**Ph.D. Dissertation Proposal: Paul Mendoza**

**Dissertation Title:** Experimental Characterization of Pu Separation by PUREX Process on a Low-Burnup, Pseudo-Fast-Neutron Irradiated DUO2 for Product Decontamination Factors and Nuclear Forensics

1. **Introduction**

In the wake of the Iran deal, North Korea’s pursuit of nuclear weapons, and the onset of the Islamic State of Iraq and Syria, nuclear proliferation concerns remain relevant. A nonproliferation focal point is the control and accounting of weapons usable material. These safeguards have been the responsibility of the International Atomic Energy Agency with the goal of promoting the safe and peaceful use of nuclear technologies1. Although these efforts, backed by an “atoms for peace” mentality and the treaty on the non-proliferation of nuclear weapons, have hindered the development of weapons in some states2, efforts beyond safeguards prevention are necessary due to “the awful arithmetic of the atomic bomb”3. An effort, some steps after, and potentially outside of, safeguards, but before detonation, is interdicted material origin attribution.

Material origin attribution, in this context, applies to deducing history of nuclear material, specifically weapons usable material. The importance and time sensitivity of which, varies widely in different circumstances, but is fundamentally an inverse problem, which are more difficult to solve.

For example, if a purified sample of plutonium were interdicted at a border between states, information from a wide variety of sources will be used to determine a likely hypothesis for where the sample came from. This hypothesis, is an informed, studied, and tested conclusion, but is only one possible explanation for the history of the sample.

The point is not to say that this exercise is hevel, but difficult and multi-layered. Multi-layered in the sense that there are many different sources of information and an infinite realm of possibility for hypothesis. This study proposes to provide another source of information for this complex problem coupled with an analysis of the range of applicability for this source of information.

Previously, computational studies have indicated that analysis of contaminants in Plutonium Uranium Recovery by Extraction (PUREX) processed plutonium could give indications of material origins.4 Unlike enrichment of U235, Pu239 “enrichment” involves neutron irradiation, which introduces radioactive fission products and hence, purification by some means. Purification by PUREX is most common for low enriched uranium fuel5 - important because U238 converts to Pu239 upon neutron capture. Material attribution is complicated by purification due to varying and unknown elemental decontamination factors (DFs, a measure of purification) for different PUREX procedures.

The problem of attribution for unpurified material has been previously studied4, 6, 7, ideally, if elemental DFs for interdicted plutonium were determined, then these previous methodologies could be used to narrow material origins by applying these correction factors, assuming DFs for different isotopes of the same element are equal.

1. **Objectives**

The objective of this research is to determine DFs and distribution coefficients (DCs, the ratio of concentrations between organic and aqueous phases) for a developed benchtop scale PUREX process for important attribution elements noted in previous research (Cs, Eu, Rb, Sr, Nd, Pm, Sm, U, and Pu).4 Both DCs and DFs will be determined for this process so that the two parameters will be experimentally connected to the specific PUREX process utilized. This establishment is important because mathematically, DC values, coupled with process information, can be used to calculate a reasonable estimate of a DF8-11, but experimentally, this connection has never been publicly discussed for the PUREX process.

This is mostly due to complications in bridging the gap between controlled experiment and practice. Most reported numbers from experiments are for steady state, equal contact volumes systems. Some practical reprocessing plants designs (example: counter current) have constantly changing concentrations, which makes mathematically determining DFs from DCs for systems difficult. Usually the DF measured from initial and final solutions and reported.

This above goal will be met by analyzing mass, gamma, and alpha spectroscopic results from the process steps of a four extraction three back-extraction PUREX process run in triplicate. Each step of the process will be analyzed for DC values, and the overall process will be analyzed so that the DF values will be determined through initial and final solutions and calculated through individual DCs.

Further, in order to determine the applicability of utilizing this information for forensic purposes the uncertainty on two important calculations will be explored. First, fuel composition calculations, have a myriad of applications, but on the research purposes to determine the uncertainty in the production of these elements in a thermal system.

1. **Present Status of the Question**

Summarize your literature review. What is the pertinent previous research in this field? Include citations.

While DCs for the various process steps of PUREX have been previously reported, details about elemental DFs for PUREX cycles have been largely limited to the major activity contributors, such as 106Ru and 95Zr12, or the heavy metal elements (Th, U, Pu)13.

1. **Procedure**

Describe your general solution method. If your work is experimental, give the nature of the data you will collect and how you will analyze it. If your work is computational, include a description of how you will verify your methodology (test cases, simulations, sensitivity analysis, etc.).

1. **Expected Results**

Include work already done and what you expect to see in the remaining work. Also talk about risk management – if something doesn’t work as expected, are there alternative soluitons?

1. Fischer D. History of the International Atomic Energy Agency. The First Forty Years. 1997.

2. Mozley R. The Politics and Technology of Nuclear Proliferation: University of Washington Press; 1998.187-188

3. Eisenhower DD. Atoms for Peace speech. Voices of Democracy. 1953.

4. Chirayath SS, Osborn JM, Coles TM. Trace Fission Product Ratios for Nuclear Forensics Attribution of Weapons-Grade Plutonium from Fast and Thermal Reactors. Science & Global Security. 2015;23(1):48-67.

5. Benedict M, Levi H, Pigford T. Nuclear chemical engineering. 2nd ed: McGraw-Hill Pulishing; 1982.477

6. Scott MR. Nuclear forensics: attributing the source of spent fuel used in an RDD event: Texas A&M University; 2005.

7. Glaser A. Isotopic Signatures of Weapon-Grade Plutonium from Dedicated Natural Uranium-Fueled Production Reactors and Their Relevance for Nuclear Forensic Analysis. Nuclear Science and Engineering. 2009;163(1):26-33.

8. Colburn AP. Simplified calculation of diffusional processes. general consideration of two-film resistances. 1939;35:211-36.

9. Sherwood TK, Pigford RL. Absorption and extraction. 2d ed: New York : McGraw-Hill; 1952.

10. Long JT. Engineering for nuclear fuel reprocessing: New York : Gordon and Breach Science Publishers; 1967.162-272

11. Perry RH, Green DW. Perry's chemical engineers' handbook. 8th ed. New York: McGraw-Hill; 2008.

12. Stoller S, Richards R. Reactor handbook, volume II, fuel reprocessing. Inter science Publishers, Inc, New York. 1961:107-234.

13. Siddall TH, Prout WE, Parker SG, Commission USAE, Savannah River L, Nemours EIdPd, et al. Equilibrium distribution data for purex and similar extraction processes: E.I. du Pont de Nemours & Co., Explosives Dept., Atomic Energy Division, Technical Division, Savannah River Laboratory; 1957. 27 p. p