Uncertainty Quantification in Neutron Multiplicity Measurements: The Impacts of Nuclear Data

A.Favalli[1*], S. Croft[2], D. Henzlova[1], M.Lockhart[1], B. Weaver[1], T.Burr[1**]

[1]Los Alamos National Laboratory, USA
[2]Oak Ridge National Laboratory, USA & Lancaster University, Bailrigg, Lancaster, UK

Corresponding authors: *afavalli@lanl.gov, **tburr@lanl.gov

Abstract

Confidence in nuclear material balances rests on the quality of physical measurements. The premise behind paired operator/declaration - inspector/measured difference analysis is that measurement data is reliable and accompanied by a well-quantified total measurement uncertainty. In the framework of neutron multiplicity counting, an established neutron-based technique for assay of Pu-item mass, neutron multiplicity equations (so called point-model equations), are solved to obtain Pu mass from three measured rates (Singles, S, Double, D, and Triples, T). The general neutron multiplicity counting approach involves two main steps: neutron detector calibration using a well characterized ^{252}Cf source followed by measurement of unknown Pu items by solution of the point model equations. The ^{252}Cf data is used to estimate detection efficiency, the Doubles gate fraction f_d, and the Triples gate fraction, f_t, on an absolute basis, each with its associated uncertainties. Recently we developed a Bayesian framework for comprehensive uncertainty quantification, which includes all the steps from neutron detector calibration to final estimation of Pu mass and (includes?) needed uncertainty distributions and correlations in all of the estimated assay-item parameters. This paper reports on the impact of the underlying nuclear data and their uncertainties on the measurement results. We also discuss our recently-developed method for high fidelity certification of the ^{252}Cf source neutron yield and the related nuclear data, including the effect of the nuclear data in the final estimation of Pu mass and in the definition of the ^{240}Pu$_{effective}$ mass. Uncertainty associated with nuclear data in all measurement steps from Cf calibration to Pu mass determination is exposed and discussed. Driven by the results, conclusions are presented regarding the impact on nuclear material verification.

Introduction

In nuclear safeguards, confidence in nuclear material balances rests on the quality of physical measurement results. The premise behind paired operator/declaration – inspector/measured difference analysis is that measurement data is reliable and accompanied by a defensible total measurement uncertainty. Passive neutron multiplicity counting (PNMC) is an established technique to assay special nuclear material (SNM), such as plutonium. Neutron multiplicity equations are solved in order to obtain a plutonium mass estimate from three observed counting rates; the so called Singles, S, Doubles, D, and Triples, T.
Neutron multiplicity equations show that under "point model" assumptions, the expected values of the S, D, and T rates (which are related to the factorial moments of the detected neutron distribution) denoted μ_S, μ_D, μ_T, respectively are given by:

$$S = F_S \varepsilon M \nu_{s1}(1+\alpha) \quad (1),$$

$$D = \frac{F_S \varepsilon^2 f_d M^2}{2}\left[\nu_{s2} + \left(\frac{M-1}{\nu_{i1}-1}\right)\nu_{s1}(1+\alpha)\nu_{i2}\right] \quad (2),$$

$$T = \frac{F_S \varepsilon^3 f_t M^3}{6}\left[\nu_{s3} + \left(\frac{M-1}{\nu_{i1}-1}\right)[3\nu_{s2}\nu_{i2} + \nu_{s1}(1+\alpha)\nu_{i3}] + 3\left(\frac{M-1}{\nu_{i1}-1}\right)^2 \nu_{s1}(1+\alpha)\nu_{i2}^2\right] \quad (3)$$

where F_S is the ^{240}Pu-effective ($^{240}Pu_{eff}$) spontaneous fission rate, ε is the neutron detection efficiency, M is the neutron leakage multiplication, α is the (α, n) to spontaneous fission neutron production ratio, f_d is the Doubles gate fraction, f_t is the Triples gate fraction, ν_{s1}, ν_{s2}, and ν_{s3} are the first, second, and third reduced factorial moments of the spontaneous fission neutron distribution, respectively, and ν_{i1}, ν_{i2}, and ν_{i3} are the first, second, and third reduced factorial moments of the induced fission neutron distribution, respectively. The unknown mass of $^{240}Pu_{eff}$ in the item is given by the ratio of F_S and the specific spontaneous fission rate of ^{240}Pu.

The ^{252}Cf spontaneous fission neutron sources also obey the point-model equations, with α = 0 and M =1, and F_s as the spontaneous fission rate of the ^{252}Cf source at the time of the measurements [1,2,3]. When representative items of Pu are not available, a general neutron multiplicity counting approach involves two main steps: detector calibration using a well characterized ^{252}Cf neutron source followed by measurement of unknown Pu items by solution of the point model equations.
How well the neutron yield from ^{252}Cf calibration source is known is an important contributor in determining the accuracy of this neutron multiplicity technique. A reliable and easy to apply in-field method to determine the neutron yield of ^{252}Cf source is needed. We have recently developed a method that provides high fidelity user-certification of ^{252}Cf source [4]. The method relies on equipment commonly used in safeguards measurements, allowing users to self-certify with high accuracy, the neutron yield of ^{252}Cf sealed sources for use in detector calibration. The results of this method and associated uncertainties, including those arising form the nuclear data, are discussed in this paper.

^{240}Pu is the primary spontaneous fission neutron source in Pu-bearing items. The source strength is usually represented by an effective $^{240}Pu_{eff}$ quantity, which is defined as a weighted sum of the ^{238}Pu, ^{240}Pu and ^{242}Pu, even Pu mass, spontaneous fission isotopes. In the definition of the $^{240}Pu_{eff}$, the relative weight of each isotope is in proportion to the product of the specific spontaneous fission rate and the second factorial moment of the prompt neutron emission distribution. The definition of $^{240}Pu_{eff}$ is derived based on the knowledge of the half-lives of the 3 Pu even isotopes [1].

In this paper we will focus our attention on those two aspects of the multiplicity counting: we will discuss the method to establish a Cf source intensity and the associated uncertainty, as well as, based on a revision of the nuclear data associated with Pu isotopes half-lives, we will propose and benchmark, a new set of coefficients for the definition of $^{240}Pu_{eff}$. A general discussion of the impact on the final uncertainties, determined using the recently developed Bayesian frameworks, of the final plutonium mass is reported.

^{252}Cf Calibration Method and Associated Uncertainties Including Nuclear Data

Uncertainty quantification in nuclear material measurements begins with the initial calibration of a detection system using a ^{252}Cf spontaneous fission neutron source. The intensity of the source is used to calculate the detector efficiency and other parameters used in the point model equations. As a result, the ^{252}Cf yield must be known with high precision (<1% uncertainty). Common techniques used by national metrology laboratories, like the National Institute of Standards and Technology (NIST), to obtain high accuracy yield measurements can be costly and time-consuming. An alternative approach, outlined in [4], uses the principles of multiplicity counting (Singles, Doubles, and Triples) and common safeguards equipment to determine the ^{252}C absolute yield, allowing calibration measurements to be performed in-house. In a previous study [5] this alternative method was evaluated using high efficiency neutron

multiplicity counters (>60%) and achieved uncertainties less than 1%. However, some facilities may not have access to high efficiency counters and must depend on lower efficiency detection systems. As a result, an additional study was conducted to apply this alternative calibration method to data acquired in common lower efficiency (coincidence) detectors. The operating parameters of the detectors of interest, including multiplicity as well as coincidence counters, are outlined in Table 1.

Table 1: Operating parameters of detectors used in evaluations

Detector	Pre-delay (μs)	Gate Width (μs)	Die Away (μs)	Efficiency (%)	Dead time coefficient A (μs)	Dead time coefficient B (ps)	Multiplicity dead time (ns)
miniENMC	1.5	24	19.2	61.9	0.155	0.006	38.6
ENMC	1.5	24	21.8	64.2	0.095	0.029	36.8
ENMC/INVS	1.5	24	18.8	80.0	0.341	0.017	100
HLNCC-II	4.5	64	43	17.1	0.768	0.248	215

The key expressions used to calculate the yield of a lightly encapsulated ^{252}Cf neutron source are shown in Equations (4)-(8). S_c represents the dead time corrected net Singles rate and $D_c(0,\infty)$ and $T_c(0,\infty)$ are the dead time corrected net Doubles and net Triples extrapolated to zero pre-delay and infinite gate width. The extrapolation effectively removes the finite pre-delay and gate parameters, specific for each counter and defined in acquisition electronics, as shown in Table 1, and provides the ideal, total correlated count rates (D and T). Parameter $\varepsilon_d/\varepsilon$ is a ratio of detection efficiencies of delayed neutrons to prompt fission neutrons, $v_i, i = 1-3$ correspond to first, second, and third factorial moments of californium spontaneous fission neutron distribution, and $v_t = v_1 + v_d$ where v_d is the mean number of delayed neutrons emitted per fission and $v_d/v_1 = 0.0023(13\%)$. Each of these parameters and the corresponding values for each detector used in the evaluations are listed in Table 2.

$$Y = \frac{1}{r} S_c \left(\frac{D_c(0,\infty)}{T_c(0,\infty)}\right)\left(\frac{v_3/6}{v_2/2}\right) \tag{4}$$

$$Y = \frac{1}{r^2} S_c \left(\frac{S_c}{D_c(0,\infty)}\right)\left(\frac{v_2/2}{v_1}\right) \tag{5}$$

$$Y = \frac{1}{r^{3/2}} S_c \left(\frac{S_c}{T(0,\infty)T_c(0,\infty)}\right)^{1/2}\left(\frac{v_3/6}{v_1}\right)^{1/2} \tag{6}$$

$$Y = D_c(0,\infty))\left(\frac{D_c(0,\infty)}{T_c(0,\infty)}\right)^2\left(\frac{v_t(v_3/6)^2}{(v_2/2)^3}\right) \tag{7}$$

Where:

$$\frac{1}{r^p} = \frac{\left(1+\frac{v_d}{v_1}\right)}{\left(1+\frac{\varepsilon_d v_d}{\varepsilon v_i}\right)^p}, p = 1, 2, \frac{3}{2} \tag{8}$$

Table 2: Nuclear data parameters for detectors in evaluations with percentage uncertainties. The factorial moments are adjusted based on estimated $^{250}Cf/^{252}Cf$ ratio in the source at the date of the measurement.

Detector	ν_1	ν_2	ν_3	$\varepsilon_d/\varepsilon$	$1/r^2$
ENMC	3.7542(0.26%)	11.935(0.16%)	31.610(0.55%)	1.210(4.50%)	0.99676(0.05%)
ENMC/INVS	3.7539(0.26%)	11.933(0.16%)	31.600(0.55%)	1.105(3.30%)	0.99724(0.04%)
miniENMC	3.7541(0.26%)	11.934(0.16%)	31.607(0.55%)	1.226(3.20%)	0.99668(0.05%)
HLNCC-II	3.7514(0.26%)	11.916(0.16%)	31.535(0.55%)	1.447(1.73%)	0.99574(0.06%)

For this paper, we will provide the yield calculations using Equation (5), which only depends on the Singles and asymptotic Doubles count rate because we are most interested in these uncertainties and how they compare to the target of <1% uncertainty needed to compete with national metrology laboratory capability but also if accurate Pu assays are to be obtained. In general, the yield calculations for Equations (4), (5), and (7) have uncertainties greater than 1%, which can be seen in the published evaluation [5].

The same ^{252}Cf neutron source, which was for reference initially certified by NIST, was used for all measurements and the uncertainties associated with the Singles, Doubles, and Triples count rates were calculated by splitting the total measurement time into short repeated measurements. For ENMC, ENMC/INVS, and miniENMC, the total measurement time was 2 hours, which was divided into 720 cycles of 10 seconds. For HLNCC-II, the total measurement time was 12 hours, divided into 2160 cycles of 20 seconds. For lower efficiency detectors, a longer measurement time is necessary to ensure that the uncertainties in the measurements will not greatly affect the yield calculation. Dead time corrected count rates are shown in Table 3. The calculated yield and associated uncertainty using Equation (5) are listed in Table 4.

Table 3: Singles and asymptotic Doubles and Triples dead time corrected rates with uncertainty.

	ENMC	ENMC/INVS	miniENMC	HLNCC-II
Date	30-Oct-15	14-Jun-16	14-Jan-16	4-Apr-19
S_c	284990(110)	307760(10)	266100(10)	32089(1)
$D_c(0,\infty)$	275500(130)	379510(330)	254760(150)	8447(4)
$T_c(0,\infty)$	157630(90)	272200(830)	134370(170)	-

Table 4: Calculated absolute yield using Equation (5) with corresponding uncertainty and decay-corrected NIST yield for comparison.

	ENMC	ENMC/INVS	miniENMC	HLNCC-II
Calculated yield	467100	395700	440400	192800
Uncertainty (%)	0.28	0.30	0.30	0.32
NIST yield	466300	396510	441800	192000

The ^{252}Cf source used in these measurements was measured by NIST in 2014 with an estimated overall uncertainty of approximately 1% at the standard deviation level. The yield calculated by NIST was decay corrected to the date when each measurement was taken and are provided in Table 4. The yield calculations are compared to the decay-corrected yield from NIST to ensure accuracy against a standardized method, and ratios between the calculated and NIST yield are shown in Figure 1.

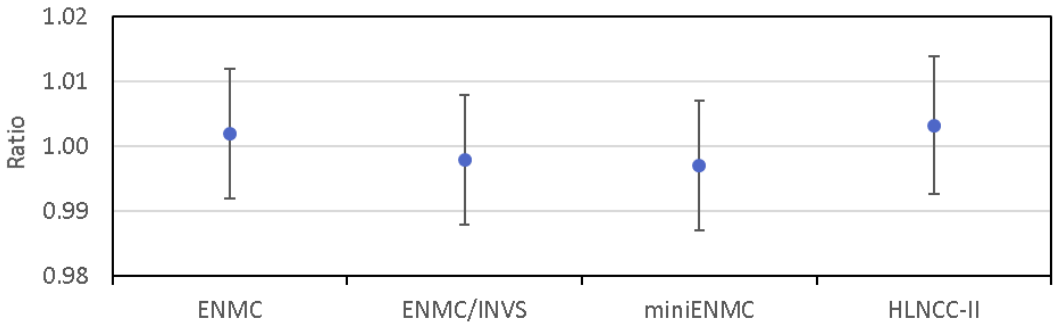

Figure 1: Ratio between calculated yield and NIST yield for each detector.

^{240}Pu effective, the impact of the revised nuclear data

Our team has recently also reviewed the available spontaneous fission half-life data, and the specific fission rate per unit mass, for $^{238, 240, 242}$Pu needed for safeguards applications [6], as they form the basis for the so called ^{240}Pu effective mass definition in terms of the basic nuclear data parameters. Based on the new evaluations the following results were obtained: for ^{238}Pu we recommend a SF half-life of $(4.745\pm0.083)\times10^{10}$ *y* corresponding to a specific SF rate of (1171±20) fis·s^{-1}·g^{-1}, for ^{240}Pu $(1.1608\pm0.0091)\times10^{11}$ *y* and (474.7±3.7) fis·s^{-1}·g^{-1}, for ^{242}Pu $(6.766\pm0.037)\times10^{10}$ *y* and (807.7±4.4) fis·s^{-1}·g^{-1}. The previous review was done by Holden and Hoffman [7] (H&H) about 20 years ago. A comparison between the two evaluations is shown in the following table.

Table 5: Comparison of the specific SF rate from the Holden and Hoffmann evaluation and the recent evaluation of our team.

Nuclide	H&H recommended	Current review	Ratio
^{238}Pu	4.75(9)x10^{10} y	4.745(83)x10^{10} y	1.0008
^{240}Pu	1.14(1)x10^{11} y	1.1608(91)x10^{11} y	1.0182
^{242}Pu	6.77(7)x10^{10} y	6.766(37)x10^{10} y	0.9994

We should notice that for ^{238}Pu and ^{242}Pu the recommended values are quite similar but for ^{240}Pu a significant shift (an increase of about 1.8%) is proposed. In all cases the uncertainty at the 68 % confidence level has been reduced with the reduction for ^{242}Pu being almost a factor of two.

The ^{240}Pu$_{\text{eff}}$ weight fraction of a Pu item, the measured quantity obtained in the neutron multiplicity counting, is defined by a weighted sum of the spontaneously fissile even Pu isotopes present in the item according to the following equation:

$$w_{eff} = \gamma_{238} \cdot w_{238} + w_{240} + \gamma_{242} \cdot w_{242} \qquad (9)$$

where w_{2xy} is the weight fraction of isotope ^{2xy}Pu present in the ^{tot}Pu composition and γ_{2xy} is the ^{240}Pu$_{eff}$ coefficient of nuclide ^{2xy}Pu relative to ^{240}Pu.

For neutron coincidence counting with a neutron detector with a flat neutron energy response, we define the ^{240}Pu$_{eff}$ coefficient of nuclide $2xy$ relative to ^{240}Pu, γ_{2xy}, on a per unit mass basis in terms of basic nuclear data for Doubles counting by the relation:

$$\gamma_{2xy} = \frac{(g \cdot \nu_2)_{2xy}}{(g \cdot \nu_2)_{240}} = \frac{\left(\frac{\nu_2}{A \cdot t_{1/2}}\right)_{2xy}}{\left(\frac{\nu_2}{A \cdot t_{1/2}}\right)_{240}} \qquad (10)$$

where g is the specific spontaneous fission rate, in fissions per unit mass, ν_2 is the second factorial moment of the spontaneous fission prompt neutron multiplicity distribution, A is the molar mass and $t_{1/2}$ the SF half-life of the nuclide.

For illustration we take the numerical values of ν_2 for ^{238}Pu, ^{240}Pu and ^{242}Pu to be 3.957, 3.789 and 3.809, respectively [8]. The corresponding γ-coefficients are given in Table 6.

Table 6: γ_{2xy} comparison based on the previous and more recent evaluation.

Nuclide	H&H recommended $t_{1/2}$	Using $t_{1/2}$ from current review	Ratio
^{238}Pu	2.528	2.576	1.0193
^{240}Pu	1	1	1 (exact)
^{242}Pu	1.679	1.710	1.0188

The impact of the recommended changes to the SF half-life can be illustrated by considering the ^{240}Pu$_{eff}$ weight fraction in % calculated across a range of isotopic compositions from low burnup to high burnup plutonium. The PIDIE set [9] provides a convenient range. The w_{eff} values calculated using both the Holden & Hoffman half-lives [7] and Croft & Favalli [6] (C&F) half-lives are shown in Table 7 along with the ratio, and double ratio with respect to the lowest burnup composition. The double ratio represents the proportionate change in Pu-mass assay value that one would observe if one calibrated using low burnup standards. For the higher burnup (reactor grade) materials the shift is quite large at several tenths of percent.

Table 7: Comparison of the calculation of the effective mass for the H&H [7] and our (C&F) [6] half-lives for sample of increasing burn-up .

	PIDIE Item Identification						
	#1.1	#1.2	#1.3	#1.4	#1.5	#1.6	#1.7
	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%
^{238}Pu	0.011007	0.022593	0.047158	0.108416	0.131373	0.92975	1.252848
^{240}Pu	5.990245	10.09933	14.14422	19.76731	21.2169	23.88933	25.5941
^{242}Pu	0.034621	0.094328	0.233843	0.566857	0.701724	3.556296	4.686304
w_{eff}(H&H)	6.076185	10.31479	14.66598	20.99295	22.72698	32.20948	36.61112
w_{eff}(C&F)	6.077818	10.31888	14.66598	21.01618	22.7556	32.3674	36.82025
Ratio	1.000269	1.000396	1.000662	1.001107	1.001259	1.004903	1.005712
Double Ratio	1	1.000128	1.000393	1.000838	1.00099	1.004633	1.005442

Note that, as discussed in [8] the traditionally used γ-coefficients are 2.52 and 1.68 while the best directly determined values, which have estimated overall uncertainties, substantially less than 1%, are 2.713 and 1.663. The present values (2.576 and 1.710) are roughly 5% lower and 3% higher than the directly determined values suggesting the situation remains far from satisfactory from a metrology perspective. Fortunately, by using representative calibration standards for practical Pu verification and assay the bias induced by the large uncertainty in the γ-coefficients can be managed, but the present re-evaluation reveals that our knowledge of the basic spontaneous fission nuclear data parameters for the even Pu isotopes remains surprisingly poor compared to the notional capability of the techniques available to determine them. It should also be noted that getting suitable Pu items for benchmarking of calibrations is increasingly difficult.

Impact of the uncertainty on the neutron yield from the ^{252}Cf Calibration Method on the ^{240}Pu$_{eff}$ mass

To quantify possible improvements in the error relative standard deviation (RSD) for ^{240}Pu$_{eff}$ mass, approximate Bayesian computation (ABC) was applied using a nominal RSD of 0.017 and a reduced relative standard deviation (RSD) of 0.005 for the assigned value of F_S for ^{252}Cf. Note that Table 4 lists RSDs of approximately 0.3% based on Eq. (5), so the 0.5% RSD used here allows for errors arising due to item-specific variation around the assumptions in Eq. (5).

ABC has been described for passive neutron multiplicity counting [10]. For example, the actual coverages of nominal 95% probability intervals is very close to 95%. In Fig. 2, the posterior RSD is 0.022 for the 0.017 RSD case and 0.014 for the 0.005 RSD case. Note that the mean values for both cases is 0.55, which is 1.7% lower than the nominal value of 0.56 g, but also note that the nominal value of 0.56 g is well within the posterior probability density functions (pdfs). The Bayesian approach allows for comprehensive treatment of all known error sources, including errors in nuclear data.

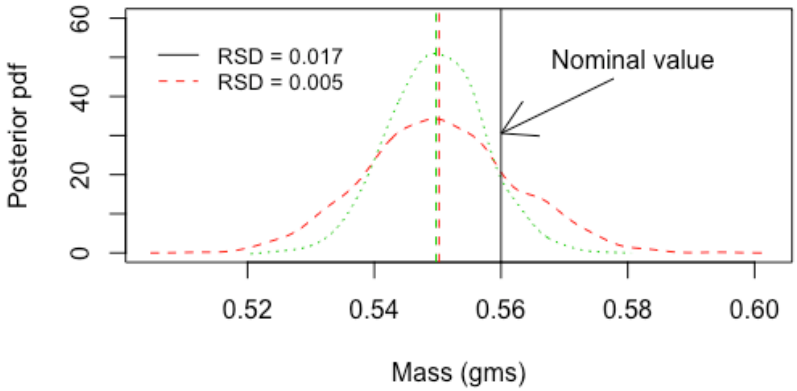

Figure 2: ABC-based posterior pdf for a real item having nominal value 0.56 gm using an RSD in F_S for ^{252}Cf of 0.017 (nominal/original) or of 0.005 (improved). The posterior pdf has RSD 0.022 using the nominal RSD of 0.017 and 0.014 using an improved RSD of 0.005.

Conclusion and remarks

In this paper we focus on two important aspects and sources of uncertainties in the ^{240}Pu mass determined by the widely used neutron multiplicity counting method. When representative Pu items are not available for empirical calibration, neutron multiplicity counting in general involves two main physics-based steps: calibration using ^{252}Cf followed by measurement of unknown Pu items by solution of the point model equations. The paper reported on recently developed method for high fidelity in-house ^{252}Cf neutron source calibration, and how the underlying uncertainties are related to nuclear data. In the second part of the paper we discussed the impact of nuclear data on the definition of ^{240}Pu$_{eff}$ mass, the assay quantity extracted by inversion of the point model equations. The new data provide first a new ^{240}Pu specific fission rate with the associated error, and a new set of coefficients for the definition of ^{240}Pu$_{eff}$ with an improvement of the determination of total Pu mass of the item by complementing the results with the gamma isotopic measurements. The results of the comparison with the previous coefficients are a reminder to the community how the nuclear data for the even Pu isotopes (spontaneous fission half-lives and specific fission rates) are overdue for re-measurement at higher accuracy and precision.

Acknowledgments. The work presented in this paper was funded by the National Nuclear Security Administration of the Department of Energy, Office of International Nuclear Safeguards (LANL release number LA-UR 21-26234)

References

[1] N. Ensslin, W.C. Harker, M.S. Krick, D.G. Langner, M.M. Pickrell and J.E. Stewart, Application Guide to Neutron Multiplicity Counting, LA-13422-M (1998).

[2] S. Croft, A. Favalli, D.K. Hauck, D. Henzlova, P.A. Santi, Feynman variance-to-mean in the context of passive neutron coincidence counting, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 686, (2012), pp136-134.

[3] A.Favalli, S.Croft, P.Santi, Point model equations for neutron correlation counting: Extension of Böhnel′s equations to any order, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 795, (2015), pp.370-375.

[4] S.Croft, D.Henzlova, Determining ^{252}Cf source strength by absolute passive neutron correlation counting, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 714, (2013), pp.5-12

[5] D.Henzlova, A.Favalli, S. Croft, In-depth evaluation of ^{252}Cf absolute calibration by passive neutron correlation counting method, Metrologia, Volume 56, Number 1.

[6] S.Croft, A.Favalli, Review and Evaluation of the Spontaneous Fission Half-lives of ^{238}Pu, ^{240}Pu, and ^{242}Pu and the corresponding specific fission rates, Nuclear Data Sheets, Volume 175 (2021), pp.

[7] N.E. Holden and D.C. Hoffman, Spontaneous fission half-lives for ground-state nuclides Pure Appl. Chem. 72(8)(2000)1525-1562. See also Errata Pure Appl. Chem. 73(7)(2001)1225-1227.

[8] S.Croft, S.Cleveland, A. Nicholson, Calculation of the ^{240}Pu-Effective Coefficients for Neutron Correlation Counting, Oak Ridge National Laboratory report ORNL/LTR-2014/559 (2014).

[9] J.Morel and B.Chauvenet, Intercomparaison des mesures de composition isotopique du plutonium par spectrometrie X et gamma Resultats de l'action "PIDIE" Rapport Final Rapport Commissariat a l'Energie Atomique CEA-R-5582(1992).

[10] T. Burr, S. Croft, D. Henzlova, B. Weaver, A. Favalli, Bayesian Bottom-up Uncertainty Quantification In Neutron Multiplicity Measurements: Providing Uncertainty Distributions And Correlations In All The Assay-item Parameters, proceeding of INMM Annual Meeting 2020 (virtual).