Contribution ID: **51** Type: **Oral**

LG-SIMS oxygen isotope and impurity distribution characterization of U-O-bearing particles as signatures of process history

Abstract: Oxygen isotopes and impurities may provide nuclear forensic signatures related to processing of uranium materials. These signatures can be evaluated by secondary ion mass spectrometry (SIMS). Here we describe such SIMS characterization of U-O-bearing materials and discuss the need for reference materials to understand signatures from real materials.

SIMS oxygen isotope characterization: The various chemical reactions for converting uranium ore concentrates (UOCs) to desired species (e.g. UO2, U3O8, etc.) can fractionate oxygen isotopes in potentially distinguishable ways. Oxygen from the local environment may also impart their isotope signatures during processing. Bulk analysis of U-O-bearing materials can provide 18O/16O ratios with a high-level of precision for evaluating process histories. SIMS can complement bulk oxygen isotope data by providing additional signatures related to isotope homogeneity at micron-level scales (e.g. particles from swipes, small fragments of fuel pellets). The isotope homogeneity or heterogeneity of a material may be related to (1) equilibrium kinetics specific to processing reactions and/or (2) the presence or lack of multiple endmember oxygen isotope sources. Figure 1 shows example SIMS particle data from New Brunswick Laboratory certified reference material (CRM) 129-A (U3O8) and two UOCs, one consisting of U3O8 and one consisting of uranyl peroxide (we note the instrument was calibrated to produce a mean δ18O value of 0‰ for CRM 129-A). Here, the two U3O8 samples have resolvable mean δ18O values, and the uranyl peroxide sample has a lower particle-to-particle variability when compared to the U3O8 sample datasets. These characteristics likely reflect the specific process histories of each material.

Mapping impurity distributions by SIMS: Distributions of impurities within U-O-bearing materials may be related to process history, and can be revealed through SIMS isotope mapping. Figure 2a shows an example of O, F, Cl, and U isotope signals from UO3 particles (sample 1). In this case, the F and Cl impurities are co-located with the uranium and oxygen signals from the particles. Figure 2b shows a three isotope plot (19F/16O vs. 35Cl/16O) of (1) sample 1 UO3 particle data (black data; left side of the plot); and (2) particle data from a second sample consisting of U3O8 material (maroon and green data; right side of the plot. Whereas the UO3 data are tightly clustered, indicating co-location of impurities with the U-O-bearing particles, the U3O8 material shows three clusters of data, including a high F residue and a high Cl residue. These impurity distribution differences are likely due low temperature processing that didn't separate F and Cl from U (e.g. the UO3 sample) compared to high-temperature sintering that separated U, F, and Cl (e.g. the U3O8 sample).

The need for reference materials: In order to interpret oxygen isotope and impurity distributions of real materials, it is vital to have reference materials with known process histories that collectively represent a meaningful parameter space of material processing. In part, this can be achieved though laboratory synthesis of materials through various chemical reactions and conditions. Subsequent characterization of these materials can then trace oxygen isotope ratios and impurity distributions back to how the material was synthesized, to better understand signatures of real materials.

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Session Classification: Oral Session #7 – Signature Research on Isotopic Signatures and Age-Dating

Track Classification: 1. Nuclear Forensics Capability Building: Initiation and Sustainability: 1.2 New Technologies, R&D and Signature Research in Nuclear Forensics