

Revisiting Classical Methods: Adapting Environmental Purification of Trace Plutonium and Neptunium to a Uranium Foil Matrix with Precipitations

Kimberly Hinrichs, [khirichs@lanl.gov](mailto:khinrichs@lanl.gov); Daniel Gurganus, Andrew Nunn
Nuclear and Radiochemistry Group, Chemistry Division
Los Alamos National Laboratory
Los Alamos, NM 87545, USA

Environmental collections containing trace levels of plutonium and neptunium are routinely analyzed by total evaporation-multiple ion counting-thermal ionization mass spectrometry (TE-MC-TIMS) in the clean labs at Los Alamos National Laboratory. Recently, adaptations to the chemistry were necessary to enable the measurement of ultra-trace plutonium and neptunium from a set of irradiated uranium foils. Due to logistical constraints preventing the introduction of the radioactive foils into the clean lab areas, an initial separation was required to remove as much uranium as possible prior to the routine chemistry. After a review of the available historical methods, a quick and easy co-precipitation step was selected.

Routine environmental chemistry proceeds from sample ashing (to 550°C), through nitric/hydrofluoric/perchloric dissolution, to a four-step purification process. The purification begins with actinide pre-concentration and separation from matrix elements (e.g. alkali metals, sulfate, and phosphate ions) using a neodymium oxalate co-precipitation followed by an iron hydroxide co-precipitation. The resulting precipitate is further purified by two anion exchange columns, and the plutonium-neptunium fraction is bound onto a single anion resin bead for loading onto a canoe-shaped Re TIMS filament. While the initial precipitations are not designed specifically to separate uranium from plutonium and neptunium, there is some separation afforded there, which is completed with the anion exchange columns.

To adapt this chemistry to an irradiated uranium foil matrix, a neodymium fluoride co-precipitation was added as a first step in the purification scheme, and separation factors between uranium and plutonium-neptunium were determined for each of the three precipitations. Results show that the neodymium fluoride co-precipitation removed approximately 90% of the initial uranium, even without a hold-back oxidant for the uranium, and the oxalate precipitation removed a further 90-95% of the remaining uranium with chemical recoveries of both plutonium and neptunium essentially quantitative. The iron hydroxide precipitation affords no separation of uranium from the other actinides.

Modern radiochemistry has a heavy dependence on ion exchange and extraction resin chromatography, and for good reason. However, co-precipitations, as well as liquid-liquid extraction and other classical methods remain valuable tools for the benchtop radiochemist that can be relied on for quick and effective separations.