Contribution ID: **134** Type: **Oral**

High Precision Isotopic Analysis of Actinide Bearing Materials: Performance of a New Generation of Purpose Built Actinide Multi-Collector ICPMS Instruments

Wednesday, 9 July 2014 14:00 (20 minutes)

Forensic trace analysis is advanced through the sensitive and precise measurement of isotope ratios. Such information has always been a critical component in nuclear forensic analysis, and mass spectrometry - traditionally TIMS - is the instrument of choice for low-level and precise isotope abundance measurements. A new class of inductively coupled plasma mass spectrometers (ICPMS) has recently become commercially available that incorporate new features to enhance the analysis of actinides, most notably, a detector array specifically designed for the isotopic abundances encountered in uranium analyses. The array/instrument can also be usefully employed for the analysis of other actinides and most of the periodic table.

The authors contributed to the realization of one of these purpose-built actinide MS instruments, the Neptune-PlusTM (ThermoFisher, Bremen, Germany). This instrument also included a new, all dry pumped vacuum system, most notably in the high speed pump used in the vacuum interface between the atmospheric pressure ICP and the mass analyzer. The authors are most familiar with instruments from ThermoFisher, however other such instruments exist, notably from Nu Instruments (Wrexham, England). Prior to the introduction of these instruments, nearly all MC-ICPMS instruments had a single ion pulse counting detection channel, surrounded by an array of Faraday Cup detectors. These prior instruments had been developed largely for the geology and geochemistry communities where very high precision measurements are the norm and samples sizes are adequately large to enable high precision measurement. The new instruments retain most of the Periodic Table coverage of the earlier generation instruments, but with the addition of these new detector arrays, precise measurement of very low abundance isotopes, e.g., 234U and 236U, becomes possible even where very little analyte is available.

The features of these instruments will be described, along with analytical performance figures of merit, selected applications and recent innovations. The latter will include the authors'research on femtosecond laser ablation (fs-LA) as a sample introduction method for MC-ICPMS as a new way to measure the isotopic composition of surfaces and particles with high spatial resolution. Comparison of the fs-LA method with more established methods such as SIMS will be provided. Conventional, solution introduction is also of interest in these instruments as are novel sample introduction methods. An example of the latter that will be described is the use of electrochemically modulated separations to effect chemical separation of interfering species like 238U1H+ from 239Pu+, 241Am+ from 241Pu+ , online, as part of the sample introduction to the ICP. Other performance figures of merit of interest include isotope ratio uncertainty as a function of analyte amount and isotopic composition, sample utilization efficiency (ion counts/atom in sample), and spectral interferences for the least abundant actinide isotopes of interest.

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Session Classification: Technical Session 3F