# PUREX separation of plutonium and uranium from fission products to measure elemental decontamination factors

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$^{239}$Pu is produced as a byproduct of $UO_2$ irradiation in a nuclear reactor, among other actinides and many fission products (FPs). These FPs are mostly elements of the 5th and 6th rows of the periodic table. The distribution of FPs relative to $^{239}$Pu in the material is dependent upon multiple factors, including the burnup of the fuel and the neutron spectrum within the reactor. Therefore, by measuring the FP:$^{239}$Pu ratios within a spent fuel sample, we can learn more about the history of its irradiation [1]. This is an important analysis for nuclear security because Pu is fissile and may be used to make nuclear weapons.

It is likely, however, that any intercepted Pu will have already been chemically separated from the FPs. The most common process for Pu purification is the PUREX process, which employs typically 30% tributyl phosphate in kerosene or n-dodecane to selectively extract Pu and U from the other species present in spent fuel. If the decontamination factor (DF) of each FP is known throughout the PUREX process, the original FP:$^{239}$Pu ratio before purification may be obtained. It is rare, however, for reprocessing facilities to publish the DFs of each FP throughout their process [2]. Recent work this year has focused on obtaining the DFs for attribution elements like Cs and Eu throughout the PUREX process used at the Barnwell Nuclear Fuel Plant (BNFP), which was built in South Carolina but never operated commercially.

The process employed at the BNFP consists of 11 major steps. Within these steps, the facility would have accomplished codecontamination (purification of Pu and U together), partitioning of the feed into separate Pu and U streams, and then final purification of each stream. The first challenge in the current work was downsizing the 11-step industrial process to a smaller scale experiment that could be completed in the laboratory. The source for the process characterizes the Ru-Rh decontamination through each step [3], which serves as a benchmark to ensure the lab-scale process matches the specifications of the industrial reprocessing plant. Afterwards, experiments were performed to maximize the oxidation of Pu(III) to Pu(IV) using sodium nitrite. This is a crucial oxidation for the first step of the process. Experiments confirmed the optimal conditions from a Hanford study to be elevated temperatures (70 $^{o}$C), low mass loading (4:1 Pu:$NaNO_2$), and an hour-long reaction time in 3 M $HNO_3$ [4]. The second challenge to overcome was the difference in U loading between the lab-scale and industrial-scale process. Because the sample of irradiated $UO_2$ in possession here is quite small (approximately 13 mg), the concentration of U in solution is roughly 1000 times less than in the industrial plant. This difference in concentration is expected to impact the DFs of the FPs. As a result, a study was performed to determine the magnitude of this impact. An initial control experiment was performed for the first step of the PUREX process as designed by the BNFP using our unaltered irradiated $UO_2$ sample. The DFs for several elements were obtained, as seen in Table I. A second experiment where depleted $UO_2$ will be added to our sample, such that the concentration of U in solution matches that used in the BNFP process, will be completed shortly hereafter and compared to the preliminary results in Table I.

**Table I.** Decontamination factors (DFs) for elements in step one of the control study. The U concentration in the initial aqueous solution was 0.93 mM.

| **Nuclide** | **Decontamination factor** |
|---|---|
| $^{241}$Am | 7.6 ± 0.9 |
| $^{144}$Ce | 11.2 ± 0.7 |
| $^{154,155}$Eu | 6.1 ± 0.5 |
| $^{125}$Sb | 62 ± 11 |
| $^{106}$Ru | 5.6 ± 0.6 |
| $^{134,137}$Cs | 3500 ± 1300 |

In most cases, errors may be improved with longer counting times. For Cs, the error is quite high because the DF is very large and the element reaches its detection limit easily. The DF of each element will continue to increase as further steps of the process are performed. Pu recovery was measured by alpha spectrometry and meets the expected 98% recovery through step one.

The results so far present new elemental DFs for step one of the BNFP process with one thousandth of the U concentration. Future work will include the elemental DFs for all 11 major steps of the BNFP process with the proper U concentration.

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