



Original Article

Plutonium mass estimation utilizing the (α, n) signature in mixed electrochemical samplesStephen N. Gilliam^{a,*}, Jamie B. Coble^a, Braden Goddard^b^a University of Tennessee-Knoxville, Nuclear Engineering Department, Knoxville, TN, 37996, USA^b Virginia Commonwealth University, Department of Mechanical and Nuclear Engineering, Richmond, VA, 23284, USA

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ABSTRACT

Quantification of sensitive material is of vital importance when it comes to the movement of nuclear fuel throughout its life cycle. Within the electrorefiner vessel of electrochemical separation facilities, the task of quantifying plutonium by neutron analysis is especially challenging due to it being in a constant mixture with curium. It is for this reason that current neutron multiplicity methods would prove ineffective as a safeguards measure. An alternative means of plutonium verification is investigated that utilizes the (α, n) signature that comes as a result of the eutectic salt within the electrorefiner. This is done by utilizing the multiplicity variable α and breaking it down into its constituent components: spontaneous fission neutrons and (α, n) yield. From there, the (α, n) signature is related to the plutonium content of the fuel.

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1. Introduction

As interest grows in alternative methods to aqueous reprocessing, safeguards and process monitoring efforts must adapt to meet the specific needs of the system environment. The system of electrochemical reprocessing introduces unique challenges within its electrorefining unit as special nuclear material, plutonium, is kept in a mixed state with other spontaneous fission components, such as curium, of the fuel. This makes verification of plutonium a difficult task when using neutron-dependent signatures as the spontaneous fission yield that arises from curium overwhelms the contribution made from plutonium. Although varying with fuel composition, approximately 95% of the neutrons seen in oxide fuels come from spontaneous fission reactions with upwards of 95% of those originating with ²⁴⁴Cm specifically [1]. Furthermore, since plutonium and curium are not held in a constant ratio due to chloride affinity during the extraction procedure; methods that utilize this ratio are not effective [2].

Although methods such as passive neutron albedo reactivity have been proposed for plutonium tracking in electrochemical systems, these techniques are applied at the end processes once ingots have already been formed in the absence of eutectic salt [3].

This leaves a gap at the electrorefiner for potential material diversion. Previous research has delved into the isotopic composition within the electrorefiner as well as a breakdown of the spontaneous fission and (α, n) source terms. It was found that for low burnups, the (α, n) signature was of significant magnitude to be considered non-negligible [2]. The proposed method aims to quantify the plutonium present in the electrorefiner via the (α, n) source term. It utilizes the unique eutectic salt of the electrorefiner that gives rise to a substantially higher (α, n) signature than has been seen in aqueous systems or end ingot products. This signature is combined with neutron multiplicity calculations to deconstruct the known but underutilized variable α which demonstrates the ratio of (α, n) to spontaneous fission neutron yield. Preliminary simulations were conducted using test samples containing curium, plutonium, and eutectic salt to demonstrate the efficacy of this approach. The mass of curium is first deduced through multiplicity point model calculations. From there, the mass of curium is related back to the plutonium mass via the multiplicity variable α .

2. Background

2.1. Electrochemical reprocessing

The core of the electrochemical process is the electrorefiner shown in Fig. 1 [4]. This unit is responsible for partitioning the chopped fuel into streams containing uranium and uranium plus

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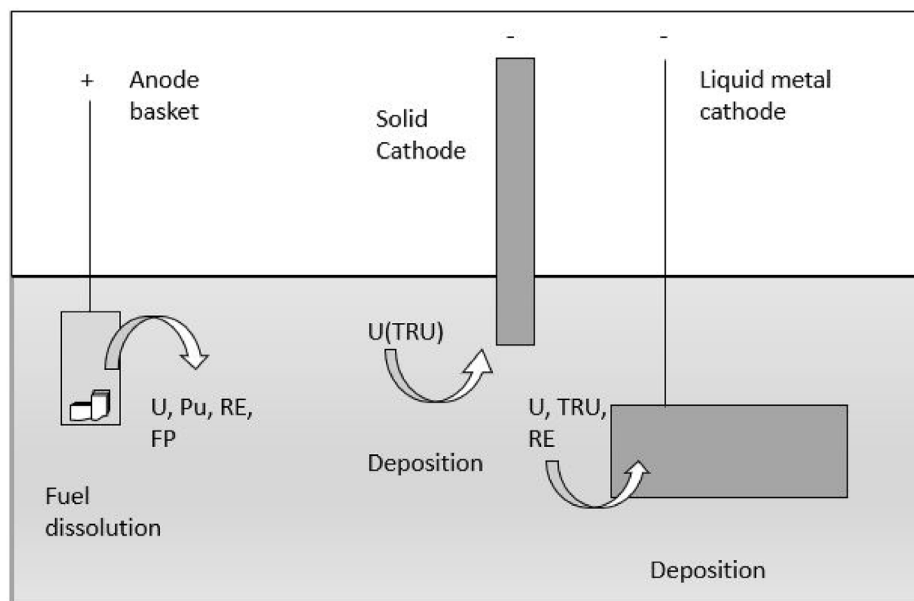


Fig. 1. Operational diagram of the electrorefiner depicting the deposition of fuel constituents, rare earths (RE), fission products (FP), uranium (U), and plutonium (Pu) into a eutectic salt. The elements are then shown to migrate onto the respective collection cathode based on extraction potential [4].

transuranics for further processing. At the beginning of the separation, metallic fuel inside an anode basket is dispensed into a pool of LiCl–KCl salt [5]. If the fuel is not in a metallic state, a reduction process is undergone before the electrorefiner unit. From there, material will transport from the basket into the salt based on the affinity for chloride formation better known as Gibbs Free Energy [6]. After migration, a solid cathode is then dispensed into the salt and a potential is run between it and the anode basket. The weaker chloride bonds formed with uranium are then broken and uranium is extracted at the surface of the cathode. The potential is set so that the majority of uranium is extracted at this time. Due to the closeness in strength of the chloride bonds, the solid cathode may also extract trace amounts of plutonium as well [4,6]. Once enough uranium has been extracted, a second cathode, typically liquid cadmium, is dispensed and the potential is increased [7]. This increased potential is now strong enough to break the chloride bonds formed by the transuranics. However, although the transuranics are extracted onto the second cathode together, the rate of extraction is based upon the strength of their chloride bond. This makes the value of the Pu/Cm ratio non-constant and eliminates it as an option for quantifying plutonium in the mixed sample [8]. Remaining fission products segregate between the anode basket and the molten salt while noble metals with lower potentials, such as rhodium and ruthenium, remain in the anode basket [9]. There is also some minor migration of the salt onto the cathodes as well as minute traces of materials with stronger chloride bonds, such as rare earths. It is for this reason a drawdown purification occurs to cleanse the cathodes before processing [6].

Under conditions present in aqueous reprocessing, oxygen that accompanies the oxide fuel acts as the main target for (α, n) reactions. If hydrofluoric acid is present then fluorine may also be a target. Neither of these aqueous targets are in a suitable abundance to produce significant interaction for a high (α, n) yield, especially when compared to the spontaneous fission neutron production. Conversely, electrochemical reprocessing proposes a vast salt pool in comparison to the batch size of the fuel entering the electrorefiner. This means that the α -particles have a higher probability for producing an (α, n) reaction as there are more targets, namely ^7Li and ^{37}Cl , to interact with [2].

2.2. Neutron multiplicity counting

In order to quantify the amount of plutonium present in a sample a nondestructive means, neutron multiplicity counting, has been used. This technique utilizes the measured number of spontaneous fission neutrons detected from a single fission event and relates it back to the quantity source term material. Assuming a well known detector efficiency, the results yield three parameters: the sample's self multiplication (M); the ratio of (α, n) neutrons produced to spontaneous fission neutrons (α); and the effective mass of the leading spontaneous fission contributor of the sample, typically $^{240}\text{Pu}_{\text{eff}}$ [10]. An alternative variable, efficiency (ϵ), can be calculated in place of the sample's self multiplication variable. For these instances, the multiplication factor is assumed to have a value of one [10]. In order to accurately measure the $^{240}\text{Pu}_{\text{eff}}$, a pure sample containing plutonium as the only spontaneous fission source must be used. If an isotope with a high spontaneous fission yield, such as the curium found in used nuclear fuel, is present it could interfere with the calculation as the spontaneous fission yield from curium would overshadow that from plutonium [1]. There are methods, however, to get around this but they require strict conditions such as a negligible (α, n) contribution or a constant ratio between the plutonium and curium content. Neither of these conditions hold true for electrochemical reprocessing due to a high (α, n) yield and an ever changing ratio due to various rates of extraction onto the cathodes [2,11].

3. Methodology

3.1. Curium mass calculation

Although the equation to solve for the $^{240}\text{Pu}_{\text{eff}}$ mass is well documented, this work relies on developing an equation for the $^{244}\text{Cm}_{\text{eff}}$ mass. What follows is a derivation of such an equation using [12]. For doubles counting with a neutron detector with a flat neutron energy response, we define the $^{244}\text{Cm}_{\text{eff}}$ coefficient of nuclide i relative to ^{244}Cm , (γ_i), on a per-unit-mass basis, in terms of

basic nuclear data by the following relation (Eq. (1)), where (g) is the specific spontaneous fission rate, in terms of fissions per unit time per unit mass, and (ν_2) is the second factorial moment of the normalized spontaneous fission prompt neutron multiplicity distribution. The specific spontaneous fission rate is calculated from the spontaneous fission half-life via Eq. (2). Here the molar mass is represented by (A) and (N_A) is Avogadro's number. Combining Eq. (1) and Eq. (2) yields the final nuclide coefficient formula (Eq. (3)).

$$\gamma_i = \frac{(g^* \nu_2)_i}{(g^* \nu_2)_{244}} \quad (1)$$

$$g = \frac{1}{A} N_A \frac{\ln(2)}{t_{1/2}} \quad (2)$$

$$\gamma_i = \frac{\left(\frac{\nu_2}{A^* t_{1/2}} \right)_i}{\left(\frac{\nu_2}{A^* t_{1/2}} \right)_{244}} \quad (3)$$

The molar masses are known to high accuracy, and so the uncertainty in the (γ_i) factors is determined, for all practical purposes, solely by the uncertainties in the second factorial moments and the spontaneous fission half-lives. Propagation of variance by first-order Taylor series expansion to the mathematical expression for (γ_i) gives Eq. (4) where (σ) is the standard deviation. The final equation for the $^{244}\text{Cm}_{\text{eff}}$ mass is given by Eq. (5) where variable $f_{,xx}$ represents the isotopic fraction for that nuclide. This equation can be rearranged to obtain the total curium mass of the sample.

$$\text{Uncertainty}_i = \left(\frac{\sigma_{\nu_2,i}}{\nu_2,i} \right)^2 + \left(\frac{\sigma_{t_{1/2,i}}}{t_{1/2,i}} \right)^2 \quad (4)$$

$$^{244}\text{Cm}_{\text{eff}} = ((1.64 \pm 0.056)f_{242} + f_{244} + (0.84 \pm 0.01)f_{246})m_{\text{Cm}} \quad (5)$$

3.2. Plutonium mass calculation

The mass of plutonium is then solved by deconstructing the variable α into its constituent parts. The method breaks down both the (α, n) and spontaneous fission components as having sources from curium and plutonium. This is shown in Eq. (6) where the spontaneous fission originates from curium with a source strength of $F_{\text{Cm}}(\text{SF})$ and plutonium with a source strength of $F_{\text{Pu}}(\text{SF})$. The numerator gives the (α, n) breakdown with $F_{\text{Cm}}(\alpha, n)$ representing the (α, n) source strength of curium and $F_{\text{Pu}}(\alpha, n)$ representing the (α, n) source strength of plutonium. Rearranging the equation to solve for the mass of plutonium (m_{Pu}) yields Eq. (7).

$$\alpha = \frac{F_{\text{Cm}}(\alpha, n)m_{\text{Cm}} + F_{\text{Pu}}(\alpha, n)m_{\text{Pu}}}{F_{\text{Cm}}(\text{SF})m_{\text{Cm}} + F_{\text{Pu}}(\text{SF})m_{\text{Pu}}} \quad (6)$$

$$m_{\text{Pu}} = \frac{F_{\text{Cm}}(\text{SF})m_{\text{Cm}}\alpha - F_{\text{Cm}}(\alpha, n)m_{\text{Cm}}}{F_{\text{Pu}}(\alpha, n) - F_{\text{Pu}}(\text{SF})\alpha} \quad (7)$$

The source strengths for the (α, n) and spontaneous fission signatures was approximated by using predetermined isotopic ratios. SCALE was then used to calculate the emission rate of the radionuclide dispensed in the accompanied salt.

3.3. MCNP simulation setup

A simulink model titled the Separations and Safeguards Performance Model for Electrochemical Reprocessing (SSPM EChem) [13] was used to obtain isotopic information for nine different fuel cases spanning a burnup of 20–60 GWd with corresponding enrichments to mirror normal operating conditions as defined by Eq. (8) [14]. The mass information obtained from the SSPM EChem was then scaled down to the sample cavity size of 4.11 g for an epithermal neutron multiplicity counter (ENMC) MCNP input deck. The ENMC input deck uses MCNP 6.2 to simulate each measurement made by the ENMC [15]. The MCNP input deck has been used before with MCNPX and examples of its capabilities can be seen in Refs. [16–18].

$$(EN) = 0.31BU^{0.65} \quad (8)$$

The internal MCNP spontaneous fission spectra, $\text{par} = \text{sf}$, was used for all spontaneous fission simulations. This consists of multiplicity data from Peter Samti and energies from a Watt spectrum [15,19]. (α, n) spectra were acquired from an internal ORNL developed program which uses SCALE 6.2 libraries to generate the spectra [20]. The information needed to generate the SCALE spectra was the mass of each constituent isotope including target isotopes. This yielded both the spontaneous fission and (α, n) neutron yield as well as the ratio of spontaneous fission neutrons to (α, n). From there the (α, n) energy distribution was manually put into MCNP. Each simulation ran for 60 min of simulated counting time. The tallies used in the simulations consisted of gated and non-gated F8 coincidence tallies to determine relevant neutron multiplicity values, as well as F4 tallies to determine the neutron flux and fission rate within the measured standard.

Prior to calculating the effective mass of curium, the proper die-away time, Eq. (9) must be determined to ensure accuracy of the resulting count rates. This is done by running the ENMC model with a fixed predelay and varying gate widths. Here, the variables are the first gate width is (G_1), the doubles counts from the first gate (D_{G1}), the doubles counts from the second gate (D_{G2}), and the die-away time (τ). A beginning gate width of $10\mu\text{s}$ was chosen and increased by $2\mu\text{s}$ up to $100\mu\text{s}$. Then the increments increased by $50\mu\text{s}$ until the final value of $250\mu\text{s}$ was reached. To obtain the optimal gate width one must multiply the die-away time by 1.257 [10]. The optimal gate width was determined to be $28\mu\text{s}$.

$$\tau = \frac{-G_1}{\ln\left(\frac{D_{G2}}{D_{G1}}\right) - 1} \quad (9)$$

4. Results

The first step in the proposed method involves the calculation of curium for the mixed sample. Using Eq. (5) the ^{244}Cm mass was obtained, shown in Fig. 2. Here the calculated and actual masses follow the same trend with increasing burnup with the calculated value being greater than that of the actual mass. Thus, an over-estimation is occurring. Examining the percent error between the calculated and actual curium mass, Fig. 3, shows that the low burnup side has the largest error value at -28% . As burnup

increases the error steadily decreases until it begins to plateau at roughly -13%.

To understand this phenomenon the α variable and a comparison made between the mass percent error and the (α, n) contribution to the neutron yield were examined. Fig. 4 shows the comparison between the calculated α from MCNP and the one produced from SCALE emission data. Here, the calculated detector response in MCNP is less than that when looking at emission rates from SCALE. This is somewhat expected due to detection efficiency of roughly 65% for the ENMC. However, a decrease in the neutrons counted when compared to the emission rate doesn't necessarily ensure that the variable α will be less. For this phenomena to occur, either the counts associated with the spontaneous fission source are being overestimated or the (α, n) counts are being underestimated. In other words, the detector doesn't discriminate based on the reaction that produced the neutron and thus some neutrons produced through (α, n) reactions will attribute to the curium mass calculation as if they were produced from spontaneous fission.

Across all nine burnup cases, the ratio between the two is on average 0.465. When the absolute value of the mass percent error is compared to the (α, n) emission rate contribution, Fig. 5, a pattern emerges. The two have similar trends as burnup increases which suggest that the percent error associated with the detector response is closely linked to the contribution made from the (α, n) neutrons. Combining the information provided by the α variable comparison and that made with the (α, n) contribution it appears that the reason for the error in the curium mass might be due to the phenomenon that (α, n) neutrons are tabulated as coming from spontaneous fission reactions. Since most samples tested in multiplicity counting have relatively low (α, n) yields this has not readily been observed nor been an issue [1]. As burnup increases the amount of curium also increases and the plutonium and overall (α, n) contribution to total neutron emissions both decrease. Thus, the mass percent error of curium becomes smaller as burnup increases due to having less influence from the (α, n) neutrons. In other words, the spontaneous fission neutrons overshadow that of the (α, n) source term for higher burnups. If false counting of (α, n) neutrons as that as coming from spontaneous fission is indeed the issue this would make sense as to why the error decreases as the spontaneous fission percent contribution to total neutrons increases.

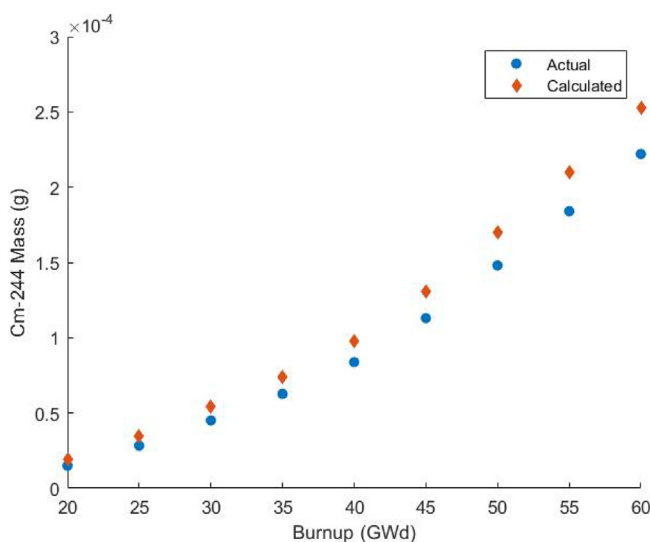


Fig. 2. Comparison between calculated mass and actual mass of Cm-244 for a mixed sample.

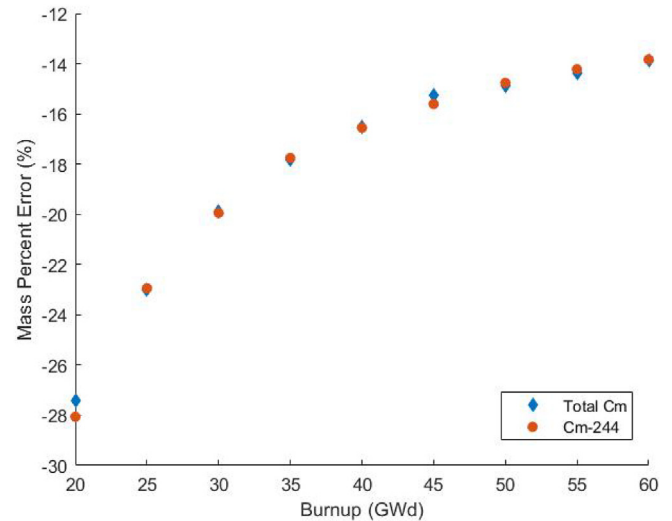


Fig. 3. Percent error between the actual and calculated Cm-244 and total curium mass estimation for a mixed sample.

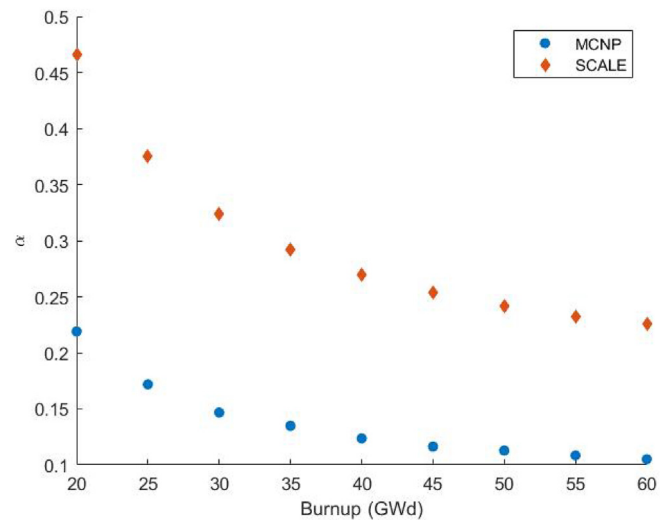


Fig. 4. α variable comparison for a mixed sample.

Next, the plutonium mass was calculated using Eq. (7) and is shown in Fig. 6. Here it is seen that the calculated mass does not match the actual mass and becomes negative starting around 30 GWd burnup. The percent error associated with this is quite high with a low value at 80%. The large error shown in Fig. 7 is due to the error associated with the curium mass calculation, shown in Fig. 3, as well as the lower calculated α value shown in Fig. 4. This also explains the negative plutonium mass cases. For these instances the error from curium mass and α variable carry through the calculations and end with a negative mass calculated.

To attempt to correct for the systematic errors, a correction factor was calculated. This factor examines the ratio between the MCNP α variable to that obtained from SCALE. This will allow a correction in the α variable inferred from the ENMC detector system. Thus, only the error associated with the curium mass calculation will affect the plutonium mass calculation. Fig. 8 shows the correction factor as a function of burnup for each of the nine cases. Each case has a correction factor given for each of its five enrichment values. The range across all cases is 0.453–0.469 with the solid blue line representing the average over the entire range at a

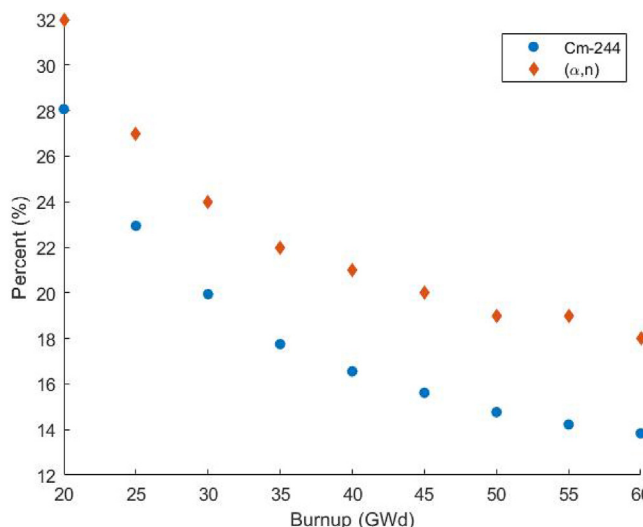


Fig. 5. Comparison between the Cm-244 absolute mass percent error and total (α,n) contribution to neutron yield.

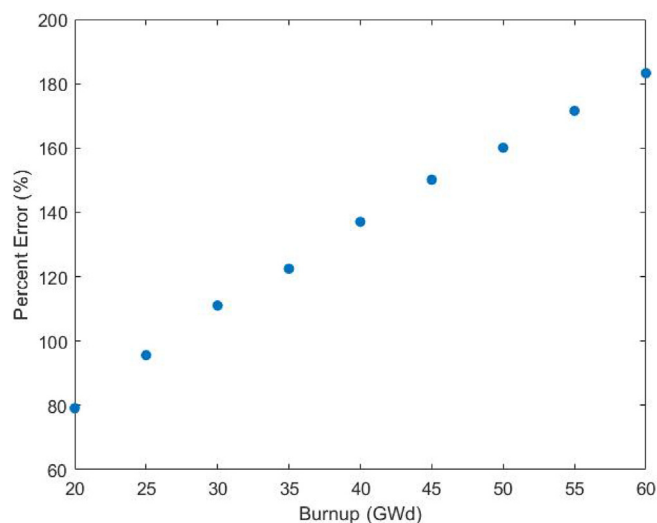


Fig. 7. Percent error for plutonium mass estimation for a mixed sample.

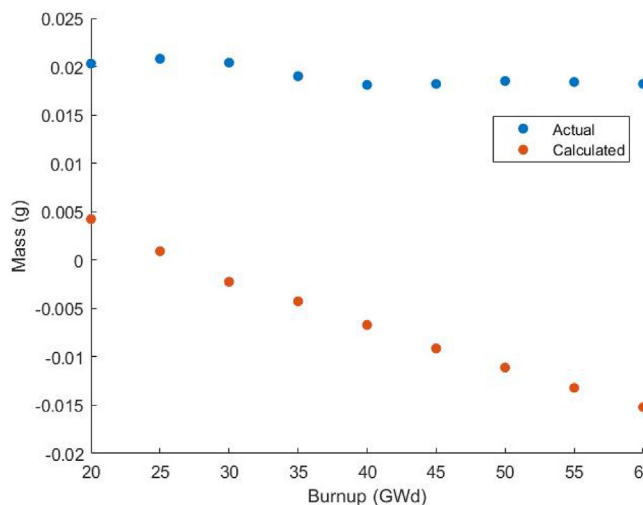


Fig. 6. Comparison between calculated mass and actual mass of plutonium for a mixed sample.

value of 0.465. It is theorized that this ratio is in part indicative of the detector system and thus might change more or less drastically if another neutron multiplicity counter was used. Also, since the ratio changes slightly for different sample compositions one would need to determine if the ratio should be recalculated for every case or if an average is appropriate if a known range of compositions is being measured. For instance, if a new sample within the given burnup and enrichment range in Fig. 8 is presented an average correction value of 0.46 might be appropriate.

To show the optimal results possible the factor associated with the specific case was used which yielded an instance of using the exact α variable. The correction factor results as well as results for a proof of concept case are seen in Fig. 9. The exact α and curium mass were used for the ideal case. Thus, the ideal case demonstrates the efficacy of Eq. (7) using the actual α and curium mass in the sample. Here, a drastic change is seen as the calculated masses are no longer negative and both follow the general trend of the actual mass. The correction factor mass is greater than the actual mass due to it still having error associated with the curium

calculation. The ideal mass matches the actual mass and demonstrates the efficacy of this method.

The percent error for the correction factor and ideal cases is shown in Fig. 10. Here it is seen that the error for the correction factor cases matches that of the percent error for the curium mass shown in Fig. 3. This stands to reason as the only error present in the calculation is that of the curium mass calculation.

5. Summary

Verification of special nuclear material is of special importance when it comes to reprocessing facilities. Certain features of the electrochemical cycle make it challenging to quantify material throughout the process. This may attract hostile adversaries and thus safeguards application must be addressed. When it comes to plutonium verification within the electrorefiner this is especially challenging as curium is also present, which could confound traditional nondestructive assay approaches based on the spontaneous fission signature. However, the eutectic salt present gives rise to a higher magnitude (α,n) signature that can be related back to plutonium content.

The (α,n) signature was examined across nine burnup cases through an MCNP detector response input deck of an ENMC. From there a method was evaluated that deconstructed the multiplicity variable α into its source components to isolate the plutonium contribution. It was seen that great error occurred during the calculation of curium due to no detector discrimination between the reaction source of neutrons. This then carried over into the plutonium mass calculations and resulted in corresponding error. A correction factor was applied to reduce associated error arising from the α variable and gave an error range of 28-13%. Although this is not ideal it demonstrates that if the error associated detector discrimination can be addressed, this method may have potential for application when used in conjunction with other techniques. Reducing error associated with detector neutron source discrimination is a challenging task and currently limits the method as a stand-alone technique.

Moving forward, the first issue to address is the neutron source discrimination associated with calculating the ²⁴⁴Cm mass. Performing measurements that isolate the natural energy distribution between the neutrons could improve mass quantification accuracy [2]. The challenge in doing this is that multiplicity techniques

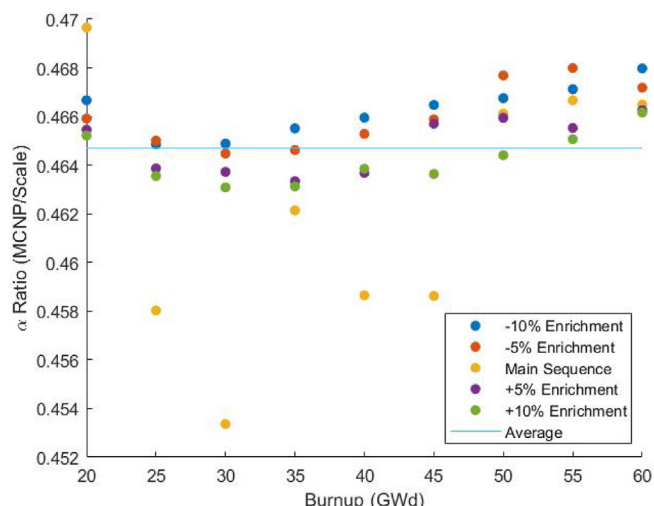


Fig. 8. α variable correction factor.

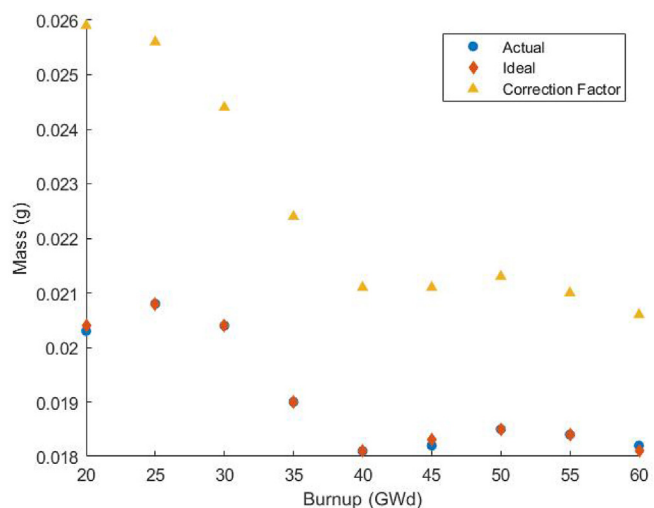


Fig. 9. Comparison between actual mass, the calculated mass with a corrected for the real α , and the ideal case which uses the actual α variable and curium mass. This is for a mixed plutonium sample.

assume the same energy spectrum due to the polyethylene moderator thermalizing the neutrons. Different detectors would need to be examined and designed to accomplish this task. If this can be done then the error using the described method for calculating plutonium will more closely mirror that of current implemented safeguards methods shown for single spontaneous fission source samples.

Once error is reduced, the next step is to simulate the process by adding ^{241}Am into the sample. This isotope is the third largest (α, n) contributor and is present within the electrorefiner. Thus, it is important to know how the higher (α, n) count contributed from this isotope will effect the steps taken in the proposed analysis explained in this document. It is important to reduce the accidental coincidence first as it is theorized adding ^{241}Am to the mixture will only increase the error associated with such. Afterwards other constituents of the fuel can be added to give a more holistic view of the composition likely to be seen within the electrorefiner. With little to no more major (α, n) or spontaneous fission sources, however, adding the remaining fuel constituents likely won't effect the overall outcome. This is still important to aid in developing the

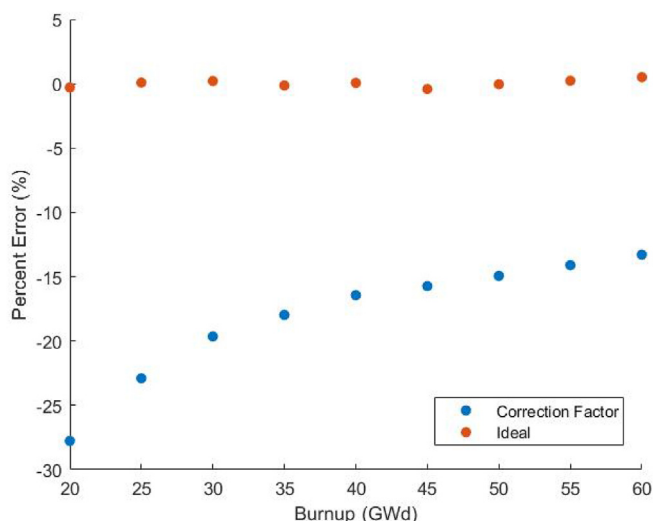


Fig. 10. Percent error for the corrected and ideal plutonium mass estimation for a mixed sample.

procedure when sampling from the electrorefiner. When taking a sample from the electrorefiner, multiple factors come into play with regard to its composition. These factors include: the location the sample was taken from (e.g. the depth as well as the proximity to either cathode or anode), the point in extraction process at which the sample is taken (e.g. mass migration, single extraction, and coextraction), and the batch number it is since the vessel was last cleaned. Performing physical experiments and simulation focusing on the thermodynamic properties of the fuel will also allow for a better understanding on the influence of where and when sampling should occur within the electrorefiner.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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