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An Analysis Technique for Active Neutron Multiplicity Measurements Based on First Principles

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Abstract

Passive neutron multiplicity counting is commonly used to quantify the total mass of plutonium in a sample, without prior knowledge of the sample geometry. However, passive neutron counting is less applicable to uranium measurements due to the low spontaneous fission rates of uranium. Active neutron multiplicity measurements are therefore used to determine the ^{235}U mass in a sample. Unfortunately, there are still additional challenges to overcome for uranium measurements, such as the coupling of the active source and the uranium sample. Techniques, such as the coupling method, have been developed to help reduce the dependence of calibration curves for active measurements on uranium samples; although, they still require similar geometry known standards. An advanced active neutron multiplicity measurement method is being developed by Texas A&M University, in collaboration with Los Alamos National Laboratory (LANL) in an attempt to overcome the calibration curve requirements. This method can be used to quantify the ^{235}U mass in a sample containing uranium without using calibration curves. Furthermore, this method is based on existing detectors and non-destructive assay (NDA) systems, such as the LANL Epithermal Neutron Multiplicity Counter (ENMC). This method uses an inexpensive boron carbide liner to shield the uranium sample from thermal and epithermal neutrons while allowing fast neutrons to reach the sample. Due to the relatively low and constant fission and absorption energy dependent cross-sections at high neutron energies for uranium isotopes, fast neutrons can penetrate the sample without significant attenuation. Fast neutron interrogation therefore creates a homogeneous fission rate in the sample, allowing for first principle methods to be used to determine the ^{235}U mass in the sample. This paper discusses the measurement method concept and development, including measurements and simulations performed to date, as well as the potential limitations.

Introduction

The ability of inspection agencies and facility operators to measure the ^{235}U content of mixed oxide (MOX) powders is increasingly necessary as plutonium becomes more commonly used in commercial nuclear reactors, either from excess nuclear weapons material or from reprocessed spent nuclear fuel. These MOX materials are difficult to measure because the spectrum and multiplicity distributions of neutrons emitted from induced and spontaneous fission of different nuclides are similar. Traditional NDA methods to quantify total ^{235}U mass include active neutron coincidence counting. Until recently, active neutron coincidence counting required calibration curves of known standards with similar geometry and material properties. Some work has been done to make this method less dependent on known standards; however, calibration curves are still required. Measurement techniques which use first principle methods and little to no calibration curves are desirable to improve the range of samples which can be measured. Preventing thermal and epithermal neutrons from entering the measured sample will create a homogeneous fission rate within the sample. This homogeneous fission rate will reduce variations in the samples spatially dependent properties, such as the microscopic fission cross-section. Using sample averaged values allows for first principle methods to be applied to the sample to determine total ^{235}U content.

Theory

Several neutron multiplicity counters can be operated in two different modes: 1) A passive mode in which spontaneous fission neutrons are detected from the sample and 2) an active mode in which fission is induced in the sample by external neutron interrogation sources e.g. americium lithium (AmLi), positioned above and below the sample. When analyzing the resultant count rates, it is common to calculate a ^{240}Pu-effective mass ($^{240}Pu_{eff}$) for passive measurements and a ^{235}U or ^{239}Pu-effective mass ($^{239}Pu_{eff}$) for active measurements. Effective mass refers to the equivalent amount of material of that isotope that would be required to produce the same doubles response.

For plutonium sample measurements there are four unknown variables to calculate: 1) Detection efficiency (ε), 2) the sample self-leakage multiplication (M), 3) the ratio of the alpha-induced to the spontaneous fission neutron production rates of the sample (α), and 4) the effective mass of the sample, $^{240}Pu_{eff}$. For each of these unknown variables, an independent equation must be created. These independent equations can be created through measurements of known standards (which is how ε is commonly determined), from computer simulations, or measured singles, doubles, or triples count rates. Active measurement of uranium can be more difficult since only the doubles count rate usually provides statistically relevant data. ε can be determined from measured standards and α is not applicable due to the nature of active measurements. This leaves M to be solved using either known standards or computer simulations, both of which require significant prior knowledge about the sample characteristics. For MOX measurements, a combination of passive and active measurements may be used to determine M, α, $^{240}Pu_{eff}$, and $^{239}Pu_{eff}$. ε is determined via measurements of known standards.

AmLi interrogation source neutrons have an average energy of 0.3 MeV. The neutron energy spectrum for AmLi is shown in Figure 1[1]. By preventing thermal and epithermal neutrons from entering the

sample, described in more detail in the next section, not only does the fission rate in the sample become homogeneous but the sample averaged fission cross-sections become less sensitive to slight changes in energy. Figure 2 shows the induced fission cross-section for several common actinides[2]. This interrogation spectrum modification was achieved by using a boron carbide liner inside the detector.

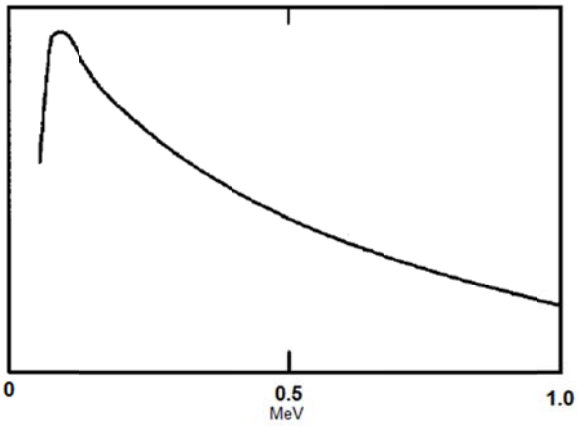

Figure 1. Neutron energy spectrum from an AmLi source. Linear scale[1].

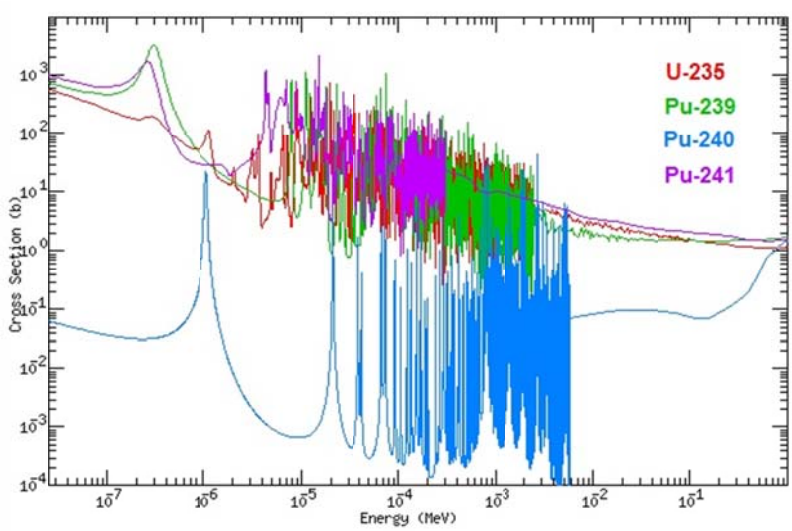

Figure 2. Fission cross-section for common isotopes of uranium and plutonium. Logarithmic scale[2].

The International Neutron Coincidence Counting (INCC) software program, Version 5.1 [3] was used when performing the measurements to obtain the neutron counting rates. The Monte Carlo N-Particle eXtended (MCNPX)[4] code was used for the detector simulations which produces the same output. The outputs of both these software programs were analyzed identically using neutron multiplicity methods[5] and several values of interest were calculated, including: $^{240}Pu_{eff}$, $^{239}Pu_{eff}$, M, and α via the following equations[1][6][7][8]:

$$0 = a + bM + cM^2 + M^3$$

$$a = \frac{-6T\nu_{s2}(\nu_{Fis1} - 1)}{\varepsilon^2 f_t S(\nu_{s2}\nu_{Fis3} - \nu_{s3}\nu_{Fis2})} \qquad \text{Eq. 1}$$

$$b = \frac{2D_{Passive}[\nu_{s3}(\nu_{Fis1} - 1) - 3\nu_{s2}\nu_{Fis2}]}{\varepsilon f_d S(\nu_{s2}\nu_{Fis3} - \nu_{s3}\nu_{Fis2})}$$

$$c = \frac{6D_{Passive}\nu_{s2}\nu_{Fis2}}{\varepsilon f_d S(\nu_{s2}\nu_{Fis3} - \nu_{s3}\nu_{Fis2})} - 1$$

$$^{240}Pu_{eff} = \frac{\frac{2D_{Passive}}{\varepsilon f_d} - \frac{M(M-1)\nu_{Fis2}S}{\nu_{Fis1} - 1}}{F_0 \varepsilon M^2 \nu_{s2}} \quad \text{Eq. 2}$$

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

$$\alpha = \frac{S}{^{240}Pu_{eff}F_0\varepsilon\nu_{s1}M} - 1 \quad \text{Eq. 4}$$

where S is the passive singles count rate, $D_{Passive}$ is the passive doubles count rate, D_{Li} is the active doubles count rate, T is the passive triples count rate, ε is the neutron detection efficiency, f_d is the doubles gate fraction, f_t is the triples gate fraction, ν_{Li1} is the first moment of induced fission for neutrons with an AmLi energy spectrum, ν_{Li2} is the second moment of induced fission for neutrons with an AmLi energy spectrum, ν_{Fis1} is the first moment of induced fission for neutrons with a fission energy spectrum, ν_{Fis2} is the second moment of induced fission for neutrons with a fission energy spectrum, ν_{Fis3} is the third moment of induced fission for neutrons with a fission energy spectrum, ν_{s1} is the first moment of spontaneous fission, ν_{s2} is the second moment of spontaneous fission, ν_{s3} is the third moment of spontaneous fission, F_0 is the specific spontaneous fission rate for ^{240}Pu, ϕ_{Li} is the neutron flux within the sample during an AmLi measurement, σ_{fLi} 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 ^{239}Pu.

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

$$^{240}Pu_{eff} = 2.52\,^{238}Pu + {}^{240}Pu + 1.68\,^{242}Pu \quad \text{Eq. 5}$$

$$^{239}Pu_{eff} = C_{U235}\,{}^{235}U + C_{U238}\,{}^{238}U + C_{Pu238}\,{}^{238}Pu + C_{Pu239}\,{}^{239}Pu + C_{Pu240}\,{}^{240}Pu + C_{Pu241}\,{}^{241}Pu + C_{Pu242}\,{}^{242}Pu \quad \text{Eq. 6}$$

where ^{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 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 ^{241}Pu is the mass of ^{241}Pu in the sample.

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

$$C_k = \frac{\left(\sigma_{fLi}\nu_{Li2}\right)_k}{\left(\sigma_{fLi}\nu_{Li2}\right)_{239}}\left(\frac{M_{molar,239}}{M_{molar,k}}\right) \qquad \text{Eq. 7}$$

where k represents the isotope of interest.

The values of ν_{Li}, σ_{fLi}, and f_d are constants for a given detector design and can be acquired through MCNPX simulations. ε can be determined from a ^{252}Cf measurement or MCNPX simulations. ϕ_{Li} is related to the active singles count rate and *M*. Table 1 shows values of the constants used in Eq. 7.

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

	σ_{fLi} [b]	ν_{Li2}	C_k
^{235}U	1.83	4.90	0.715
^{238}U	0.00	5.38	0.001
^{238}Pu	1.12	7.29	0.642
^{239}Pu	1.78	7.17	1.000
^{240}Pu	0.37	7.09	0.202
^{241}Pu	2.36	7.17	1.1315
^{242}Pu	0.25	7.09	0.134

The passive singles, doubles, and triples neutron multiplicity equations 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 above values with Eq. 3, 4, and 5, the ^{235}U mass can be determined without knowing the uranium heavy metal fraction or isotopic composition, assuming that the ^{238}U fission contribution is negligible. From Table 1, we can see that C_{U238} is 0.001, which is much smaller than the value of C_{U235}, which is 0.715.

Boron-Carbide

Due to the design of many active neutron coincidence counters, a large portion of the neutrons entering the measurement sample are epithermal. This is due to scattering interactions within the polyethylene. Figure 3 shows the energy dependent fission rate, with and without B_4C, within a 100g U_3O_8 sample using an active AmLi source. A large number of epithermal fissions in the sample occur predominately on the exterior of the sample, thus creating a heterogeneous fission rate within the sample. This not only violates the point model assumption but causes ϕ_{Li} and σ_{fLi} to be geometry dependent. To solve this problem, the measurement samples can be placed inside a B_4C can. The B_4C used here was a 2.5cm thick natural isotopic composition B_4C can with an outside height and diameter of 22cm and 18cm, respectively.

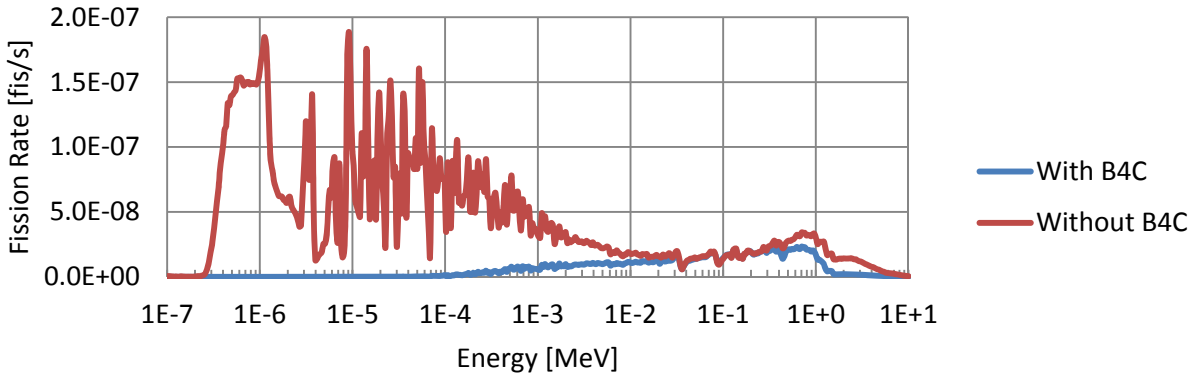

Figure 3. Energy dependent fission rate of a uranium sample using an active AmLi source.

Experiment

All neutron measurements described in this paper were performed at LANL using the Epithermal Neutron Multiplicity Counter (ENMC)[9]. The ENMC has a higher efficiency and shorter die-away time than other commercially available neutron counting systems. The ENMC has a 20cm diameter cylindrical sample cavity surrounded by 121 ^{3}He tubes at 10 atm embedded in high density polyethylene. The ^{252}Cf efficiency of the ENMC in an active configuration is 59% with a die-away time of 19µs. The output of this detector was sent to a JSR-15 multiplicity shift register module[10] and analyzed with INCC.

Passive and active AmLi measurements were performed on six different plutonium standards of varying isotopic and chemical compositions to benchmark an MCNPX model of the ENMC. Table 2 shows the total plutonium mass, ^{240}Pu fraction to total plutonium mass, and chemical composition of each standard.

Table 2. Total plutonium mass, ^{240}Pu fraction to total plutonium mass, and chemical composition of each standard.

Sample Name	Total Pu Mass [g]	^{240}Pu Fraction	Composition
STDIS03	10.96	0.0356	PuO_2 powder
STDIS09	11.85	0.0689	PuO_2 powder
STDIS12	20.09	0.1185	PuO_2 powder
CBNM-Pu61	5.45	0.2669	PuO_2 ceramic
STD11	59.74	0.0662	PuO_2 powder with F impurities
LAO250C10	59.36	0.1630	PuO_2 powder

All measurements, passive and active, were performed with the ENMC in an active configuration with the B_4C insert. The measurement times were all 3600s long, with the exception of a 12800s AmLi background measurement. The plutonium standards were placed on a stand in approximately the center of the B_4C insert. The B_4C insert was also placed on a stand in approximately the center of the ENMC cavity. The AmLi interrogation sources had neutron emission rates of approximately 4.1×10^4 ±

20% n/s each. The JSR-15 was set with a pre-delay of 1.5µs, a gate width of 2 µs, and a cycle length of 60s.

Results and Analysis

The values of $^{240}Pu_{eff}$, α, and M, were all in reasonable agreement for the measurements and MCNPX simulations, seen in Figures 4-6. As expected, the measured α value for the STD11 plutonium standard is significantly larger than the declared or MCNPX values. This is because the fluorine impurities were not accounted for in the MCNPX or declared values. There is a large discrepancy in the value of M for the same STD11 standard, which is thought to come from the large α value. It is not known why the STDIS12 standard has a significantly larger measured M value than the MCNPX value.

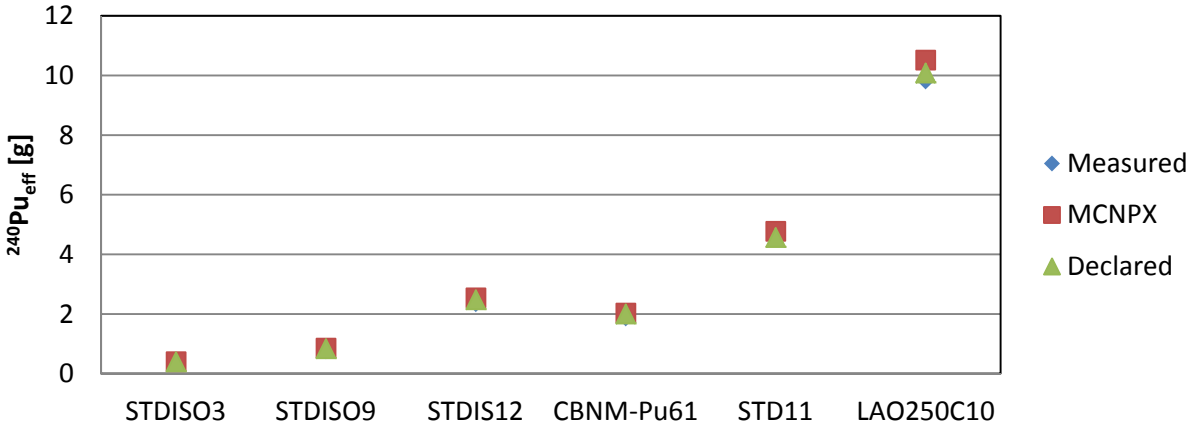

Figure 4. $^{240}Pu_{eff}$ values from measured and MCNPX simulated counts rates as well as the declared values. Error bars smaller than data points.

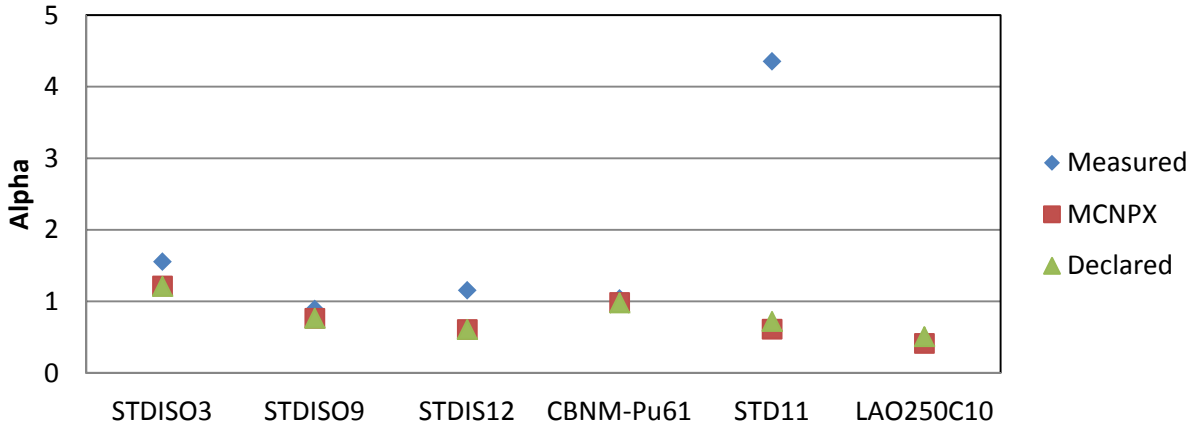

Figure 5. Alpha values from measured and MCNPX simulated counts rates as well as the declared values. Error bars smaller than data points.

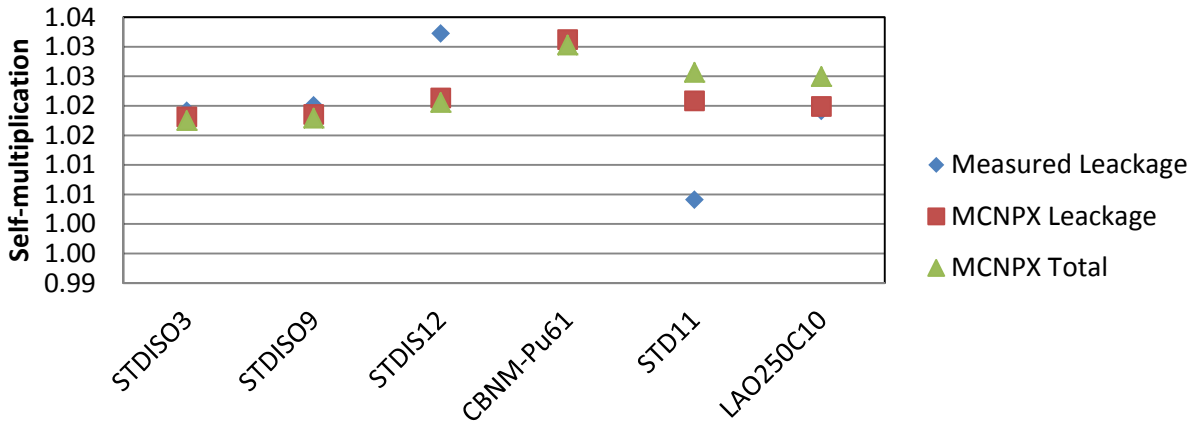

Figure 6. Self-multiplication values from measured and MCNPX simulated counts rates as well as the declared values.

The measured and MCNPX simulated $^{239}Pu_{eff}$ values calculated from Eq. 3, along with the declared values from Eq. 6, can be seen in Figure 7. It is clear that the MCNPX simulated values match well with the declared, although it is difficult to draw conclusions about the measured data. The measured $^{239}Pu_{eff}$ values have much larger uncertainties than those of other neutron multiplicity values. This is due to the fact that the doubles count rates from passive and active measurements vary from -0.8% to 1.9%. In order to reduce this uncertainty, either the difference between the doubles count rates, or the precision to which they are known, needs to be increased.

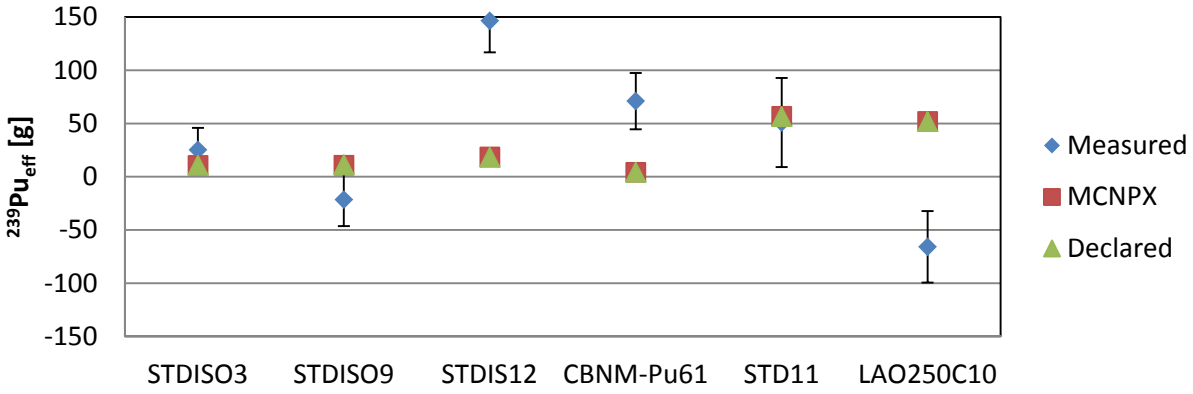

Figure 7. $^{239}Pu_{eff}$ values from measured and MCNPX simulated counts rates as well as the declared values.

Although many of the $^{239}Pu_{eff}$ values in Figure 7 appear to be statistically different, they do not incorporate all sources of uncertainty. Unrelated measurements using a low burn-up plutonium boron (PuB) interrogation source have shown that systematic uncertainties contribute to variations of measured active doubles count rates. These measurements were performed with the same plutonium standards and detector setup, but had much higher energy interrogation neutrons, approximately 2.8 MeV. Figure 8 shows the ratio of the doubles count rates for a 600s and a 3600s measurement. It should be noted that these measurements were nonconsecutive and the ENMC end plugs and B_4C insert

were removed between each measurement. It can be seen that 3 out of the 5 data points have ratios statistically different than 1.

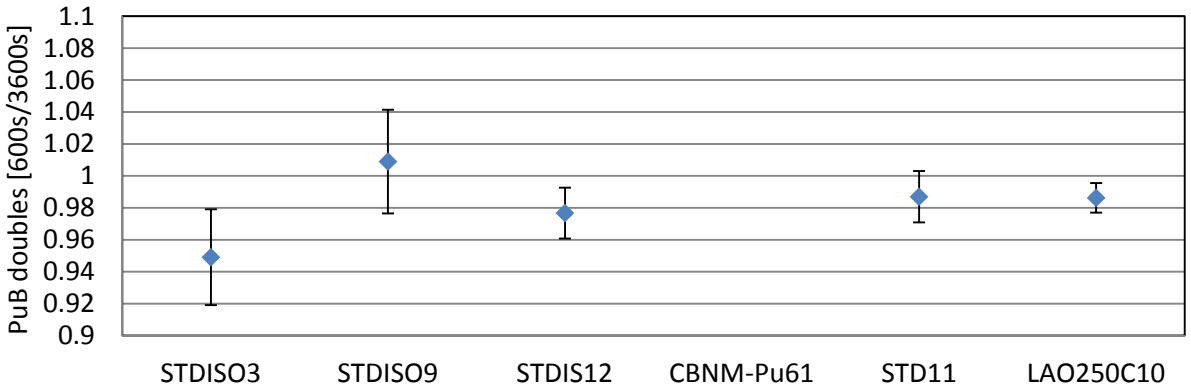

Figure 8. Ratio of the doubles count rates for a 600s and a 3600s measurement of the ENMC with the B_4C insert in an active configuration using a PuB interrogation source. The CBNM-Pu61 standard was not measured, and thus not shown.

Even if systematic uncertainties could be minimized, the measured $^{239}Pu_{eff}$ values in Figure 7 still have large counting statistical uncertainties. An alternative to performing longer measurement times would be to increase the neutron emission rate of the AmLi sources. MCNPX simulations were performed for each measured plutonium standard to estimate how the counting statistical uncertainties would change with variable neutron emission rates. From Table 3, it can be seen that for the two low burn-up low plutonium mass STDIS03 and STDIS09 standards there are only marginal gains by using AmLi sources with neutron emission rates above $6x10^4$ n/s each. However, for the higher burn-up larger plutonium mass standards there is a significant reduction in the uncertainty estimates.

Table 3. MCNPX simulations estimating counting statistical uncertainties for various AmLi neutron emission rates.

AmLi strength [n/s each]	$5x10^3$	$1x10^4$	$6x10^4$	$1x10^5$	$5x10^5$	$1x10^6$	$1x10^7$
STDIS03	639%	373%	211%	205%	198%	198%	197%
STDIS09	952%	516%	222%	210%	196%	194%	193%
STDIS12	1274%	652%	169%	145%	117%	115%	113%
CBNM-Pu61	4445%	2311%	690%	610%	517%	507%	498%
STD11	476%	241%	49%	38%	25%	24%	23%
LAO250C10	634%	323%	65%	50%	29%	27%	25%

Conclusion

The theory and analytical methods for measuring the $^{239}Pu_{eff}$ mass of a sample have been developed and shown to agree with MCNPX predictions. It is less clear that the measurements also statistically agree with the theory. Three potential solutions to improving the results of the measurements are: 1) Increase the count time of the active measurements, 2) Reduce systematic uncertainties by reducing

variations in sample and B_4C geometric positioning, and 3) increase the AmLi source strength for high burn-up large mass plutonium samples.

This method for determining a sample $^{239}Pu_{eff}$ mass is expected to work for measurements of plutonium, uranium, and MOX, although the concept has only been tested with plutonium samples. Measurements of uranium standards present the additional challenge of determining the sample self-leakage multiplication without measurement of the sample passive spontaneous fission. Although, it is suspected that the $^{239}Pu_{eff}$ uncertainties will be much smaller without subtracting the large spontaneous fission rate which occurs in plutonium bearing samples. Additional work needs to be performed before an implementation of this method can be evaluated for plutonium, uranium, or MOX facilities.

Acknowledgements

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References

1. D. Reilly, N. Ensslin, and H. Smith, "Passive Nondestructive Assay of Nuclear Materials," Ch. 16, Los Alamos National Laboratory, LA-UR-90-732, March (1991)
2. J. Chang, ENDFPLOT-2.0, Korea Atomic Energy Research Institute, accessed July 13, 2011, http://atom.kaeri.re.kr/endfplot.shtml
3. "International Neutron Coincidence Counting 5.1.2.3," Los Alamos National Laboratory, LA-CC-10-092, September (2010)
4. J. Hendricks, "MCNPX 2.6.0 Extensions," Los Alamos National Laboratory, LA-UR-08-2216, April 2008.
5. K. Boehnel, "The Effect of Multiplication on the Quantitative Determination of Spontaneously Fissioning Isotopes by Neutron Correlation Analysis," Nuclear Science and Engineering, 90, pp. 75-82 (1985)
6. D. Reilly, "Passive Nondestructive Assay of Nuclear Materials Addendum," Ch. 6, Los Alamos National Laboratory, LA-UR-07-1402, (2007)
7. D. Reilly, "Passive Nondestructive Assay of Nuclear Materials Addendum," Ch. 7, Los Alamos National Laboratory, LA-UR-07-1403, (2007)
8. B. Harker, M. Krick, W. Geist, and J. Longo, "INCC Software Users Manual," Los Alamos National Laboratory, LA-UR-10-6227, March (2009)
9. J. Stewart, H. Menlove, D. Mayo, W. Geist, L. Carrillo, G. Herrera, "The Epithermal Neutron Multiplicity Counter Design and Performance Manual: More Rapid Plutonium and Uranium Inventory Verifications by Factors of 5-20," Los Alamos National Laboratory, LA-13743-M, August (2000)
10. N. Menaa, M. Villani, S. Croft, B. McElroy, R. Venkataraman, S. Philips, and M. Newell, "Evaluation of the LANL Hand Held Multiplicity Shift Register and Canberra JSR-15," Proc. NSS/MIC IEEE, Honolulu, HI, October 28 - November 3 (2007)