Cover letter

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Dear Sir or Madam,

We wish to submit an original research article entitled “Modeling oxygen-void interactions in uranium nitride” for consideration by the Journal of Nuclear Materials.

We confirm that this work is original and has not been published elsewhere, nor is it currently under consideration for publication elsewhere.

Oxygen impurities in uranium nitride (UN) are reported to influence its swelling behavior under irradiation, yet the underlying mechanism remains unknown. In this work, we develop a first-principles model that quantifies the interaction of oxygen with voids and fission gas bubbles in UN, leading to a reduction in surface energy that can promote swelling. The analysis reveals that segregation of substitutional oxygen at surface nitrogen sites is the primary driver of surface energy reduction, $|\Delta\sigma|$, while oxygen in surface hollow sites plays a minor and sometimes counteracting role. $|\Delta\sigma|$ is most pronounced for small cavities (R_v = 1–10 nm) at intermediate temperatures that coincide with the onset of breakaway swelling in UN. Larger voids require higher temperatures for oxygen adsorption to significantly lower their surface energy. The temperature dependence of $|\Delta\sigma|$ exhibits three regimes: negligible reduction at low temperatures due to sluggish oxygen diffusion, a maximum at intermediate temperatures where oxygen incorporation is optimal, and a decline at high temperatures due to enhanced bulk solubility. A parametric analysis reveals that $|\Delta\sigma|$ depends strongly on both oxygen concentration and cavity size, but is largely insensitive to porosity. Our results suggest that oxygen-induced surface energy reduction is essential for reconciling the mechanistic swelling model of UN with experimental observations.

We have no conflicts of interest to disclose.

Please address all correspondence concerning this manuscript to bwbeeler@ncsu.edu (Benjamin Beeler) and cooper_m@lanl.gov (Michael W.D. Cooper).

Thank you for your consideration of this manuscript.

Sincerely,
Mohamed AbdulHameed