A Concept for Quantitative NDA MOX Measurements Using Only Neutron Radiation

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Abstract

There is a desire in the world to reduce the quantity of Pu in storage by burning it as mixed oxide (MOX) fuel in nuclear reactors. For safeguards reasons, the masses of both the ^{235}U and Pu in MOX fuel must be quantified, preferably using non-destructive assay (NDA) techniques. Current NDA techniques require both gamma ray and neutron measurements to be performed. This research investigates the ability to quantify the ^{235}U and Pu content of MOX fuel using only neutron measurements. This would reduce the amount of equipment needed and subsequently the cost required to conduct the measurement. This measurement technique is based on performing three different neutron measurements and analyzing their neutron multiplicity response. The first measurement is a passive measurement of a ^{252}Cf standard to determine the detector efficiency (ε). The second measurement, in a passive configuration, will determine the effective ^{240}Pu ($^{240}Pu_{eff}$) content, self multiplication (M), and alpha-neutron reaction rate (α). The third measurement, in an active configuration using interrogation neutrons from an AmLi and AmBe source, will determine the ^{239}Pu and ^{235}U content. Simulated and measured results will illustrate the viability of this technique and its practical limitations.

Introduction

The ability of inspection agencies and facility operators to measure MOX powders is increasingly necessary as Pu becomes more commonly used in commercial nuclear reactors, either from excess nuclear weapons material or from reprocessed used nuclear fuel. These MOX materials are difficult to measure because neutrons emitted from induced and spontaneous fission of different nuclides are very similar. Traditional NDA methods require additional gamma ray spectroscopy equipment to acquire the isotopic information of the measured material. Aside from the additional cost of this added equipment, only gamma rays from the outer most edge of the material can be detected due to the large photon absorption cross-section of high atomic number materials, such as U and Pu. These methods have the implicit assumption that the isotopic composition of the outside of the material is the same throughout the material. However, quantitative measurements of MOX materials are possible without using additional gamma ray equipment by exploiting isotope-specific nuclear properties, such as the energy-dependent fission cross sections and the neutron fission multiplicity.

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 reducing nuclear weapons material stockpiles and reprocessing used nuclear fuel. However, as reprocessing and weapons stockpile reduction becomes more popular, the production of MOX fuels is likely to increase.

Theory

The Active Well Coincidence Counter (AWCC), shown in Fig. 1, is a neutron multiplicity counter which can detect neutrons created individually or in multiples. 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 AWCC 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 AWCC detects neutrons through neutron capture via 42 ^{3}He tubes embedded in polyethylene (which is used for moderating, or slowing down, neutrons to increase detection probability).

Figure 1. An AWCC located at the Joint Research Center in Ispra, Italy. Left: Braden Goddard, Center: Chris Ryan, Right: Dr. Paolo Peerani.

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, or thermalize, 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 64 μs, 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 multiplication of the sample (M) and is accounted for when determining the Pu mass of the sample.

The AWCC can be operated in two different modes, 1) a passive mode in which it passively detects spontaneous fission neutrons from the sample and 2) an active mode in which fission is induced in the sample by external neutron sources. 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 on the composition of the samples but merely an equivalent mass.

When determining the mass of a sample which contains only one actinide, there are four unknown variables that must be solved for, 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, an independent equation must be created. These independent equations can be created through measurements of known standards, computer simulations, or singles, doubles, or triples count rates from the sample being measured.

For samples containing multiple isotopes of interest, an additional unknown effective mass is added for each additional isotope contained in the sample. Because of this, MOX samples require more independent equations. To create these independent equations, a passive and an active measurement, as well as an additional active measurement containing AmBe interrogation sources, 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 AWCC, the masses of ^{235}U, ^{238}U, $^{290}Pu_{eff}$, and $^{240}Pu_{eff}$ can be quantified.

Active measurements made with either the traditionally-used AmLi source or an AmBe 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. This average energy and distribution are quite different from an AmBe source which has an average energy of 5 MeV and an energy distribution as seen in Fig. 3.[1] 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 5 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.[2]

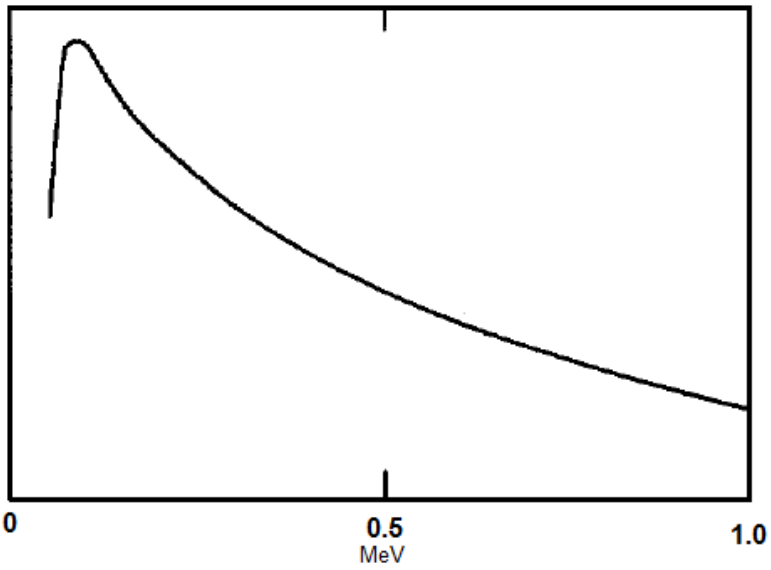

Figure 2. Neutron energy spectrum from an AmLi source.[1]

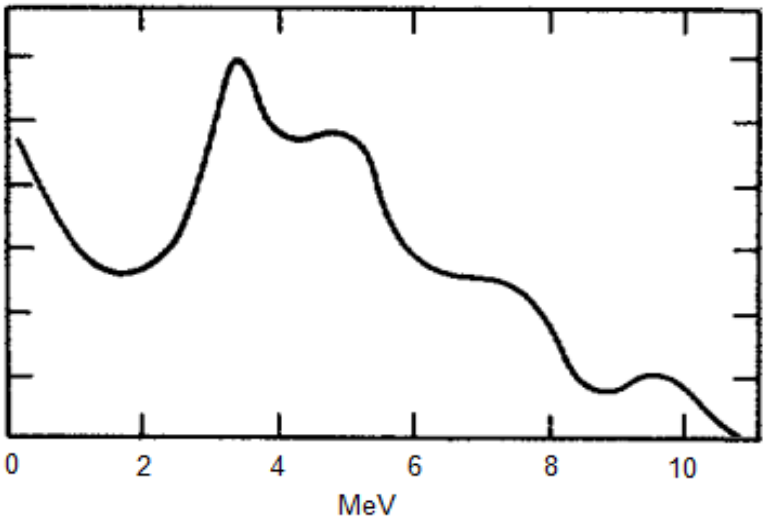

Figure 3. Neutron energy spectrum from an AmBe source.[1]

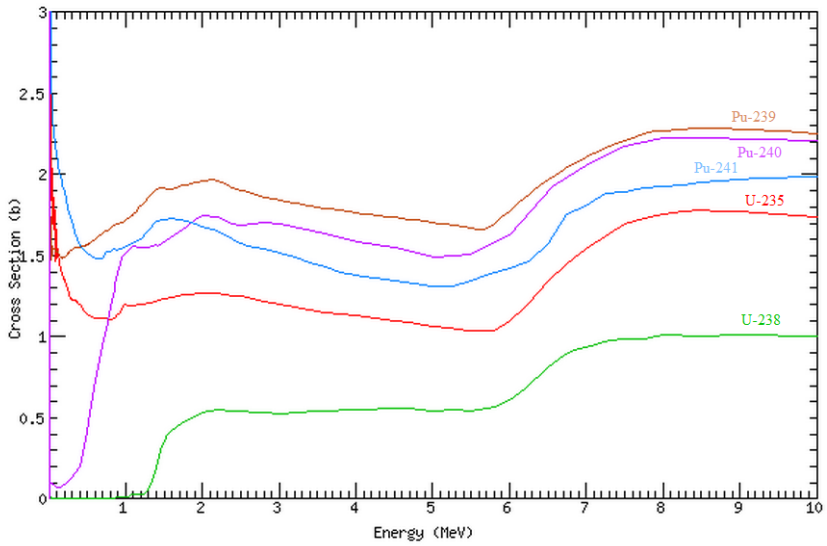

Figure 4. Fission cross-section for common isotopes of U and Pu.[2]

Boron-Carbide

Due to the design of the AWCC, a large 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 flux within a U sample using an active AmLi and AmBe source. These flux distributions are not only similar, thus reducing the independence of each other, but cause the fission rate in the sample to be predominantly on the periphery of the sample due to U and Pu epithermal resonances. To solve this problem, the measurement samples were placed inside of a boron-carbide (B_4C) cylinder. Figure 6 shows the energy dependent flux within a U sample, which has been placed in a B_4C cylinder, using an active AmLi and AmBe source. The B_4C used consisted of a boron enrichment of 96% in ^{10}B, a density of 90% theoretical, and a container thickness of 2 cm on the side, top and, bottom.[3] The outside height and diameter of the B_4C container were 16 cm and 14 cm, respectively.

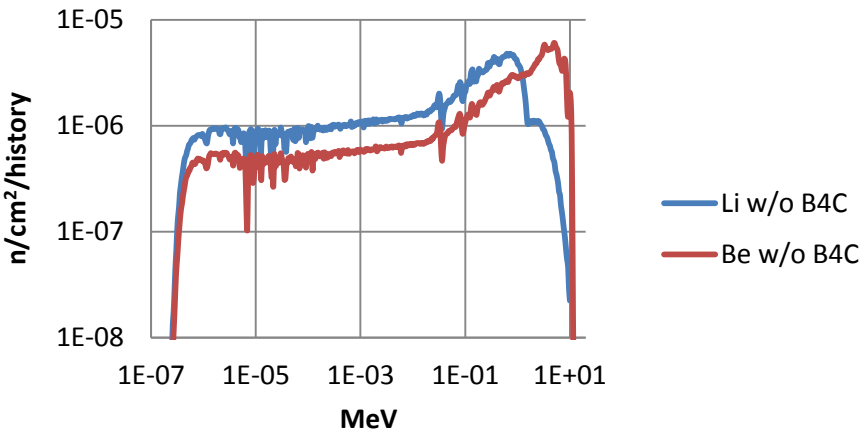

Figure 5. Energy dependent flux within a U sample, without B_4C shielding, using an active AmLi and AmBe source.

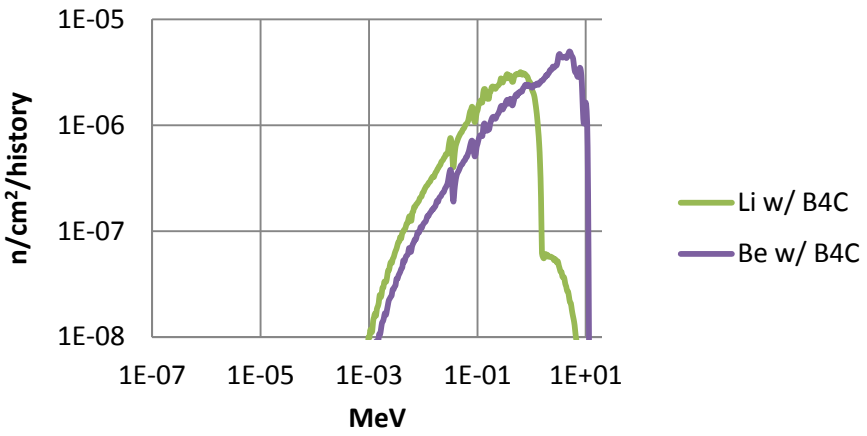

Figure 6. Energy dependent flux within a U sample, with B_4C shielding, using an active AmLi and AmBe source.

Calculations of Effective Mass

The active neutron coincidence counting doubles equation is given by[4]:

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

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 an isotope of interest. However, instead of using a coupling parameter to determine the specific fission rate, F_0, a first principles approach was used:

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

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:

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

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

$$^{240}Pu_{eff} = 2.52^{238}Pu + {}^{240}Pu + 1.68^{242}Pu \quad \text{Eq. 4}$$

$$^{239}Pu_{eff\_Li} = C_{U235\_Li}{}^{235}U + C_{U238\_Li}{}^{238}U + C_{Pu239\_Li}{}^{239}Pu + C_{Pu240\_Li}{}^{240}Pu \quad \text{Eq. 5}$$

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, and ^{240}Pu is the mass of ^{240}Pu in the sample.

It should be noted that Eq. 5 assumes that there are only four fissionable isotopes in the measured sample, ^{235}U, ^{238}U, ^{239}Pu, and ^{240}Pu. This assumption is valid when the isotopic fractions of ^{238}Pu and ^{242}Pu are small, and due to similar C_{k_Li} values for ^{239}Pu and ^{241}Pu, Shown in able 1.

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

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

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)[6] 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 AmBe neutrons for four isotopes of interest.

	σ_{f_Li} [b]	σ_{f_Be} [b]	ν_{Li2}	ν_{Be2}	C_{k_Li}	C_{k_Be}
^{235}U	1.44	1.31	3.87	5.20	0.603	0.499
^{238}U	0.00	0.35	4.11	6.53	0.002	0.164
^{239}Pu	1.62	1.78	5.82	7.80	1.000	1.000
^{240}Pu	0.43	1.24	5.73	8.18	0.260	0.728
^{241}Pu						

The passive singles, doubles, and triples neutron multiplicity equations[1] can be used with a passive measurement of a MOX sample to determine α, M, and $^{240}Pu_{eff}$. A passive ^{252}Cf measurement can be made to determine ε. By using the declared enrichment of the MOX uranium with the AmLi and AmBe versions of Eq. 3 and Eq. 5, the ^{235}U, ^{238}U, and ^{239}Pu masses can be determined. Thus resulting in a complete set of equations to determine the ^{235}U, ^{238}U, ^{239}Pu, and ^{240}Pu$_{eff}$ masses without using a gamma spectroscopy measurement.

Verification of Uranium Enrichment Using α

When performing safeguards measurements, it is necessary to verify any values used by the inspector which were provided by the facility operator. For MOX measurements made using the above mentioned methods, the uranium enrichment must be verified. This can be done indirectly by

comparing the α value determined by the passive measurement to the $^{240}Pu_{eff}/^{239}Pu$ ratio. By knowing the burn-up history of the Pu, a $^{240}Pu_{eff}/^{239}Pu$ ratio versus α plot can be created, as seen in Fig. 7 for a Pressurized Water Reactor (PWR). If the declared U enrichment is correct, the measured α value will match the $^{240}Pu_{eff}/^{239}Pu$ plot value, within statistical uncertainty. It should be noted that the α values were determined using:

$$\alpha = \frac{13400 f_{238} + 38.1 f_{239} + 141 f_{240} + 1.3 f_{241} + 2.0 f_{242}}{1020(2.54 f_{238} + f_{240} + 1.69 f_{242})} \qquad \text{Eq. 7}$$

where f_k is the Pu isotopic fraction for isotope k.[1] The data in Fig. 7 was obtained using the Oak Ridge Isotope Generation (ORIGEN) code.[7]

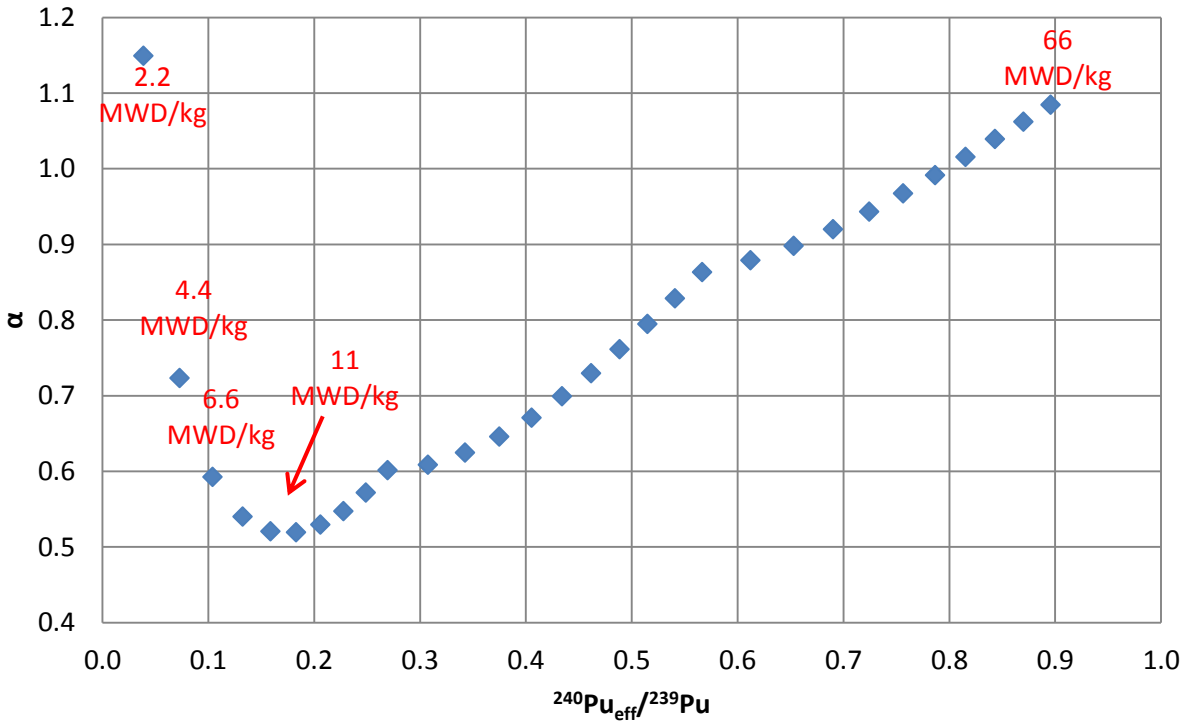

Figure 7. Plot of α versus $^{240}Pu_{eff}/^{239}Pu$ ratio for a PWR.

Results

Passive, active AmLi, and active AmBe measurements were performed on U, Pu, and MOX samples of varying isotopic and chemical compositions to benchmark an MCNPX model for an AWCC. Tables 2, 3, and 4 show the percent difference, with statistical uncertainty, between the measured and declared effective masses for: 1) $^{239}Pu_{eff\_Li}$, $^{239}Pu_{eff\_Be}$, and $^{240}Pu_{eff}$ for several different isotopic compositions; 2) $^{235}U_{eff\_Li}$ and $^{235}U_{eff\_Be}$ for several different enrichments; and 3) ^{235}U, ^{238}U, ^{239}Pu, and $^{240}Pu_{eff}$ for two different MOX compositions. The MOX compositions contain 170g of heavy metal in an oxide composition, with a Pu fraction of 6%, and a uranium enrichment of 0.256%. The MOX sample with a Pu isotopic of: 0% ^{238}Pu, 95% ^{239}Pu, 5% ^{240}Pu, 0% ^{241}Pu, and 0% ^{242}Pu (0,95,5,0,0) contains weapons grade Pu,[8] while the MOX (2,55,22,15,6) sample contains reactor grade Pu.[7] The U, Pu, and MOX results shown in tables 2, 3 and,4 were acquired from MCNPX simulations.

Table 2. Percent difference, with statistical uncertainty, between the measured and declared effective masses for $^{239}Pu_{eff\_Li}$, $^{239}Pu_{eff\_Be}$, and $^{240}Pu_{eff}$ for several different isotopic compositions.

Material Composition	$^{239}Pu_{eff\_Li}$ [%]	$^{239}Pu_{eff\_Be}$ [%]	$^{240}Pu_{eff}$ [%]
Pu (0,70,30,0,0)	-11.8 ± 2.1	-4.6 ± 4.6	2.1 ± 0.01
Pu (0,55,30,15,0)	-7.0 ±2.2	-5.3 ± 2.5	2.0 ± 0.01
Pu (0,70,24,0,6)	-11.9 ± 2.2	-5.1 ± 2.5	2.3 ± 0.01
Pu (2,55,22,15,6)	-7.6 ± 2.1	-3.4 ± 4.7	4.7 ± 0.01
Pu (0,100,0,0,0)	-6.8 ± 1.9	-2.5 ± 2.3	-
Pu (0,0,100,0,0)	-3.3 ± 4.7	-4.1 ± 5.8	0.6 ± 0.01
MOX (0,95,5,0,0)	-2.4 ± 12.0	6.3 ± 11.3	3.9 ± 0.01
MOX (2,55,22,15,6)	-4.2 ± 13.9	4.9 ± 11.5	12.2 ± 0.01

Table 3. Percent difference, with statistical uncertainty, between the measured and declared effective masses for $^{235}U_{eff\_Li}$ and $^{235}U_{eff\_Be}$ for several different enrichments.

Material Composition	$^{235}U_{eff\_Li}$ [%]	$^{235}U_{eff\_Be}$ [%]
U 36% (metal)	-3.9 ± 3.9	1.7 ± 9.2
U 90%	-0.1 ± 4.4	20.9 ± 15.8
U 36%	-2.2 ± 3.4	3.3 ± 7.7
U 20%	5.5 ± 6.0	9.4 ± 9.8
U 0.3%	90.4 ± 370.6	13.1 ± 14.3

Table 4. Percent difference, with statistical uncertainty, between the measured and declared masses for ^{235}U, ^{238}U, ^{239}Pu, and $^{240}Pu_{eff}$ for two different MOX compositions.

Material Composition	^{235}U [%]	^{238}U [%]	$^{239}Pu^{(a)}$ [%]	$^{240}Pu_{eff}$ [%]
MOX (0,95,5,0,0)	8.1 ± 15.5	8.1 ± 15.5	0.4 ± 13.0	3.9 ± 0.01
MOX (2,55,22,15,6)	3.4 ± 15.6	3.4 ± 15.6	-4.4 ± 16.6	12.2 ± 0.01

[(a)]Includes 241Pu equivalent mass

From Tables 2, 3, and 4 it can be seen that the effective and actual masses of the different isotopes are mostly accurate given their statistical uncertainty. This is especially true for the two MOX simulations, which are the most practical application of this measurement method.

Conclusion

The initial assessment of the feasibility to perform quantitative NDA measurements on MOX fuel without using gamma spectroscopy has been completed and shown to be a potentially useful NDA technique. Before this technique can be implemented in a non-research setting further work may be needed. The AmBe neutron source should be replaced with a non-beryllium material due to the large (n,2n) cross-section of beryllium. Implementing this change would significantly reduce the uncertainty of high neutron energy active measurements. A more appropriate detector needs to be chosen. A large portion of the interrogation flux in the AWCC is in the epithermal range and is thus filtered out by the B_4C. This filtering leads to a lower fission rate and thus longer counting times. A more thorough investigation of the expected α values from different reactor designs with varying burn-up is needed. A

wider range of samples need to be simulated in addition to performing physical measurements. The optimum thickness and shape of the B_4C container needs to be determined. The significance to safeguards of knowing the content of ^{239}Pu and $^{240}Pu_{eff}$ without knowing the full Pu isotopic composition needs further investigation. One of the most useful aspects of this research is the methodology that was used, which can be applied to other NDA challenges.

Acknowledgements

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