**Determination of Decontamination Factors for Various Radioisotopes during the PUREX Process of Irradiated DUO2**

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**Abstract –** Conclusions. Describe briefly the chemistry behind process. PUREX process actually simulated in experiment. Proven by XX % separation from Pu. Isotopic compositions within irradiated sample agreed with simulation results to within XX %. Decontamination factors ranged from XX % to YY % per PUREX cycle. Briefly discuss chemical reasons for particular isotopic contaminates.

I. BACKGROUND

*I.A Objective*

This project proposes to study how indicator fission products (FP) and actinides propagate through the industrial Plutonium Uranium Redox Extraction (PUREX) process. The specific application will utilize low burnup (< 4000 MWd/MTU) depleted uranium irradiated in the fast neutron spectrum of Oakridge National Laboratories’ (ORNL) High Flux Isotope Reactor (HFIR). The fuel, along with FP and minute amounts of weapons grade plutonium, will be dissolved in 4 M nitric acid and put through three PUREX process cycles and analyzed at every step with high resolution gamma spectroscopy, and ICP-MS analysis.[[1](#_ENREF_1), [2](#_ENREF_2)] This information will be useful in trace element analysis of interdicted weapons grade plutonium.[[3](#_ENREF_3)] Analysis will determine appropriate decontamination factors for various elements at each step in the process as well as important endogenous information about the power history for the depleted uranium.[[4](#_ENREF_4)]

*I.B Significance*

The overarching push behind this project stems from the need to increase our nuclear forensics attribution capabilities in the United States.[[17](#_ENREF_17), [18](#_ENREF_18)] Nuclear Forensics is the investigative activity that surrounds the search for attributes of undetermined radioactive specimens. Its primary goal is to answer key questions surrounding attribution and future expectations and consequences so that decision makers are adequately equipped to respond to either an intercepted material or a nuclear explosion. The overarching question behind this project is: can basic spectroscopy analysis of PUREX processed interdicted weapons grade plutonium give indication to what type of reactor the material was produced in, the cycle history and burnup of the fuel, or the last processing date? This information would be vitally useful in light of our continued push to increase our nuclear forensics analysis base.

*I.B Introduction*

Little Boy’s decent into Hiroshima introduced the world to a new level of destruction that outclassed all previous mediums for taking life. Projectiles and chemical explosives paled in comparison to the direct conversion of mass into energy that nuclear weapons entail. The arms race of the ensuing Cold War was an indicator showing that the new tier of nuclear weapons will be an aspiration and desire for many people groups, whether good or bad. Therefore, it is the duty of mankind to limit accessibility to these weapons.

Accessibility can be limited through a number of ways, including but not limited to the control of technology, information or material. In light of our postindustrial and academically proliferate societies limiting access to technology and information is not always ensured and therefore all forms of arms control should be seriously considered. The most important being limiting access to material that could be used to make a nuclear weapon, namely, 235U and 239Pu. This project focuses on the more deadly of the two, which is most efficiently and almost uniquely produced in nuclear reactors through the neutron capture of 238U.[[4-6](#_ENREF_4)]

[[7](#_ENREF_7)]

It might seem counterintuitive, but a method to help limit the production of this dangerous isotope is to learn how its most optimally produced.

Other isotopes of plutonium, which are mostly weapon deterring, are produced through subsequent neutron absorptions of the 239Pu. Thus, the resulting plutonium isotopic composition is strongly dependent on how long the fuel remains in the reactor. This composition is also dependent on relative reaction cross sections and half-lives. The irradiated fuel will contain plutonium isotopes ranging from 238 up to 242. It should be noted that as a result of this, the resulting composition of the plutonium can provide important information about the reactor and generally, lower neutron irradiation levels (< 4000 MWd/MTU) lead to higher isotopic concentrations of 239Pu.

The reaction shown above occurs more optimally in a fast neutron spectrum as opposed to a thermal neutron spectrum and also subsequent neutron absorptions for 239Pu happen less optimally in a fast neutron spectrum.[[7](#_ENREF_7)] These two facts means that plutonium with high concentrations ( > 93%) of 239Pu (weapons grade plutonium) is most optimally produced in a fast neutron spectrum.[[6](#_ENREF_6)]

The two conditions just now described, which optimally produce 239Pu, are met in the outer blanket of a Fast Breeder Reactors (FBR).[[8-10](#_ENREF_8)] This project will irradiate depleted uranium to a low burnup in a fast spectrum comparable to the new fast reactors being built in India.[[8](#_ENREF_8)] Depleted uranium was used so that the concentration of 238U will be as high as easily achievable.

It should be noted that thermal reactors with various different fuel types will produce different concentrations of FP. An important goal of this project is to determine how much residue signature is passed to the end plutonium product for a fast reactor depleted fuel type, and determine if that residue is sufficient so that distinction between fast and thermal reactors can be made.

The next step in producing isolated 239Pu would be to chemically isolate plutonium. The most common method for this is the PUREX process. Which utilizes the ionization states of plutonium and in nitric acid and tributyl phosphate. The PUREX process was developed as part of the Manhattan Project in 1947 and focuses on isolating plutonium from fission products and uranium.[[1](#_ENREF_1), [3](#_ENREF_3), [11](#_ENREF_11)]

After chemical isolation, the weapons grade plutonium is then converted to a form that is usable for a weapon. Given a significant quantity (SQ) of this material, this is the last stop before a very real possibility of a post detonation scenario and is the hypothetical interception point for this project. The importance of being able to identify the origins of this material is evident and even if less than a SQ were intercepted from an unauthorized person or country, prompt termination of all activates that led to the materials procurement will be necessary.

Analysis of the intercepted material will have trace isotopes present. These isotopes will come from the process the plutonium underwent. It is the hope of this project to mimic and study these processes so that ties can be drawn back to the originating reactor and provide enough information so that these activates will be stopped.

*I.C Previous Nuclear Forensics Studies*

There have been lots of previous work in the area of using isotopic compositions in order to determine or verify important origin and process information of reactor fuel.[[1](#_ENREF_1), [3](#_ENREF_3), [4](#_ENREF_4), [12-15](#_ENREF_12)] These works are varied in implementation and scope.

An application for using isotopic composition and ratios in spent reactor fuel is in the area burnup verification. One such study utilized measurement of isotopic ratios of stable noble fission gases during reprocessing in order to verify burnup.[[12](#_ENREF_12)] This study utilized data analysis methods to determine specific fuel parameters and then compared with measured values of stable noble gases. It is important to verify fuel burnup for reasons described above. If a fuel assembly were burned to a lower level than reported, then the isotopic concentrations of plutonium could be higher in 239Pu.

Others in the field of nuclear forensics have looked a little further ahead in the processing of used nuclear fuel. Given spent fuel from a reactor that is used for a radiological dispersal device (RDD), one looked to determine the burnup, enrichment, and age for the fuel.[[13](#_ENREF_13)] These combined data would then be subsequently used for reactor attribution purposes. This situation is similar to our scenario in that it looks at FP in the matrix of the fuel itself and deduces reactor type and information about the history of the fuel, but this method does not consider any chemical processing.

Analysis of plutonium isotopics alone have been suggested to help identify reactor type at the same processing step as our proposed project.[[16](#_ENREF_16)] The reactors considered were of the fast and thermal type. This study concluded that while it is possible to identify with a high level of confidence differences in plutonium composition between fast and thermal reactors, it is difficult to make this same distinction in “dedicated production reactors fueled with natural uranium.” This should be contrasted with our study with depleted uranium in a fast neutron spectrum.

A specific paper looked at PUREX co-processing of spent LWR fuels.[[15](#_ENREF_15)] A couple of decontamination factors and production yields for uranium and plutonium were determined. The specific goal of this paper was to demonstrate that uranium and plutonium could be co-extracted for reprocessing. This study was also limited in determining decontamination factors for a wide array of isotopes. Which is not important in terms of reprocessing, because the standard seeks to minimize activity contamination in the sample, but is important in forensic analysis, because trace isotopes, whether radioactive or not, could give indication of origin.

There also have been various studies about overall decontamination factors that result from the overall PUREX process, there are however, no studies that study the intermediate steps of the PUREX process. This is important because there are slightly different methods towards implementing the PUREX process and understanding the intermediate steps are helpful because they produce different decontamination factors.

Nuclear proliferation is a growing concern worldwide due to the increased availability of nuclear materials, knowledge of sensitive technologies, and the possibility of diverting nuclear materials such as uranium and plutonium away from peaceful uses. Due to this increasing risk of nuclear threats, nuclear forensics capabilities are being developed at Texas A&M University with sponsorship from the Department of Homeland Security. This nuclear forensics capability development will aid in improving deterrence capabilities and in educating the next generation of scientists and engineers. One of the nuclear forensics activities that we are currently pursuing is the use of computational and experimental methods to determine the isotopic characteristics of weapons-grade plutonium separated from low-burnup uranium that could be produced in different neutron spectral environments available in foreign nuclear reactors.

Combating Nuclear Terrorism (lecture 21 NUEN 610)

Improve interpretation capabilities. “nuclear forensic science relies to a large extent on the expertise and experience of the investigating scientists. Knowledge in areas such as radiochemistry, nuclear physics, reactor physics, material science and in the nuclear fuel cycle are required” (Ref1)

Our objective was to determine the differences in fission products and actinides characteristics for uranium samples irradiated in different type of nuclear reactors (thermal and fast reactors).

The final goal of these experiments was to analyze trace elements and isotopes present in the residual matrix produced by each and every step of industrial PUREX chemical reprocess by alpha and gamma spectroscopy and other analytical tools. The weapon grade plutonium separated from other actinides and fission products such as Cs, Ce and Sm was determined by ICP-MS with great precision to match the computational results.

*I.B Methodology*

This study will be accomplished by performing the following steps:

1. Perform a literature review to ascertain specific details behind the industrial PUREX process.

2. Develop laboratory procedures to mimic the industrial PUREX process with nanogram quantities of plutonium.

3. Check laboratory conditions and acquire and set up if necessary all laboratory materials needed including: a glove box, a fume hood, appropriate chemicals, spectroscopy systems, appropriate shielding, and other basic laboratory equipment.

4. Machining and irradiating depleted uranium at HFIR to an appropriate level as determined by a colleagues MCNP calculations.

5. Dissolve unirradiated depleted uranium (UDU) in 4 M nitric.

6. Dissolve irradiated depleted uranium (IDU) in 4 M nitric acid

7. Perform PUREX experiment with UDU and verify uranium separation with ICP-MS

8. Perform PUREX experiment with IDU, verify Pu is being separated with co-precipitation by alpha spectroscopy.

9. Perform three cycles with two separate aliquots and collect appropriate spectroscopy and ICP-MS data after each step.

10. Analyze data to determine decontamination factors for at least 5 isotopes, burn up for the fuel and power history for the reactor.

11. Determine variance for results.

Every fission product cannot be tracked through this analysis and thus the scope will be limited to detection capabilities of the gamma spectroscopy system, and the indicator isotopes in the ICP-MS

Irradiation of fuel leads to certain byproducts. (fission fragments and higher actinides). Complex and governed by X Y Z processes.

Two of the most important variables that determine the resulting composition of fuel leaving a flux field are initial composition and the characteristics of the flux field itself. Talk about these importances and how differences will change the outcome. (this introduces variables that we can study, different fuel types and different types of reactors).

Discuss several uses for irradiating fissile isotopes and some consequences for irradiating such fuel. Producing electricity, radiopharmaceutical (a radioactive compound used for diagnostic or therapeutic purposes), detector systems, and many other applications in industry (ref2)

One particular use is to produce material for nuclear weapons production. Most isotopes of concern are Pu, U233, and U235 (Lecture 12 NUEN 610, IAEA safeguards). Talk about methods for producing these isotopes. Mention that a hydrogen bomb typically needs a type of fission explosion to start.

Narrow down on Pu, and specifically enriched Pu. Explain why it is good for an explosive weapon. Mention ways for producing Pu in question. Talk about our particular method (irradiation and chemical separation).

Discuss various methods for chemical separation. How it can change with fuel form.

Discuss most commonly used form of chemical separation, PUREX. Discuss how the chemistry works. Discuss the differences for different isotopes. Discuss different stages and scrubbing steps.

Talk about how in nuclear forensics trace isotopes can be used to determine which methods of enrichment were used and what type of reactor produced the material (NUEN 610 Lecture, forensics book).

Talk about Gamma spectroscopy and how results were determined

Talk about ICP-MS and how results were determined

II. EXPERIMENTAL

*II.A. Instrumentation*

Fume hood, glovebox, centrifuge. Production quality and where they were purchased from. Talk about gamma detector and GENIE software. These operations were performed inside a heavily shielded glove box.

*II.B. Reagents and Materials*

List of chemicals and where they were bought, stock solutions impurities. , pipettes, vials.

The uranium samples for this project were irradiated at the High Flux Isotope Reactor (HFIR) of Oak Ridge National Laboratory. After chemical processing of the irradiated uranium samples, the isotopic concentrations (and ratios) of selected fission products and actinides were measured and the burnup of nuclear material was determined

*II.C. Procedure*

Pu/U separation experiment (talk about how we wanted to make sure Uranium and Pu were separated)

PUREX process experiment. Talk about all the great details behind pipetting and transferring, counting and diluting.

III. RESULTS AND DISCUSSION

Talk about the Uranium experiment. The results, talk about what you think about the results. How the results helped us move forward with confidence.

Present the isotopic results. Gamma spectrums, whatever we got. Let them have all the gory details.

IV. SUMMARY AND CONCLUSIONS

In the future…I would like to be from the future.

FUNDING

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**Results and Discussion:**

U/Pu separation Experiment (results and discussion)

Chemistry? States in solution? Function of concentration?

The results for the uranium/plutonium separation are shown in Fig. XX. Separation of plutonium from uranium is a maximum around XX M HNO3. Uranium tends towards one phase or another based on (chemistry lingo). This is supported by (reference).

The concentration of HNO3 is extremely important because as a variable, it can vary widely (put in theory? Get rid of?)

Nitric Acid separation experiment (results discussion)

Chemistry? After this partition was confirmed the percentage of Pu (III) present in solution was determined through two experiments. Plutonium exists in HNO3 solution in both (III) and (IV) oxidation states in these forms. (chemistry). Sodium nitrite converts Pu (IV) to Pu (III) by the following means. Two different experiments were conducted with and without sodium nitrite added.

Pu and U results

As a function of contact volume and time. And number of contacts.

Describe as a function. Relate to literature.

Describe overall results (at the end of the day how much Pu separated out)

Decontamination factors at different nitric acid concentrations.

These are the trends noticed

Chemistry?

These will be useful for nonproliferation because…

Should we make comparisons to industrial column separation? (Compare volumes and contact time?) Isotope separation as a function of contact?