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A Concept for Quantitative NDA Measurements of Advanced Reprocessing Product Materials

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ABSTRACT: As new reprocessing methods for spent nuclear fuel are developed, such as the uranium extraction (UREX) process, methods using nondestructive assay (NDA) techniques must also be developed to allow for quantitative measurements of product materials. Currently developed NDA techniques cannot directly quantify materials containing U, Np, Pu, and Am. This research investigates the ability to quantify these actinides in an oxide form using neutron multiplicity measurements. This technique assumes that the isotopic composition of the sample is known, either through gamma spectroscopy or other means. This measurement technique is based on performing three different neutron measurements and analyzing their neutron multiplicity response. The first is a passive measurement of the product material to determine the effective plutonium-240 ($^{240}Pu_{eff}$) content, self multiplication (M), and alpha-neutron reaction rate (α). The second is an active, AmLi (α, n) source, measurement of the product material to determine the effective ^{235}U content. The third is an active, AmB (α, n) source, measurement of the product material to determine the effective ^{237}Np content. The quantity of Am in the sample can be determined from α. Simulated results using Monte Carlo N-Particle eXtended (MCNPX) version 2.6 will illustrate the viability of this technique and its practical limitations.

KEYWORDS: *nonproliferation, safeguards, nondestructive assay, neutron multiplicity, reprocessing, MCNPX, epithermal neutron multiplicity counter*

I. INTRODUCTION

The ability of inspection agencies and facility operators to measure powders containing several actinides is increasingly necessary as new reprocessing techniques and fuel forms are being developed[(1-5)]. These powders are difficult to measure because neutrons emitted from induced and spontaneous fission of different nuclides are very similar. Traditional methods require destructive assay (DA) of the sample. Nondestructive assay (NDA) methods are often preferable to DA methods because they are generally faster, cheaper, and have a smaller impact on the facilities operations. Quantitative measurements of these powders are possible without using DA methods by exploiting isotope-specific nuclear properties, such as the energy-dependent fission cross sections and the neutron fission multiplicity.

II. Background

Neutron multiplicity measurements have been used for many years to measure a variety of samples ranging from fuel rods to bulk U or Pu oxide powders. These measurements have traditionally been made on pure samples consisting of only one actinide element, usually Pu, due to the difficulty of determining the source of neutrons. Neutrons produced from U or Pu are distinguished by their energies and the number of neutrons produced per event. For many years, such drawbacks of neutron multiplicity counters have been non-problematic due to the disadvantages of reprocessing used nuclear fuel. However, as reprocessing becomes more popular, and new reprocessing methods are investigated, the production of multi actinide fuels and waste forms are likely to increase.

III. Theory

The Epithermal Neutron Multiplicity Counter (ENMC), shown in Fig. 1, is a neutron multiplicity counter which can detect neutrons created individually or in multiples[(6)]. Since fission is the dominant source of neutrons created in multiplicity, when at least two neutrons are detected within a short time interval, they are considered to come from a single fission event. When the ENMC detects two or three neutrons within a predetermined time gate, they are counted as a doubles or triples count, respectively. When only one neutron is detected in the time interval, it is counted as a singles count. The ENMC detects neutrons through neutron capture via 121 10-atm ^{3}He tubes in closely packed rings embedded in polyethylene[(7)].

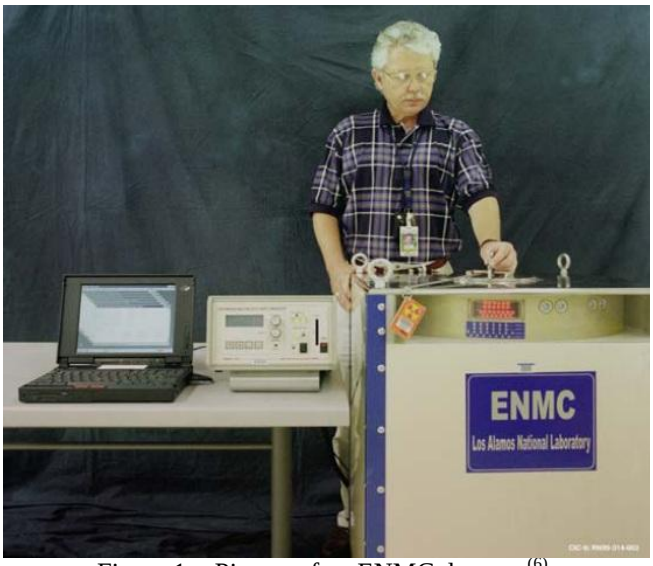

Figure 1. Picture of an ENMC detector[6].

When a fission event occurs, all neutrons released from the event are created at the same instant, yet they are not all detected at the same instant. This is due to the fact that fission neutrons are born fast and must slow down to epithermal or thermal energies via the aforementioned polyethylene before being captured by the ^{3}He tubes. Because of this, the time interval, or gate width, must be defined so that a large portion of neutrons from a fission event can be captured. Typically set at 22 μs for the ENMC[7], this gate width depends on the detector and the type of measurement. This is approximately equal to the time it takes for a neutron to thermalize and be captured. It is common for only one of the neutrons released from a fission event to be detected thus being counted as a single. In addition to this, two different (α,n) neutrons can be detected within the gate width and counted as a double, this is referred to as an accidental. To discount these accidental events, a second neutron count gate is set several seconds after and subtracted from the first gates count rate. This method has been proven precise for determining the Pu mass of a sample. Another source of neutron counts that must be accounted for are self induced fissions in the actinide sample. This is referred to as the self-multiplication of the sample (M) and is accounted for when determining the Pu mass of the sample.

The ENMC is traditionally operated in a passive mode, in which it passively detects spontaneous fission neutrons from the sample. However, design modifications have been developed to operate the ENMC in an active mode, in which fission is induced in the sample by external neutron sources above and below the sample[8]. The most common material used as a neutron source for active interrogation is AmLi, which is placed above and below the sample of interest. When using the International Atomic Energy Agency (IAEA) Neutron Coincidence Counting (INCC) software program, the data output of the active interrogation measurement is a ^{235}U mass or, in passive mode, a ^{240}Pu-effective mass ($^{240}Pu_{eff}$). Effective mass refers to the amount of material of the effective mass isotope that would be required to produce the same doubles count rate. This output does not give any information about the composition of the samples.

When determining the mass of a sample which contains only one actinide, there are four unknown variables that must be determined, 1) the efficiency of the detector (ε), 2) the self-multiplication of the sample (M), 3) the ratio of the alpha-induced neutron production rate to the spontaneous fission rate of the sample (α), and 4) the effective mass of the sample, ^{235}U or $^{240}Pu_{eff}$. For each of these unknown variables, either an independent equation must be created or an educated guess must be made. An educated guess can be made through measurements of known standards or computer simulations, while the independent equations are formed from the singles, doubles, or triples count rates from the sample being measured.

For samples containing multiple actinides, an additional unknown effective mass is added for each additional actinide contained in the sample. Because of this, materials containing U, Np, Pu, and Am require more independent equations. To create these independent equations, a passive and an active measurement, as well as an additional active measurement using an AmB interrogation source, were made. From these three measurement modes, five independent equations can be acquired: one from the passive singles count rate, three from the doubles count rates of each measurement mode, and one from the passive triples count rate. Active mode singles count rates are dominated by the (α,n) interrogation sources, thus making them useless in most situations. The active triples count rates often have statistical uncertainties that are too large for practical use. By combining the five useful independent equations with known information about the sample and the ENMC, the masses of U, Np, Pu, and Am can be quantified, assuming that their isotopic fractions are known.

Active measurements made with either the traditionally-used AmLi source or an AmB source would be identical with the exception of the energy of the fission-inducing neutrons. Neutrons emitted from an AmLi source have an average energy of 0.3 MeV and have an energy distribution as seen in Fig. 2[9]. This average energy and distribution are quite different from an AmB source which has an average energy of 3 MeV and an energy distribution as seen in Fig. 3. Due to the difference in neutron energies, the neutron count rates for these sources yield independent equations. When comparing the induced fission cross-sections of different actinides at 0.3 MeV and 3 MeV, it can be seen that there would be a significant difference in the fission rate depending on the energy of the neutrons used. Figure 4 shows the induced fission cross-section for several common actinides[10].

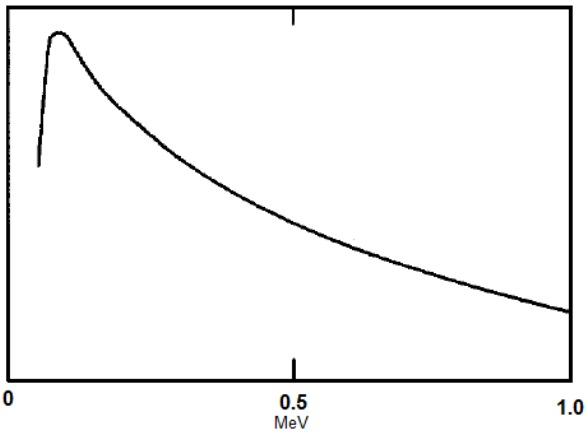

Figure 2. Neutron energy spectrum from an AmLi source[9].

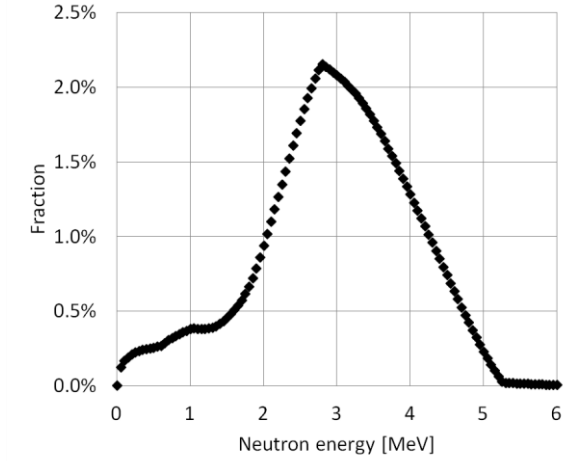

Figure 3. Neutron energy spectrum, generated from SOURCES4C, for an AmB source[11].

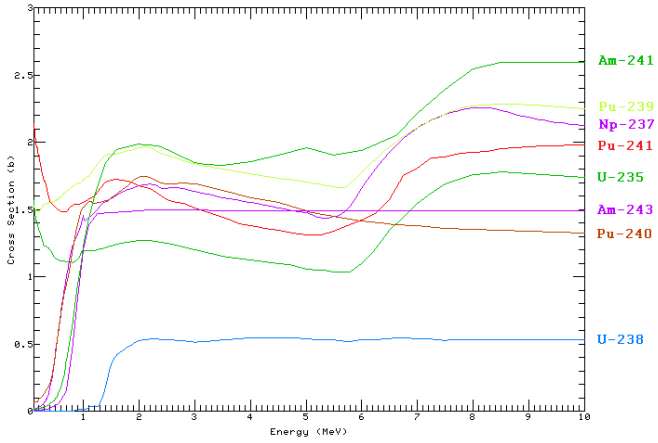

Figure 4. Fission cross-section for common isotopes of U, Np, Pu, and Am[10].

IV. Boron-Carbide

Due to the design of the ENMC, a significant portion of the neutron flux which enters the measurement sample has epithermal energy. This is due to scattering within the polyethylene. Figure 5 shows the energy dependent fission rate within a 1kg ^{235}U oxide sample for different thickness of boron-carbide (B_4C) using an active AmB source. It can be seen in Fig. 5 that without any B_4C shielding a significant portion of the fissions occurring in the ^{235}U are caused by epithermal neutrons being absorbed in fission resonances. Neutrons at these energies are absorbed predominately on the surface of the sample, thus creating a heterogeneous fission rate in the sample. Instead of using a sample coupling method[12], it was determined that placing a B_4C cylinder around the sample would eliminate heterogeneous fissions in the sample without requiring the use of calibration curves. From Fig. 5 it can be seen that a B_4C thickness of at least 0.25 cm greatly reduces the epithermal neutron induced fission rate in the sample. An optimal thickness of 0.5 cm was chosen based on reducing epithermal fissions while minimizing the reduction in fast fissions. The B_4C used consisted of a boron enrichment of 96% in ^{10}B, a density of 90% theoretical, and a container thickness of 0.5 cm on the side, top and, bottom[13]. The outside height and diameter of the B_4C container were 13 cm and 11 cm, respectively.

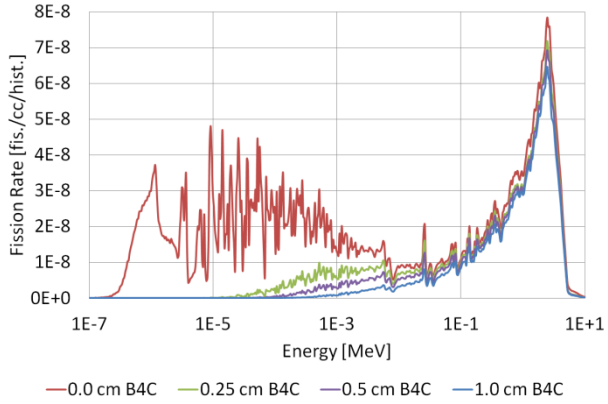

Figure 5. Energy dependent fission rate within a 1kg ^{235}U oxide sample for different thickness of a B_4C using an active AmB source.

V. Calculations of Effective Mass

The active neutron coincidence counting doubles equation is given by[14]:

$$\left(D_{Li}\text{-}D_{Passive}\right) = \frac{F_o\,{}^{239}Pu_{eff\_Li}\varepsilon^2 f_d M^2 \nu_{Li2}}{2}\left[1+\frac{(M\text{-}1)\nu_{Li1}\nu_{Fis2}}{(\nu_{Fis1}\text{-}1)\nu_{Li2}}\right] \quad \text{(Eq. 1)}$$

where D_{Li} is the doubles count rate from the AmLi measurement, $D_{Passive}$ is the doubles count rate from the passive measurement, F_0 is the specific fission rate, $^{239}Pu_{eff\_Li}$ is the effective mass of ^{239}Pu in the sample that would produce the same AmLi doubles count rate, ε is the neutron detection efficiency of the detector, f_d is the doubles gate fraction, M is the self multiplication of the sample, ν_{Li2} is the second moment of induced fission for neutrons with an AmLi energy spectrum, ν_{Li1} is the first moment of induced fission for neutrons with an AmLi energy spectrum, ν_{Fis2} is the second moment of induced fission for neutrons with a fission energy spectrum, and ν_{Fis1} is the first moment of induced fission for neutrons with a fission energy spectrum. Equation 1 was used to determine the effective mass of ^{239}Pu. However, instead of using a coupling parameter to determine

the specific fission rate[12], F_0, a first principles approach was used:

$$F_o = \frac{\varphi_{Li}\sigma_{f\_Li}N_A}{M_{molar}}$$
(Eq. 2)

where ϕ_{Li}, is the neutron flux within the sample during an AmLi measurement, σ_{f_Li} is the average fission cross-section for neutrons with an AmLi energy spectrum, N_A is Avogadro's Number, and M_{molar} is the molar mass of the effective mass isotope.

By combining Eq. 1 with Eq. 2 and solving for $^{239}Pu_{eff\_Li}$ we acquired:

$$^{239}Pu_{eff\_Li} = \frac{2(D_{Li}-D_{Passive})M_{molar}}{\varphi_{Li}\sigma_{f\_Li}N_A\varepsilon^2 f_d M^2 \nu_{Li2}\left[1+\frac{(M-1)\nu_{Li1}\nu_{Fis2}}{(\nu_{Fis1}-1)\nu_{Li2}}\right]}$$
(Eq. 3)

Similar to passive coincidence counting, we used the following to convert from effective masses to the masses of isotopes of interest:

$$^{240}Pu_{eff} = 2.52\,^{238}Pu + {}^{240}Pu + 1.68\,^{242}Pu$$
(Eq. 4)

$$\begin{aligned} &^{239}Pu_{eff\_Li} \\ &= C_{U235\_Li}\,^{235}U + C_{U238\_Li}\,^{238}U + C_{Np237\_Li}\,^{237}Np \\ &+ C_{Pu238\_Li}\,^{238}Pu + C_{Pu239\_Li}\,^{239}Pu + C_{Pu240\_Li}\,^{240}Pu \\ &+ C_{Pu241\_Li}\,^{241}Pu + C_{Pu242\_Li}\,^{242}Pu + C_{Am241\_Li}\,^{241}Am \\ &+ C_{Am243\_Li}\,^{243}Am \end{aligned}$$
(Eq. 5)

where $^{240}Pu_{eff}$ is the effective mass of ^{240}Pu in the sample that would produce the same passive doubles count rate, ^{238}Pu is the mass of ^{238}Pu in the sample, ^{240}Pu is the mass of ^{240}Pu in the sample, ^{242}Pu is the mass of ^{242}Pu in the sample, C_{k_Li} is an equivalent worth constant for isotope k, ^{235}U is the mass of ^{235}U in the sample, ^{238}U is the mass of ^{238}U in the sample, ^{239}Pu is the mass of ^{239}Pu in the sample, ^{241}Pu is the mass of ^{241}Pu in the sample, ^{241}Am is the mass of ^{241}Am in the sample, ^{243}Am is the mass of ^{243}Am in the sample.

The constants, C_{k_Li}, were determined by using a ratio of nuclear properties of the effective isotope and the constant's isotope:

$$C_{k\_Li} = \frac{(\sigma_{f\_Li}\nu_{Li2})_k}{(\sigma_{f\_Li}\nu_{Li2})_{239}}\left(\frac{M_{molar,239}}{M_{molar,k}}\right)$$
(Eq. 6)

where k represents the isotope of interest.

The values of ν, σ_f, *and* f_d are constants for a given detector design and can be acquired through Monte Carlo Neutral Particle eXtended (MCNPX)[15] code simulations. ε can be determined from a ^{252}Cf measurement or through MCNPX simulations. ϕ is related to the active singles count rate. Table 1 shows values of various constants used in the above equations.

Table 1. Microscopic fission cross-sections, reduced factorial moments, and ^{239}Pu effective worth constants of both AmLi and AmB neutrons for ten isotopes of interest.

	σ_{f_Li}	σ_{f_B}	ν_{Li2}	ν_{Be2}	C_{k_Li}	C_{k_B}
^{235}U	1.67	1.40	3.87	4.30	0.658	0.510
^{238}U	0.00	0.23	4.21	4.84	0.001	0.092
^{237}Np	0.31	1.08	5.06	5.88	0.160	0.532
^{238}Pu	1.11	1.74	6.01	7.01	0.673	1.020
^{239}Pu	1.71	1.80	5.84	6.67	1.000	1.000
^{240}Pu	0.34	1.09	5.72	6.74	0.196	0.661
^{241}Pu	2.16	1.82	5.93	6.61	1.278	0.993
^{242}Pu	0.22	0.88	5.82	6.85	0.128	0.497
^{241}Am	0.17	1.08	7.39	8.61	0.127	0.772
^{243}Am	0.11	0.84	8.48	9.55	0.089	0.658

The passive singles, doubles, and triples neutron multiplicity equations[16] can be used with a passive measurement of a sample to determine α, M, and $^{240}Pu_{eff}$. To determine the Am content in a sample, the following equation was used:

$$\alpha = \frac{(13400f_{238}+38.1f_{239}+141f_{240}+1.3f_{241}+2.0f_{242})m_{Pu}+(2690f'_{241}+133f'_{243})m_{Am}}{1020(2.54f_{238}+f_{240}+1.69f_{242})m_{Pu}}$$
(Eq. 7)

where f_k is the Pu isotopic fraction for isotope k, f_k' is the Am isotopic fraction for isotope k, m_{Pu} is the mass of Pu in the sample, and m_{Am} is the mass of Am in the sample. The coefficient of 133 in front of the ^{243}Am isotopic fraction, f_{243}', was determined, through use of Oak Ridge Isotope GENeration-Automatic Rapid Process (ORIGEN-ARP) calculations[17], by relating the number of (α,n) neutrons produced in on oxide matrix by ^{243}Am to those produced by the Pu isotopes. Although Np and U do not appear in Eq. 7, the equation is still valid even if those elements exist in the sample. This is due to the fact that common Np and U isotopes have half-lives large enough that their (α,n) neutron production rates are small enough that they can be ignored. It should be noted that Eq. 7 assumes that there are not light element impurities in the oxide sample to produce additional (α,n) neutrons. Equation 7 can be used along with the isotopic fractions of Pu and Am, the Pu mass in the sample, and the measured α value to determine the Am mass in the sample.

In order to determine the U and Np masses in the sample, the AmLi and AmB versions of Eq. 3, Eq. 5, and the U and Np isotopic compositions were used. This measurement method results in a complete set of equations to determine the U, Np, Pu, and Am masses, assuming the isotopic composition of the sample is know.

VI. Results

Passive, active AmLi, and active AmB simulations were performed using MCNPX. These simulations were of the ENMC detector with five different oxide samples. 1) U, Np,

Pu, and Am mixture with spent fuel isotopic. This is representative of a possible UREX product. 2) Np, Pu, and Am mixture with 50% heavy metal content of depleted U. This mixture represents a potential intermediate product from a reprocessing plant to a fuel fabrication facility. 3) Np, Pu, and Am mixture with 50% heavy metal content of 4% enriched U. This mixture also represents a potential intermediate product from a reprocessing plant to a fuel fabrication facility. 4) Np, Pu, and Am mixture with 90% heavy metal content of 4% enriched U. This mixture represents a potential fuel material. 5) Pu and Am mixture with 90% heavy metal content of 4% enriched U. This mixture also represents a potential fuel material. It should be noted that the isotopic compositions not explicitly stated were taken from a typical Pressurized Water Reactor (PWR)[1].

When the values for $^{240}Pu_{eff}$, $^{239}Pu_{eff\_Li}$, and $^{239}Pu_{\mathrm{eff}\_Li}$ based off the measured singles, doubles, and triples rates are compared to theoretical values, a bias can be seen. This bias appears to be linearly proportional to the Pu mass in the sample, as seen in Fig. 5 for $^{240}Pu_{eff}$.

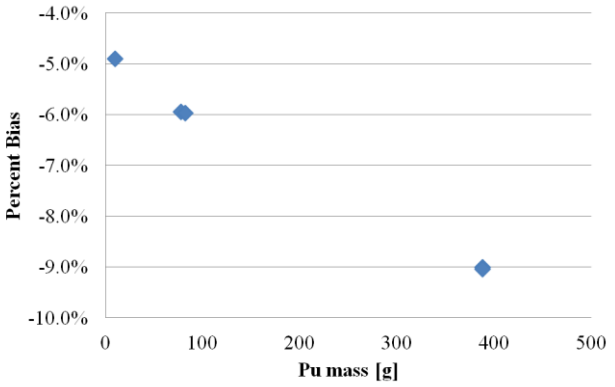

Figure 5. Pu mass vs. $^{240}Pu_{eff}$ bias, indicating a linear relationship.

Tables 2, 3, 4, and 5 show the measured and declared masses for Pu, Am, U, and Np, respectively. These tables also have the MCNPX statistical uncertainty and the predicted uncertainty from a 1 hour measurement, respectively[18].

Table 2. Measured and declared mass for Pu with MCNPX and measured uncertainties.

	Pu mass measured [g]			Pu mass declared [g]
1) U, Np, Pu, Am	9.40	±	0.00 , 0.04	9.89
2) Np, Pu, Am + 50%U @ 0.27%	353	±	0.08 , 1.13	388
3) Np, Pu, Am + 50%U @ 4.0%	353	±	0.08 , 1.13	388
4) Np, Pu, Am + 90%U @ 4.0%	73.07	±	0.01 , 0.23	77.69
5) Pu, Am + 90%U @ 4.0%	77.12	±	0.02 , 0.24	82.02

Table 3. Measured and declared mass for Am with MCNPX and measured uncertainties.

	Am mass measured [g]			Am mass declared [g]
1) U, Np, Pu, Am	0.71	±	0.00 , 0.01	0.41
2) Np, Pu, Am + 50%U @ 0.27%	33.75	±	0.02 , 0.22	16.22
3) Np, Pu, Am + 50%U @ 4.0%	33.82	±	0.02 , 0.22	16.22
4) Np, Pu, Am + 90%U @ 4.0%	5.82	±	0.00 , 0.04	3.24
5) Pu, Am + 90%U @ 4.0%	6.15	±	0.00 , 0.04	3.43

Table 4. Measured and declared mass for ^{235}U with MCNPX and measured uncertainties.

	^{235}U mass measured [g]			^{235}U mass declared [g]
1) U, Np, Pu, Am	6.34	±	17 , 33	7.57
2) Np, Pu, Am + 50%U @ 0.27%	-0.18	±	7 , 92	1.15
3) Np, Pu, Am + 50%U @ 4.0%	-80.90	±	513 , 6342	17.09
4) Np, Pu, Am + 90%U @ 4.0%	26.41	±	313 , 1575	30.76
5) Pu, Am + 90%U @ 4.0%	26.07	±	314 , 1637	30.76

Table 5. Measured and declared mass for ^{237}Np with MCNPX and measured uncertainties.

	^{237}Np mass measured [g]			^{237}Np mass declared [g]
1) U, Np, Pu, Am	1.70	±	349 , 669	0.57
2) Np, Pu, Am + 50%U @ 0.27%	89.73	±	494 , 6166	22.57
3) Np, Pu, Am + 50%U @ 4.0%	505	±	2626 , 32459	22.57
4) Np, Pu, Am + 90%U @ 4.0%	-4.63	±	1604 , 8061	4.51
5) Pu, Am + 90%U @ 4.0%	-7.64	±	1609 , 8377	0.00

From Tables 2 and 3 it can be seen that measuring the Pu and Am masses in an oxide sample with a 1 hour measurement produces expected measurement uncertainties which would be acceptable for most measurements. Tables 4 and 5 have expected measurement uncertainties for the ^{235}U and ^{237}Np masses which are unacceptably large.

VIII. CONCLUSIONS

The measurement methodology to determine the isotopic masses of a U, Np, Pu, and Am oxide sample has been developed. The measurement uncertainties associated with the masses of ^{235}U and ^{237}Np are too large for practical applications, using current neutron multiplicity technology. Optimizing the AmLi and AmB source strengths along with implementing the redesigned end plugs of the ENMC for an active configuration will reduce the measurement

uncertainties of ^{235}U and ^{237}Np. In addition to this, samples which do not contain Np do not require the additional active AmB measurement, thus reducing the number of measurements required and the measurement uncertainties of the ^{235}U mass in the sample.

The measurement methodology presented here might only be practical with fast neutron multiplicity counters, which have shorter die-away times, and thus fewer accidental counts[6].

The measurement methodology presented here requires the full isotopic fractions in the sample to be know. It is currently unknown if traditional gamma ray detectors with existing software programs[19] can achieve the required measurement uncertainties, or if more advanced techniques will be needed.

ACKNOWLEDGMENTS

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NOMENCLATURE

$^{239}Pu_{eff\_B}$	– Plutonium-239 effective mass for AmB active measurements
$^{239}Pu_{eff\_Li}$	– Plutonium-239 effective mass for AmLi active measurements
$^{240}Pu_{eff}$	– Plutonium-240 effective mass
Am	– Americium
AmB	– Americium-Boron
AmLi	– Americium-Lithium
B	– Boron
B_4C	– Boron-Carbide
DA	– Destructive Assay
ENMC	– Epithermal Neutron Multiplicity Counter
He	– Helium
IAEA	– International Atomic Energy Agency
INCC	– IAEA Neutron Coincidence Counting
M	– Self-Multiplication
MCNPX	– Monte Carlo N-Particle eXtended
NDA	– Nondestructive Assay
Np	– Neptunium
ORIGEN-ARP	– Oak Ridge Isotope GENeration-Automatic Rapid Process
Pu	– Plutonium
U	– Uranium
UREX	– URanium EXtraction
α	– Alpha-neutron reaction rate
ε	– Neutron detector efficiency

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