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Nanoscale Spatially Resolved Mapping of Uranium Enrichment in Actinide-Bearing Materials

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Uranium (U) isotopes are central to a diverse set of scientific disciplines, most notably earth and planetary sciences, toxicology, environmental monitoring and bioremediation, and the nuclear fuel cycle, forensics, and safeguards [1-4]. Specifically in nuclear fuel cycle applications, the amount of the fissionable U isotope (^{235}U) relative to all U (defined as enrichment) is a critical parameter in fuel performance, and thus impacts economic viability of nuclear power. The distribution of ^{235}U in a nuclear fuel before irradiation can directly influence the nucleation and distribution of fission products and fission gas bubbles leading to impact on fuel swelling kinetics [5]. This make it critical to analyze the distribution of ^{235}U in a nuclear fuel at a high spatial resolution to account for ^{235}U enrichment variation across all possible nanoscale heterogenities in the microstructure.

The small volume (nanometers to micrometers in diameter) of precipitates or interfacial regions that must be analyzed limits the ability of many bulk analysis techniques currently used for quantifying ^{235}U enrichment, introducing a crucial technological and resultant knowledge gap. Our work aims to address this gap, while also demonstrating a methodology by which ^{235}U isotopic abundances in actinide-bearing materials can be measured quantitatively with sub-nanometer scale spatial resolution to gain uniquely powerful insight into material radioactivity, origin, or processing history.

Here, we demonstrate unprecidented nanoscale mapping of U isotopic enrichment with high sensitivity across various microstructural interfaces within small volumes (100 nm^3) of depleted, low, and high enriched U with different nominal enrichments of 0.20, 19.75, and 90% ^{235}U respectively. Results from APT analysis are summarized in Figure 1, where element distribution maps for U isotopes, and the portion of the mass spectra illustrating the main U peak is presented.

Results indicate that uranium carbide (UC) inclusions have a similar enrichment to the surrounding matrix, and are thus likely formed during material processing. We also find that the most accurate way to quantify the enrichment is by using the main U peak (U^{3+} charge state), and the ion counts for each isotope, due to the presence of strong U hydride peaks associated with the U^{2+} charge state as shown in Figure 2.

The approach presented here can be applied to study nanoscale variations of isotopic abundances in the broad class of actinide-bearing materials, providing unique insights into their origin and thermo-mechanical processing routes [6].

References:

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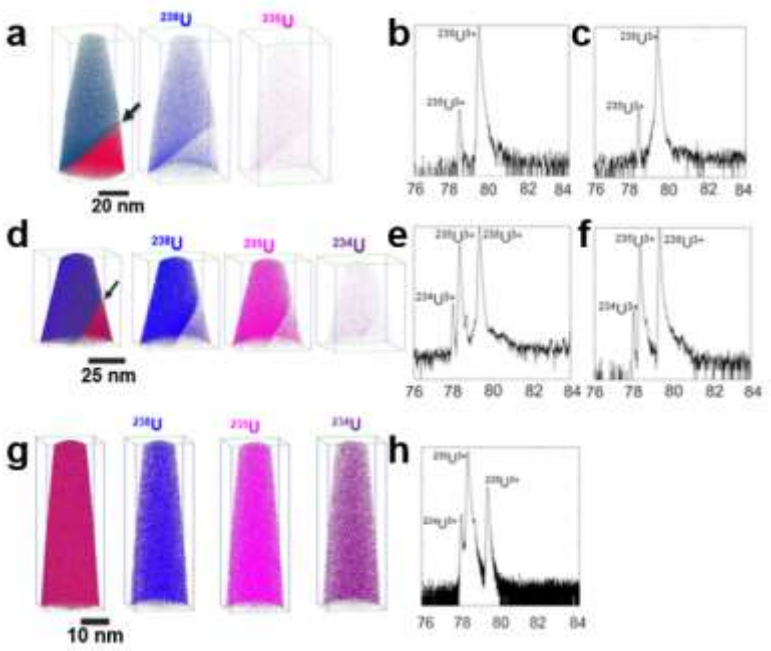

Figure 1. Summary of results from APT analysis of depleted, low, and highly enriched uranium, where sub-figures (a,b,c) are depleted U, (d,e,f) are LEU, and (g,h) are HEU.

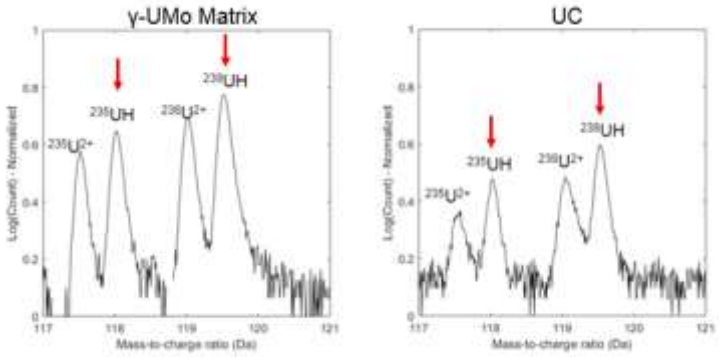

Figure 2. Example of U hydride peaks in the mass spectra of low-enriched U matrix and UC phases.

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