ALPHA-DECAY-INDUCED DAMAGE AND RECOVERY IN FUEL-LIKE ACTINIDE DIOXIDES

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The significant structural stability of 12 $^{238}PuO_2$, $^{239}PuO_2$, $^{240}PuO_2$, $^{241}AmO_2$ and $^{244}CmO_2$ samples and 14 fuel-like (U,Pu)O_2, (U,^{241}Am)O_2, (Pu,^{241}Am)O_2 and (Pu,^{244}Cm)O_2 samples under self α decay is explored by a radiation annealing model [1]. At ambient temperature, following exposure of ~0.2 dpa (displacements per atom), lattice expansion is limited to $\Delta V/V = 0.0090\pm0.0009$. This behavior indicates limited accumulation of Ac (actinide) and O Fps (Frenkel pairs), and thereby suggests the importance of a steady-state of damage and recovery.

In our model, event-by-event production of Ac and O Fps is combined with prompt removal of randomly-occurring closely-spaced interstitial-vacancy pairs. Recombination is controlled by critical annihilation volumes, ν_{ac} and ν_o. Guided defect mobility is generated by electronic and mechanical distortions. The fractional volume expansion is given by $\Delta V/V = [\Omega_{ac}/\nu_{ac} + \Omega_o/\nu_o] \exp(-w\lambda Nt)$. Here, Ω_{ac} and Ω_o represent volume increases per surviving Ac and O Fp, N is the density of the Ac atoms, λ is the weighted average decay constant of an atom, and t is the exposure time. This expression reproduces all experimental data.

Our MD simulations on UO_2 at <5K [2], and two more recent similar computations [3,4], reveal that interstitial (or vacancy) in the O sublattice recombine promptly with its 1st or 2nd closest vacancy (or interstitial). Therefore, the joint crystal volume that is occupied by a stable pair of non-overlapping O interstitial and vacancy is ν_o=1.218 nm^3. Thus, apparently the density of surviving O Fps at defect saturation is ν_o^{-1} = 0.82 nm^{-3}. Some close U interstitials and vacancies also recombine promptly. However, there is evidence that U Fps are efficiently eliminated via thermal diffusion. Thus, our model yields Ω_{Θ}=0.0110±0.0018 nm^3. This result coincides with high values of this parameter as derived by static computations [5], and is close to 0.0137 nm^3 as derived by the elastic continuum model [6].

Briefly, the mean volume damaged per α-decay event is w=490±80 nm^3 (~3,000 unit cells). The simple inverse exponential dependence of ΔV/V on time indicates that defect saturation is achieved even in a first-exposed lattice region. Under any large number of Fps that are created per α-decay event, assumed ~1,500, about 400 O Fps survive.

We find that the stability of the AcO_2 lattice is independent of the composition, chemical properties and the decay half-lives of the actinide cations. These properties are an advantage in the context of safe use of dvanced complex AcO_2 reactor fuels as well as direct disposal of burnt reactor fuels. Long-term stability is assured by the presence of ancient highly crystalline uraninite and thorianite [7].

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