**Separating super-grade plutonium for forensic analysis**

K.J. Glennon,[1,2,3] E.M. Bond,[3] T.A. Bredeweg,[3] S.S. Chirayath,[4,5] and C.M. Folden III[1,2]

[1]*Department of Chemistry, Texas A&M University, College Station, Texas 77843*

[2]*Cyclotron Institute, Texas A&M University, College Station, Texas 77843*

[3]*Nuclear and Radiochemistry, Los Alamos National Laboratory, Los Alamos, New Mexico 87545*

[4]*Center for Nuclear and Security Science and Policy Initiatives, Texas A&M University, College Station, Texas 77843*

[5]*Department of Nuclear Engineering, Texas A&M University, College Station, Texas 77843*

Discriminating the origin of special nuclear material (SNM) such as Pu and $^{233,235}$U is of key importance in the field of pre-detonation nuclear forensics. Discriminating the irradiation conditions under which proliferated Pu material was produced can help lead intelligence officials to the source of such proliferated material. One Pu discrimination technique previously investigated by our group is a maximum likelihood methodology which compares a set of measured fission product (FP) isotope ratios in unidentified Pu material to a set of simulated FP isotope ratios produced under a variety of reactor conditions [1,2]. The use of FP isotope ratios in addition to Pu isotope ratios allows this discrimination approach to distinguish between many reactor types over a large range of fuel burnups without considering assumptions about the material's origin. Our previous study suggested that at least 5 g of PUREX separated Pu would be necessary to contain a sufficient mass of the FPs for analysis [3].

Recently, four samples of super-grade Pu between 4 – 60 mg have been used to develop a chemical methodology to isolate and concentrate the FP elements of interest for maximum likelihood reactor-type discrimination. The four samples of Pu were dissolved in 8 M $HNO_3$ and loaded onto an anion exchange column to retain $Pu^{IV}$ while U, Cs, Ba, and the rare-earth elements (REEs) eluted. The U was isolated into its own fraction at this step; the Cs, Ba, and REEs were further separated in concentrated $HNO_3$ using a cation exchange column to isolate the REEs. Cs and Ba were then isolated into their own fractions using a Sr specific extraction chromatography resin (SR Resin by Eichrom) in 1 and 0.1 M $HNO_3$. Fig. 1 describes the isolation of these elements into their own fractions with their reported yields. In general, the FP elements were recovered with excellent yields greater than 80%. This procedure was shown to recover quantities of FP elements as low as $10^{-10}$ g from the 60 mg sample of Pu. A previous characterization of irradiated $UO_2$ material [4] suggests that 5 g of PUREX separated weapons-grade Pu discharged at a burnup of 1 GWd/MTU would contain at least $10^{-10}$ g of most FPs of interest, indicating that this chemical methodology may be sufficient to isolate and measure FP isotope ratios in large samples of PUREX separated Pu.

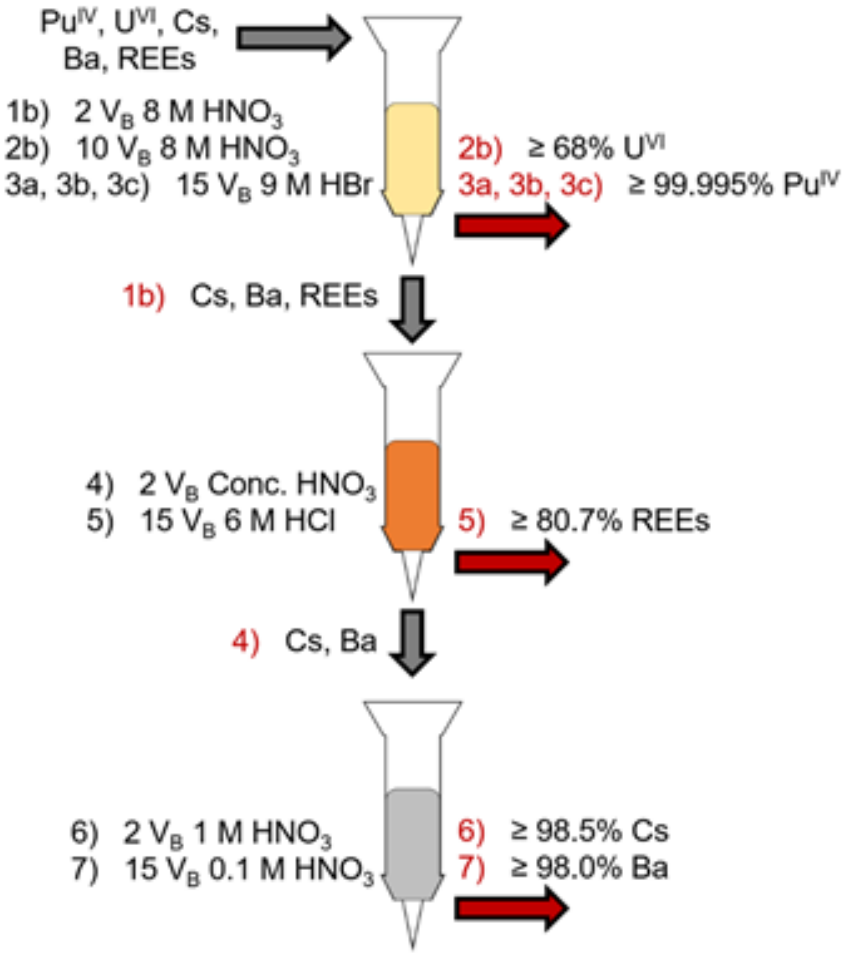


**Fig. 1**. Chromatography separations of the four Pu samples to isolate select FP elements. The first column used anion exchange resin (yellow), the second cation exchange resin (orange), and the third SR resin by Eichrom (gray).


[1] J.M. Osborn, K.J. Glennon, E.D. Kitcher, J.D. Burns, C.M. Folden III, and S.S. Chirayath, Nucl. Eng. Technol. **51**, 384 (2019). doi:10.1016/j.net.2018.11.003

[2] J.M. Osborn, E.D. Kitcher, J.D. Burns, C.M. Folden III, and S.S. Chirayath, Nucl. Technol. **201**, 1 (2018), doi:10.1080/00295450.2017.1401442

[3] K.J. Glennon, J.M. Osborn, J.D. Burns, E.D. Kitcher, S.S. Chirayath, and C.M. Folden III, J. Radioanal. Nucl. Chem. **320**, 405 (2019). doi:10.1007/s10967-019-06486-w

[4] J.M. Osborn, K.J. Glennon, E.D. Kitcher, J.D. Burns, C.M. Folden III, and S.S. Chirayath, Nucl. Eng. Technol. **50**, 820 (2019). doi:10.1016/j.net.2018.04.017