Method Optimization to Extract 1,000,000 Pu Atoms From Lunar Soil

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Resonance ionization mass spectrometry (RIMS) has the potential to select and count ~1100 ^{244}Pu atoms using the Laser Ionization of Neturals (LION) RIMS instrument at LLNL.[1] Successful analysis of such ultra-trace samples requires both high sample purity and recovery through chemistry that are typically not achieved nor quanitified in conventional environmental-level Pu extractions. Furthermore, sample mounting for RIMS requires the full Pu aliquot in a ~250 micron spot, which demands an exceptionally clean sample in a very small final volume (<1 μl). Here, optimizaton of ultra-trace (<0.6 fg) Pu extraction from 10 grams of lunar soil simulant JSC-1A are achieved using a simple chemistry that minimizes reagent volume while maintaining reproducibile recoveries of >70%.

Tests were performed on fully digested aliquots of JSC-1A spiked with pg to fg quantities of the Pu standard CRM-130. Extraction chromotography using Eichrom TEVA resin was optimized by varying matrix ion concentrations, redox reagents ($NaNO_2$, $NH_2OH{\cdot}HCl$, H_2O_2), eluent reagents (HCl:HF, HCl:HI), and preconcentration steps (LaF, CaF, metal carriers). Recovery of Pu was determined via isotope dilution mass spectometry on a Nu III MC-ICP-MS at LLNL with a detection limit of ~0.5 fg Pu/mL solution. Preliminary results show that the Pu oxidation state is vitally important for co-precipitation reactions and Pu resin adsorption ($k'_{Pu[III]} \approx 0$, $k'_{Pu[IV]} > 10{,}000$)[2], and was significantly impacted by high cation concentrations in the digested sample matrix. The best results are achieved with the use of $NaNO_2$ and repetetive TEVA separations.

The optimized method will be used to quantatitively extract 150,000-1,500,000 atoms of ^{244}Pu from 10 g of lunar soil to constrain the contemporary interstellar flux of the r-process nuclide ^{244}Pu ($t_{1/2} \approx 80Ma$).[1] This work will constrain the r-process nucleosynethesis source by selecting between two possible r-process production scenarios: 1) neutron stars (i.e., a rare ^{244}Pu source) or 2) supernovae events (i.e., a frequent, steady state ^{244}Pu source). This method also has potential nuclear forensic and paleoclimate applications.

[1]Trappitsch et al. (2019) *LPSC Abstract* 2132.
[2]Horwitz et al. (1995) *Analytica Chimica Acta*, **310**, 63-78.