Analysis of mixed U/Pu materials

with Large-Geometry SIMS

U/Pu within nuclear debris

Accurate characterization of actinide ratios can provide valuable information for safeguards and other nuclear applications. Glassy nuclear debris forms during the dynamic interplay between the fireball from a nuclear detonation and the Earth's surface. Secondary Ion Mass Spectrometry (SIMS) is one of the few techniques that can resolve micron scale elemental and isotopic heterogeneities in such materials. In this study, dynamic multicollector Large-Geometry SIMS (LG-SIMS) was applied to the measurement of U/Pu within individual debris pieces. Recent development in LG-SIMS analysis software enables the measurement protocol described here and opens opportunities for the concurrent measurement of other multi-isotope systems.

Innovative dynamic multicollector mode

For small or inhomogeneous materials, U and Pu isotopes must be measured together using the multicollector system for adequate counting statistics and optimized accuracy and precision. While U isotopic measurements are established, the concurrent addition of Pu is challenging because the difference in mass dispersion between U and Pu causes misalignment of the Pu isotopes on the detectors (Fig. 1a).

Using the LG-SIMS multicollection Optional Instrument Parameters (OIP) software option, mass dispersion can be adjusted and signals aligned on the detectors for each isotope system (Fig. 1b). For each magnet jump (mass dispersion setting) the peak shape is optimized by adjusting additional coupling parameters.

Relative Sensitivity Factor

One challenge of accurate quantification between interelement ratios is the difference in ionization efficiency for each element. A relative sensitivity factor (RSF) is used to correct for this difference. Uranium- and plutonium-doped glasses of known U/Pu were measured and compared to the known value to calculate a $RSF_{U:Pu}$ of ~2.3, similar to other published values.

Fine scale U/Pu measurements

The U/Pu ratio was measured within individual debris from the Trinity nuclear test. U/Pu ranges from 4 to 143, with a maximum uncertainty of ± 0.34% (Fig. 2). SIMS microanalysis reveals a large spatial range in U/Pu that would be averaged by bulk analysis.

The innovative method demonstrated here can be utilized for other materials and isotope systems and is therefore of high interest to nuclear forensics, safeguards, and geochemistry communities.

Data collected on IMS 1280-HR at Pacific Northwest National Laboratory (PNNL), USA. Courtesy of Mindy Zimmer and Heather Cunningham, PNNL-SA-151803 (US).

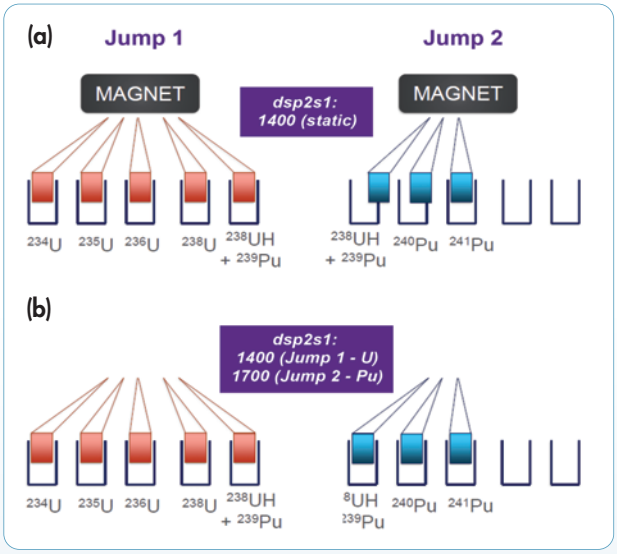


Figure 1: U and Pu isotopes measured in LG-SIMS dynamic multicollection mode.
a) Difference in mass dispersion for isotopes of U and Pu.
b) Using the OIP software option, the mass dispersion is optimized for each isotope system / magnet jump by adjusting the dsp2s1 stigmator after the magnet.

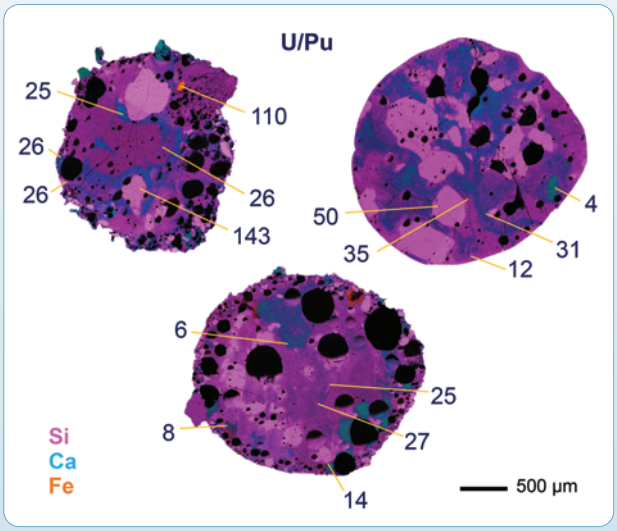
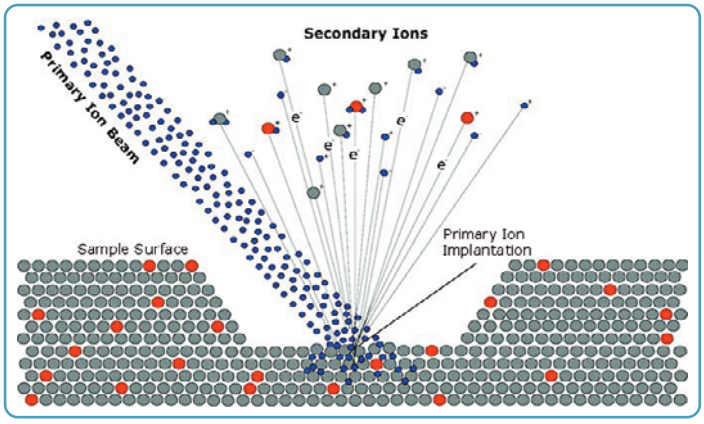


Figure 2: Backscatter electron images of three pieces of glassy nuclear debris from the Trinity test, showing the U/Pu ratio obtained at different locations of the sample. LG-SIMS dynamic multicollector mode with OIP option (Fig. 1b). Spot size 25 µm.

The Technique Behind



Dynamic SIMS

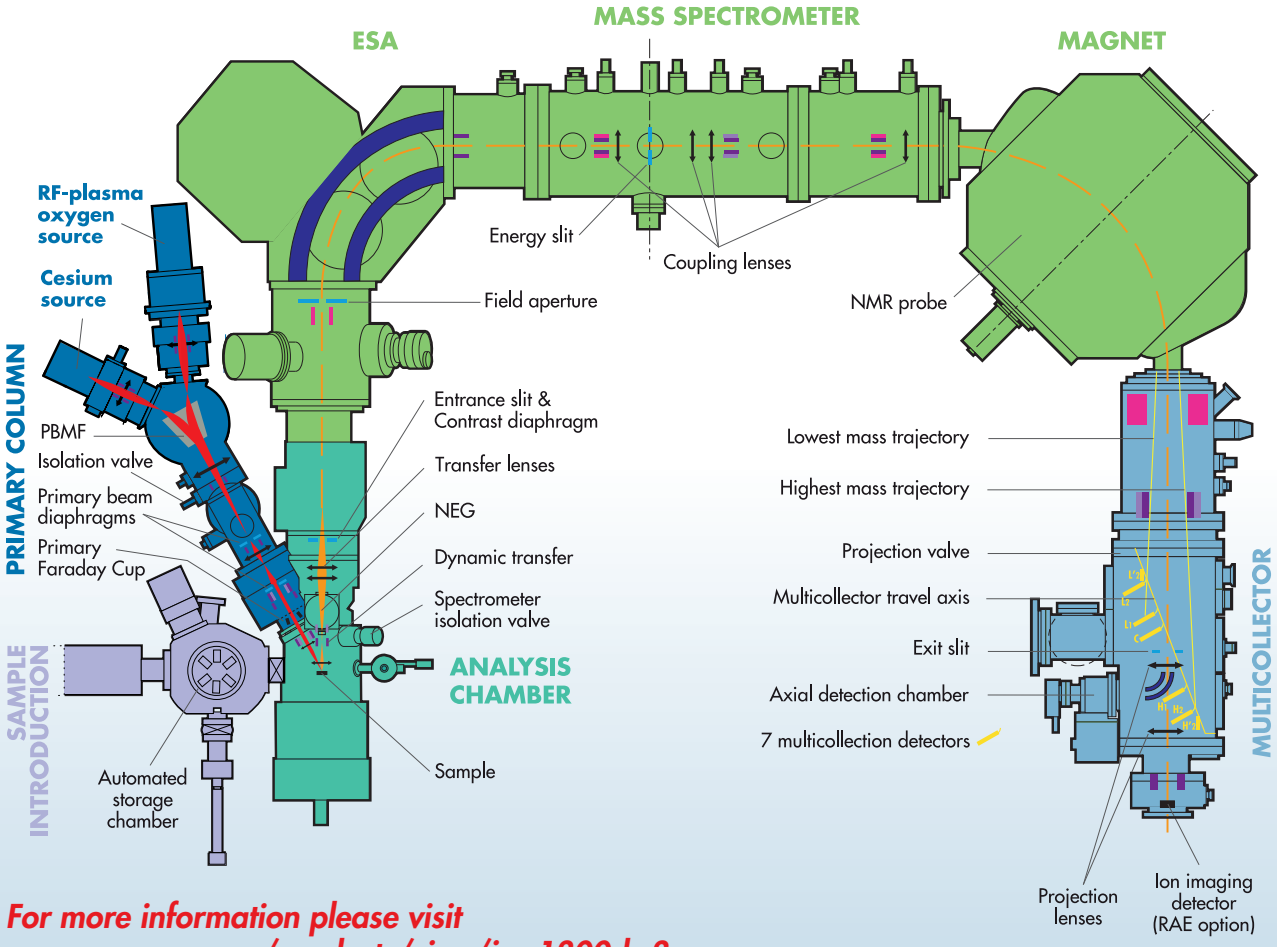
When a solid sample is sputtered by primary ions of few keV energy, a fraction of the particles emitted from the target is ionized. Secondary Ion Mass Spectrometry consists of analyzing these secondary ions with a mass spectrometer.

The SIMS technique is "destructive" by its nature (sputtering of material). It can be applied to any type of flat, solid material that can be kept under vacuum.

In dynamic SIMS, bulk composition and in-depth distribution of trace elements are investigated with a depth resolution ranging from sub-nm to tens of nm. SIMS is recognized as the most sensitive elemental and isotopic surface analysis technique.

CAMECA IMS 1300-HR³

The latest generation multicollection Large-Geometry SIMS is equipped with a high-brightness RF-plasma oxygen ion source in addition to the cesium microbeam source. It also includes an automated storage chamber for analysis of multiple mounts in chained mode.



For more information please visit
www.cameca.com/products/sims/ims1300-hr3

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