sizes of several thousand atoms, allowing us to model defects in plutonium ranging in size from a mono-vacancy to helium-filled bubbles of over 1 nm in diameter.

In order to identify the defects that account for the observed lifetime values, we have performed positron lifetime calculations for a set of vacancies, vacancy clusters, and helium-filled vacancy clusters in delta-plutonium. The calculations produced values of 143ps for defect-free delta-Pu and 255ps for a mono-vacancy in Pu, both of which are inconsistent with the dominant experimental lifetime component of 182ps. Larger vacancy clusters have even longer lifetimes. The observed positron lifetime is significantly shorter than the calculated lifetimes for mono-vacancies and larger vacancy clusters, indicating that open vacancy clusters are not the dominant defect in the aged plutonium samples.

When helium atoms are introduced into the vacancy cluster, the positron lifetime is reduced due to the increased density of electrons available for annihilation. For a mono-vacancy in Pu containing one helium atom, the calculated lifetime is 190 ps, while a di-vacancy containing two helium atoms has a positron lifetime of 205 ps. In general, increasing the helium density in a vacancy cluster or He-filled bubble reduces the positron lifetime, so that the same lifetime value can arise from a range of vacancy cluster sizes with different helium densities.

In order to understand the variation of positron lifetime with vacancy cluster size and helium density in the defect, we have performed over 60 positron lifetime calculations with vacancy cluster sizes ranging from 1 to 55 vacancies and helium densities ranging from zero to five helium atoms per vacancy. The results indicate that the experimental lifetime of 182 ps is consistent with the theoretical value of 190 ps for a mono-vacancy with a single helium atom, but that slightly better agreement is obtained for larger clusters of 6 or more vacancies containing 2-3 helium atoms per vacancy. For larger vacancy clusters with diameters of about 3-5 nm or more, the annihilation with helium electrons dominates the positron annihilation rate; the observed lifetime of 180ps is then consistent with a helium concentration in the range of 3 to 3.5 He/vacancy, setting an upper bound on the helium concentration in the vacancy clusters. In practice, the single lifetime component is most probably associated with a family of helium-filled bubbles rather than with a specific unique defect size.

The longer 350-400ps lifetime component is consistent with a relatively narrow range of defect sizes and He concentration. At zero He concentration, the lifetime values are matched by small vacancy clusters containing 6-12 vacancies. With increasing vacancy cluster size, a small amount of He is required to keep the lifetime in the 350-400 ps range, until the value saturates for larger helium bubbles of more than 50 vacancies (bubble diameter > 1.3 nm) at a helium concentration close to 1 He/vacancy.

These results, taken together with the experimental data, indicate that the features observed in TEM data by Schwartz et al are not voids, but are in fact helium-filled bubbles with a helium pressure of around 2-3 helium atoms per vacancy, depending on the bubble size. This is consistent with the conclusions of recently developed models of He-bubble growth in aged plutonium.

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References

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