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Experimental Characterization of Pu Separation by PUREX Process on a Low-Burnup, Fast-Neutron Irradiated DUO₂ Surrogate for Nuclear Forensics

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Abstract

This project investigated how fission products separate through the Plutonium Uranium Redox Extraction (PUREX) process. A depleted uranium surrogate pellet with 0.25% enrichment, weighing 12.9 mg was irradiated in a fast flux spectrum to 3000 MWd/t over the course of three months at ORNL's HFIR reactor. The irradiated pellet was dissolved in 8 M HNO₃ and carried through a laboratory scale PUREX process. A 30 vol% TBP/kerosene mixture was used for the organic phase. **XX** fission isotopes were analyzed with gamma spectroscopy along with inductively coupled plasma mass spectrometry (ICP-MS) at every stage of the plutonium extraction. Isotopes **X X X** which are useful for nonproliferation were determined to have an overall decontamination factors of **Y Y Y**. Distribution coefficients and ratios of separation for isotopes were also determined.

Keywords

Nonproliferation, Irradiated Uranium, Decontamination Factors, Plutonium Extraction

Introduction

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. This project focuses specifically on plutonium extraction by processing neutron irradiated depleted uranium dioxide (DUO_2) fuel, which contains a spectrum of fission products that are systematically separated from the plutonium by PUREX processing. The question of whether trace-level isotopes or isotope ratio signatures exist as a result of naturally occurring contamination or trace residues associated with the chemical separation process was examined. These signatures, when coupled with reactor burnup data, could potentially be used to determine plutonium origin.

Previous work in the area of attribution has been carried out with varied implementation and scope.[1-7] Related work in the area of decontamination of plutonium has also been studied to some extent.[2, 7-9]

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.[10] This study utilized data analysis methods to determine specific fuel parameters that gave indication for burnup and then compared with measured values of stable noble gases. The importance of burnup verification is due to the fact that the isotopic concentrations and amount of Pu in spent fuel vary as a function of burnup.

Others, in the field of nuclear forensics, have studied the processing of used nuclear fuel. Given spent fuel from a reactor that is used for a radiological dispersal device (RDD), the burnup, enrichment, and age for the fuel was determined.[5] 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.[11] The reactors considered were of the fast and thermal type. This study concluded that

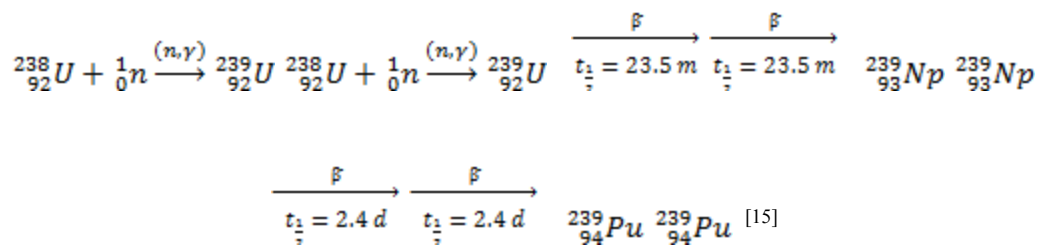
while it is possible to identify with a high level of confidence differences in plutonium composition between fast and thermal reactors, it is difficult to make this same distinction in “dedicated production reactors fueled with natural uranium.” This should be contrasted with our study with depleted uranium in a fast neutron spectrum.

A specific paper looked at PUREX co-processing of spent LWR fuels.^[7] 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.

There have been many books and papers that have studied the PUREX process. Variations in solvent concentrations and flow rates coupled with Uranium saturation have been studied extensively. The primary focus being optimizing Pu and U extraction. Distribution coefficients between organic and aqueous phases in 30 v/o TBP have been reported for several isotopes under different circumstances.[12] Our results agree with these findings and present a more comprehensive list distribution coefficients of not only TBP in contact with nitric acid, but in contact with a ferrous environment.

Theory

This project focuses on the production and characterization of plutonium obtained through neutron capture of ^{238}U , to be specific, depleted UO_2 irradiated in a fast neutron environment.^[3, 13, 14]



The irradiated UO_2 could also contain plutonium isotopes ranging from mass numbers 238 up to 242 which arise from subsequent neutron absorptions. Plutonium isotopic composition could also vary depending on the neutron spectrum (i.e. the type of reactor). Hence, an investigation of the composition of the plutonium can provide important information about the reactor type and burnup. Generally, lower burnup levels (<5000 MWd/ MTU) lead to higher isotopic concentrations of ^{239}Pu (90% or higher)

Theoretical basis: Cross sections for fission and absorption coupled with energy dependence (how they lead to different concentrations in the reprocessed product), chemistry behind the PUREX process (U and Pu, but also decontamination factors), coupling chemistry and concentrations to lead to information about material origin (cite handbooks, no common knowledge Eqs.), theory behind **gamma/alpha spectroscopy?** **PUREX chemistry** Theory behind ICP-MS? Theory behind MCNP?

Experimental

The later referenced initially dissolved solution and the stock solution were stored in 20 ml glass disposable scintillation vials with urea caps from Fisher Scientific. Each experiment used VWR ultra high performance centrifuge conical-bottom tubes made of ultra-clear polypropylene copolymer with caps made of high density polyethylene. 50 ml centrifuge tubes were used for **prepared reused** solutions and 15 ml centrifuge tubes were used to hold samples from the stock solution. These centrifuge tubes were parafilm wrapped while samples were not in use to reduce evaporation losses, marked with fine point sharpie permanent marker to distinguish vials. The Champion F-33D fixed angle digital variable speed centrifuge from Ample Scientific was used for centrifuging samples after mixing in Fisher Scientific's Digital Vortex Mixer.

Most operations were conducted in a mBraun's LABmaster Pro Glove Box Workstation with four glove stations and an approximate volume of 1.4 m^3 . The glove box was equipped with a mBraun HEPA filter, a 29 kg Edwards Pump model number RV12, 0.4

mm gloves, and two antechambers. Both the glove box gloves and the 15 ml centrifuge tubes were cleaned periodically with Radiawash Towelettes from BioDex Medical Systems Incorporation. This was to maintain a contamination free laboratory. Pure argon and a hydrogen/argon fill gases was acquired from Praxair. Argon was used for normal operation while the combination gas was used to regenerate the filter.

Shielding was provided in the glove box by a lead shielded workstation, lead bricks, and a lead pig for the glass scintillation vial containing the stock solution. Extra support was provided underneath the glove box so that the stainless steel base would not give under the notable weight of the lead.

Basic laboratory procedures were conducted with standard personal protective equipment, including stretch ease powder-free nitrile examination gloves from Denville Scientific Incorporation. Pipetting used an Eppendorf 1000 μ l adjustable pipette with dualfilter 1250 μ l, certified clean and sterile purity grade tips.

The DUO_2 used for this experiment was acquired from Dr. McDeavitt (a nice guy), had an enrichment of 0.28 wt%, and was surrogated into a pellet of density XX g cm^{-3} . The distilled water used for dilutions was acquired from a 03/08/2011 manufactured (filter hasn't changed...just saying) Purelab flex water purification system with a model number PF3XXXXM1 to purify College Station tap water. The kerosene (100%), sodium nitrite (100%), and hydrofluoric acid (48%) were acquired from sciencelab.com, 69% nitric acid was acquired from Mallinckrodt Chemicals, Tri-n-butyl phosphate (>99%) was acquired from Fisher Scientific, iron sulfamate (40.26%) was acquired from Strem Chemicals Incorporation, and neodymium fluoride (99.95%) was acquired from GuideChem.

The pellet both prior to dissolution and after, were counted on a standard Canberra electrode coaxial Ge detector model number CC4018 which was connected to a Canberra Lynx MCA. GENIE software was used to analyze spectrum with samples typically encased in a lead tomb. Inductively coupled plasma mass spectrometry (ICP-MS) was also conducted for some samples with use of the University of Missouri's PerkinElmer

NexION 300X quadrupole ICP-MS operated in standard mode by Dr. James McKamey. A Canberra Passivated Implanted Planar Silicon (PIPS) detector coupled with an alpha spectrometer, MCA and a vacuum pump were also used qualitatively to verify plutonium separation of co-precipitated plutonium. **Plutonium co-precipitation was accomplished with neodymium fluoride, 2.2 M HF, and 0.22 μ m millipore paper.**

Detailed three dimensional models of Oakridge National Laboratories' (ORNL) High Flux Isotope Reactor (HFIR) were developed using the Monte Carlo transport computer code, MCNPX, coupled with the burn-up/depletion computer code, CINDER90. These were used to model the irradiation of a DUO_2 surrogate pellet so that expected activities and concentrations could be determined. A measured 12.9 ± 0.05 mg of DUO_2 was irradiated over the course of three months with two shut down periods in the HFIR flux spectrum to a calculated 3000 MWd/t. This produced 0.194 ± 0.035 mg of Pu.

After the short lived radioisotopes had opportunity to decay, the irradiated DUO_2 pellet was weighed in a weighing boat on an electronic balance and transferred to a round bottom flask. 5 ml of 8 M HNO_3 was added to the round bottom flask and the flask was heated at 50°C with constant 100 rpm stirring for two hours. The dissolution apparatus is shown in Fig. 1.

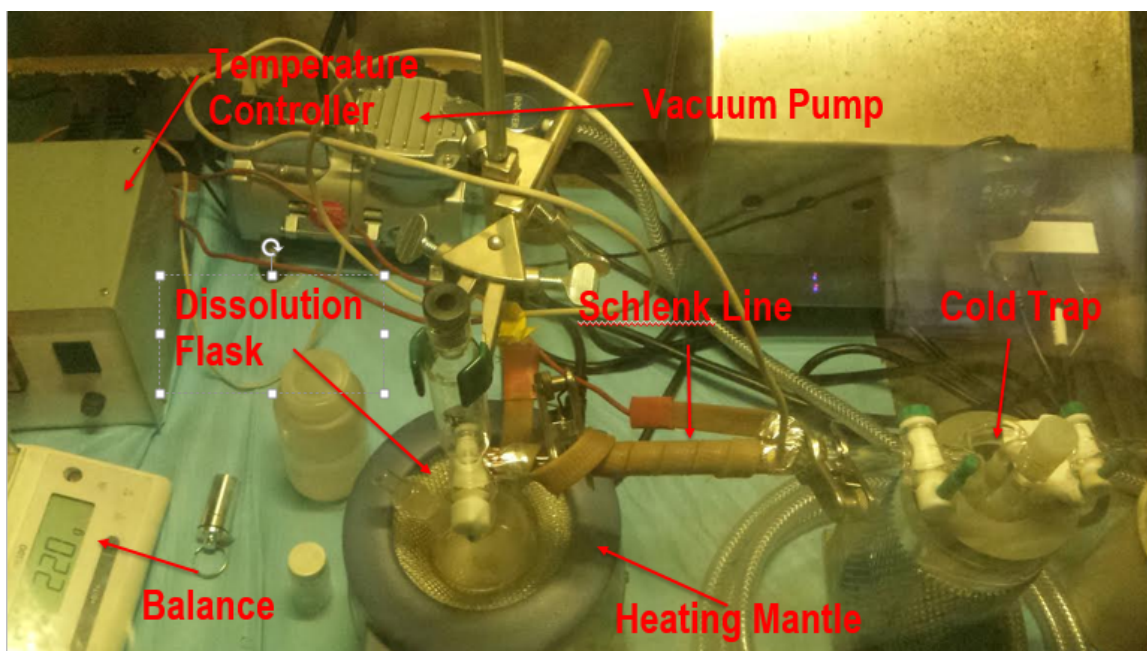


Figure 1. Dissolution of the Irradiated DUO₂ Pellet.

The round bottom flask was connected to a cold trap with the help of a schlenk line. The fission product gases such as H₂, CO₂, Kr, Br, I and N₂O were captured in the cold trap inside the molecular sieves which were surrounded by liquid nitrogen. The 5 ml concentrated 8 M HNO₃ solution was transferred from the round bottom flask to a 20 ml glass scintillation vial and kept heavily shielded. This solution is referred to as the “dissolution solution”.

| Gas | % (μg) | Freezing Point (K) |
|---------------------------------|-----------|--------------------|
| Tritium | 1 (0.001) | 14.01 |
| Carbon-14 (as CO ₂) | 1 (0.001) | 194.7 |

| | | |
|------------------|---------|--------|
| Krypton-85 | 15 (5) | 115.79 |
| Xenon-131 to 136 | | 161.4 |
| Iodine-129 | 1 (0.3) | 386.85 |
| Bromine-81 | | 265.8 |
| N_xO_v | | |
| N_2O | 25 CC | 182.35 |
| NO | | 110.15 |
| N_2O_3 | | 170.15 |
| N_2O_4 | | 262.15 |
| N_2O_5 | | 232.15 |

In order to reduce the amount of activity per sample, 500 μ l from the dissolution solution was diluted to 5 ml and the concentration was changed to 4 M HNO_3 . This was subsequently called the, “stock solution”. The stock solution was stored in its own 20 ml glass scintillation vial in a lead pig inside the glovebox. Each PUREX process used 500 μ l aliquots from this stock solution.

The flowchart for a PUREX cycle for one of three experiments is shown in Fig. 2 below. The other two experiments had variants from this base flow chart. The step titled, “Diluted to 4 ml with 4 M HNO_3 ” has been previously described. The first experiment proceeded with transferring a 500 μ l aliquot into a 15 ml centrifuge tube.

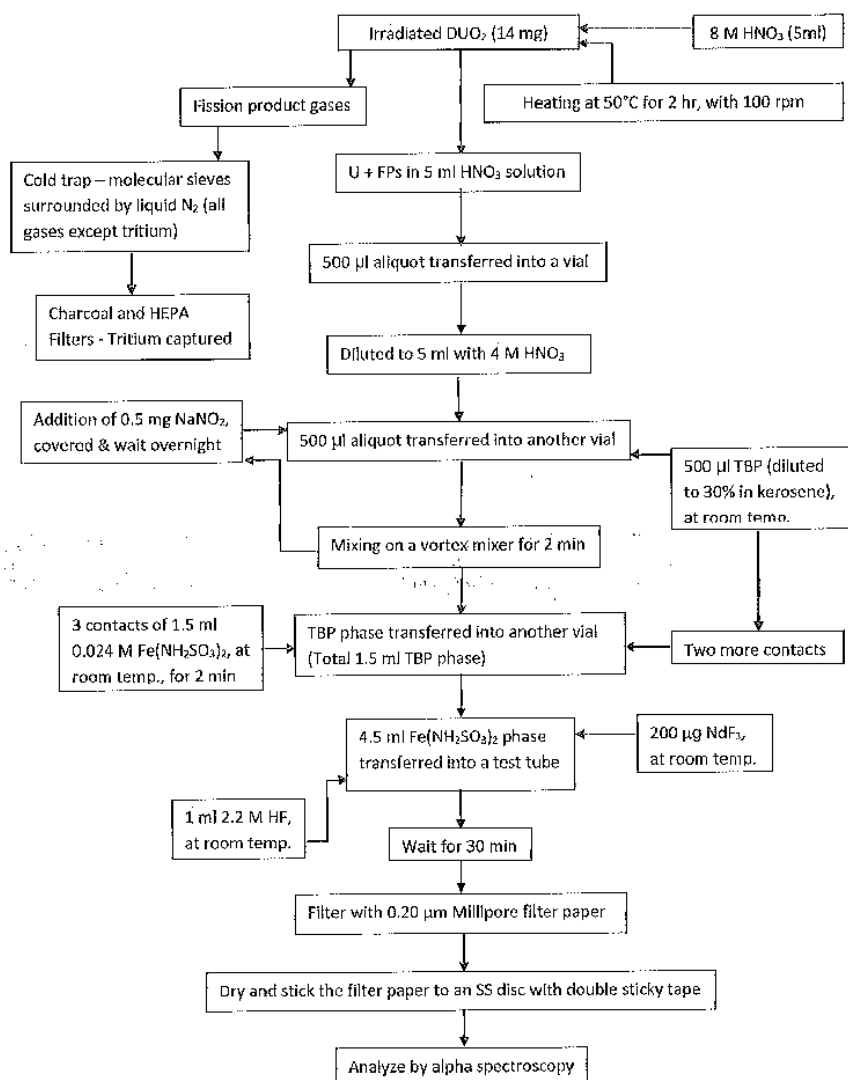


Figure 2. Flow Chart for Experiment.

The aliquot's Pu (III) was converted to Pu (IV) by adding 0.5 mg of NaNO₂ to 500 µl of stock solution with subsequent stirring, and covering. The solution was left overnight so that NO₂ gas completely oxidized Pu (III).

Break Describe the experiments. Describe how you measured data.

This 500 µl aliquot was then contacted with 700 µl of 30% by volume Tri-Butyl Phosphate (TBP), diluted in kerosene, at room temperature. Equal volumes of TBP and stock solution were not used, and an extra 200 µl of TBP was added at contact to create a buffer layer of sorts during pipetting. A similar procedure was used during the plutonium and uranium separation. Both the aqueous and organic phase were mixed on a vortex mixer for 15 minutes at 1500 rpm, and then separated with the help of a centrifuge. Subsequent contacts of 500 µl TBP occurred to ensure the quantitative recovery of heavy metal from the aqueous phase.

The total organic TBP phase was then contacted with an equal volume plus 200 µl of 0.024 M $\text{Fe}(\text{NH}_2\text{SO}_3)_2$ and 0.75 M HNO_3 solution to separate the plutonium from the uranium. Multiple contacts were also done at this stage to extract a large portion of the Pu.

Each of these steps were analyzed with gamma spectroscopy ICP-MS analysis.

Should I talk about verification of U and Pu separation?

Should I talk about Pu extraction verification with alpha spec?

What figures should I include?

ICP - MS Calculations

I could write a long paper on this...probably nobody cares. contaminates, subtractions, approximations.

Results and discussion

Enrichment U

The ICP-MS results indicated an enrichment of $0.283 \text{ wt}\% \pm \text{XX} \%$ and $89.3 \text{ wt}\% \pm \text{XX} \%$ of ^{235}U and ^{239}Pu in the samples. The DUO_2 had an initial ^{235}U concentration of $0.25 \text{ wt}\% \pm \text{XX} \%$. The increase in enrichment occurred due to the fact that the Resonance Integral (RI) (n, γ) cross section for ^{235}U is 133.0 b as opposed to 278.1 b for ^{238}U .^{kaeri} Although ^{235}U has an almost equal RI (n,f) cross section, compared to the ^{238}U (n, γ), the resonance region for ^{238}U begins before ^{235}U . Which is shown in Fig. XX below.^{janis} This, coupled with the larger concentration of ^{238}U atoms, yields an increased enrichment of ^{235}U for DUO_2 leaving a resonance region neutron spectrum. Other phenomena, namely ^{234}U 's non-negligible RI (n, γ) cross section (631.3 b), and build up of other uranium isotopes, either enhance this effect with fresh fuel, because ^{235}U is produced from (n, γ) of ^{234}U , or deter it as burn up increases due to ^{237}U 's large RI (n, γ) cross section of 1084 b. These effects would be weighted differently in a thermal reactor (should I talk about this?). ^{237}U , short half life could give information on cycles. Other people have studied this...citation, blah blah blah. ^{242}Cm and ^{244}Cm both give indication on cycles leaving a reactor (reaches new equilibrium), could U237 contribute to something like that for us? How can I tell how many cycles my fuel went through? I need to know!!

Figure XX. Fission and Capture cross section data for ^{235}U and ^{238}U .

Fuel Burn Up

The burn up of this fuel was roughly estimated (could do better by knowing how many fissions came from where) by analyzing the ^{137}Cs and ^{148}Nd concentrations. Both isotopes were picked due to their relatively constant fission yields across fissile material and due to their relatively stable status. Concentrations were determined with mass spectroscopy and ^{137}Cs additionally was determined with gamma spectroscopy. The results are shown

in Table XX below. These calculations assumed negligible RI absorption cross sections (0.4 b - Cs, 14.7 b Nd) .

Enrichment Pu

Plutonium enrichment was also determined through ICP-MS and was found to be 89.3% \pm XX %. This was expected due to prior simulation results.^{swinney} What was not expected was the percentage that was due to Pu.

PUREX Process

Experiment 1 Actinides

Three runs were carried out under similar experimental conditions described above. The uranium saturation in all solutions was less than 0.023% and thus its variable condition was negligible throughout the experiment. Under these conditions three different experiments were conducted. The first experiment with 4 contacts of TBP and 3 contacts of 0.024 M iron sulfamate, 4 M HNO₃ extracted 99.1% \pm XX% and 99.9% \pm XX% of the original Pu and U into TBP and 96.0% \pm XX% and 6.9% \pm XX% respectively were extracted into the iron sulfamate. The iron sulfamate was treated with neodymium(III) fluoride and 2.2 M HF to co-precipitate the plutonium with neodymium. The precipitate was filtered through a 0.22 μ m millipore filter and is shown in Fig. XX.

Figure XX. Co-Precipitated Plutonium (OTHER PRECIPITATE?) on 0.22 μ m millipore filter paper.

The plutonium precipitate was qualitatively analyzed using a PIPS alpha detecting system. Due to layering of the precipitate and partial absorption into the filter paper, the spectrum, shown in Fig. XX, had energy peak at at 5.15 MeV with a continuum

extending to 3.5 MeV. This clearly indicates ^{239}Pu was extracted. Smaller amounts of ^{241}Am were extracted as indicated by the smaller 5.5 MeV peak. A peak wide count determines that roughly $10\% \pm \text{XX}\%$ (correct for different isotopes $0.26\ \mu\text{g}$) of the extracted plutonium was counted, geometric considerations considered, by the alpha detector - while the rest was caught up in the filter matrix.

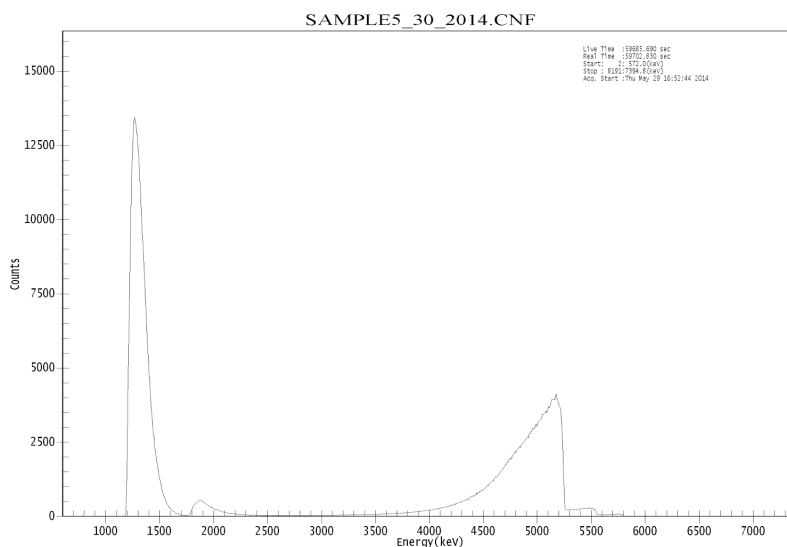


Figure XX. Alpha spectrum of co-precipitated plutonium.

More details about first experiment. second cycle (U and Pu)

Experiment 2 Actinides

The second experiment, with a single contact of TBP and a single contact of 0.75 M HNO_3 , 0.024 M iron sulfamate solution did not utilize sodium nitrite to convert Pu(III) to Pu(IV) . A calculated $94.2\% \pm \text{XX}\%$ and $97.4\% \pm \text{XX}\%$ of Pu and U were extracted into TBP and $158\% \pm \text{XX}\%$ and $23.1\% \pm \text{XX}\%$ were respectfully extracted into the iron sulfamate.

This experiment was able to isolate $50 \pm \text{XX}\%$ of the original Pu and separate out $80\% \pm \text{XX}\%$ of the original uranium. Overall, 94.4% plutonium was recovered from the

irradiated UO₂ pellet. While, during the last step of PUREX process, 94% uranium was separated from plutonium. The separated plutonium hasPu-239,

Experiment 3 Actinides

Experiment 1 FP (radioactive - stable - compare to books, amounts, DF factors)

Experiment 2 FP (radioactive - stable - compare to books, amounts, DF factors)

Experiment 3 FP (radioactive - stable - compare to books, amounts, DF factors)

| Sample results (ng/mL) | ²³⁴ U | ²³⁵ U | ²³⁶ U | ²³⁷ Np | ²³⁸ U | ²³⁹ Pu | ²⁴⁰ Pu | ²⁴¹ Pu + ²⁴¹ Am | ²⁴² Pu |
|---|------------------|------------------|------------------|-------------------|------------------|-------------------|-------------------|--|-------------------|
| 239Pu sample 140620 | 0.208 | 37.2 | 1.83 | 0.439 | 13000 | 217 | 18.3 | 6.89 | 0.394 |
| <i>Italicized results are approximate</i> | | | | | | | | | |
| Sample limits of detection (ng/mL) | ²³⁴ U | ²³⁵ U | ²³⁶ U | ²³⁷ Np | ²³⁸ U | ²³⁹ Pu | ²⁴⁰ Pu | ²⁴¹ Pu + ²⁴¹ Am | ²⁴² Pu |
| 239Pu sample 140620 | 0.00095 | 0.00095 | 0.00097 | 0.00095 | 0.65 | 0.019 | 0.0025 | 0.0011 | 0.00097 |

Fission product separation percentage by isotope is shown in the first table.

Table I

| Isotope | Activity predicted by MCNPX (Ci/gm) | Final Measured Activity (Ci/gm) | Measured Uncertainty (Ci/gm) | Percent Error (%) |
|-------------------|---|------------------------------------|------------------------------------|----------------------|
| ¹⁴⁴ Ce | 7.29E-02 | 9.14E-02 | 3.E-03 | 20 |
| ¹³⁴ Cs | 2.02E-03 | 2.01E-03 | 4.E-05 | -0.6 |
| ¹³⁷ Cs | 1.15E-02 | 1.47E-02 | 4.E-04 | 21 |
| ¹⁵⁴ Eu | 1.79E-04 | 1.70E-04 | 7.E-06 | -5.3 |
| ¹⁰⁶ Rh | 7.38E-02 | 6.90E-02 | 2.E-03 | -6.8 |
| ¹²⁵ Sb | 1.55E-03 | 1.27E-03 | 4.E-05 | -22 |
| ⁹⁵ Zr | 5.07E-03 | 4.82E-03 | 1.E-04 | -5.3 |

These results agree well with previous literature for PUREX experiments^{ref}. Each contact of TBP and iron sulfamate should yield similar separation percentages in subsequent experiments. The waste stream concentrations of U and Pu show that each contact of TBP should remove **XX% ± XX%** of plutonium and **XX% ± XX%**. These numbers would correspond to separation ratio(?) numbers of XX and YY, respectively.

The second experiment repeated the first, with four contacts of TBP and three iron sulfamate contacts. Sodium nitrite was used to convert **Pu (IV) to Pu (III)** and the cycle

was repeated a second time in order to further reduce contaminants. These results isolated $87\% \pm XX\%$ of the original Pu and separated out $99.7\% \pm XX\%$ of the original uranium. Fission products propagated through the solution in a similar fashion as before with these noted differences (any noted differences?).

The third experiment was a variant on the second, conducted at a different nitric acid concentration. Three cycles were performed and $87\% \pm XX\%$ of the original Pu was produced with $99.7\% \pm XX\%$ of the original uranium separated out. Fission products propagated through this solution in a with slightly different trends because of the different nitric acid concentrations. These results are shown in table XX

Table xx

Isotopes x and x are related in this way.

Conclusions

It was discovered that isotopes X Y and Z propagate through the PUREX process as such...Pu was able to be isolated at this efficiency per contact. In Conclusions you should not repeat sentences from the Abstract, Introduction, and the Results sections. It should summarize the most important results, their novelty advantages, and limitations. Here you may also mention planned future work and/or recommendations to others.

Based on the results, it was determined that the fuel sample was originated in a fast-spectrum reactor with a burn-up of 3.5 GWD/MTU.

Acknowledgements

Acknowledgments of people, grants, funds, *etc.* should be placed here. The names of funding organizations should be written in full.

References

Supplementary information

This section will not appear in the printed version of your paper but it will contain a link; the webpage containing the electronic supplementary information will appear when one clicks on the hyperlink. Here you can list the details of your research which would be too long for the main text, *e.g.* a larger number of spectra *etc.* Start with 1 for Figure and Table numbers in this section.

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