



## Identifying neutron shielding in neutron multiplicity counting

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### ABSTRACT

Neutron Multiplicity Counting (NMC) and passive neutron interrogation is becoming a standard procedure in special nuclear material control and accountability, due to the relative optical transparency of other structure materials to neutron flux, making it very effective for measuring composite poorly-characterized samples. Currently, all applicable neutron multiplicity counting methods assume that both the detection efficiency and the neutron die-away time are system parameters, independent of the sample. Clearly, if the detection efficiency of the system is reduced due to neutron absorption or moderation inside the sample, the measurement will be biased. Therefore, detecting a reduction in the detection efficiency, either accidental or deliberate, is a well motivated problem. In the present study we develop, through both theory and implementation, a new method for detecting a reduced effective detection efficiency in NMC by sampling the fourth central moment of the count distribution. One of the attributes that make the proposed method appealing is that it does not require any additional operation and may be easily applied to any standard NMC counter, assuming that the data is recorded in LIST mode. The method is implemented on a set of 18 measurements, 7 of which are standard NMC measurements, and in the remaining 11 the sample is covered by a moderator, noticeably reducing the effective detection efficiency. The new method successfully discriminates between them.

### 1. Introduction

In the standard practice of Neutron Multiplicity Counting (NMC), the first three sampled factorial moments of the neutron count distribution are used in an inversion model to extract the spontaneous fission rate, the  $(\alpha, n)$  rate, and the multiplication of the sample [1].

In recent years, passive neutron interrogation and NMC is becoming a standard tool in the community of nuclear safeguards, safety and security, mainly due to the relative optical transparency of many structure materials to neutron flux, proving NMC to be very useful in measuring poorly characterized, composite samples. In respect, extensive work is done in recent years improving the existing methods, in terms of simulation method [2,3], spatial corrections [4], uncertainty analysis and quantification [5,6] and more.

One of the basic parameters in any NMC measurements is the detection efficiency, defined as the probability of a neutron to be detected, which explicitly appears in the inversion formula. Currently, all NMC methods assume that the detection efficiency is a system

parameter independent of the sample. Typically, the detection efficiency is either known or calibrated using a well characterized neutron source. In return, if the detection efficiency is compromised due to absorbing or scattering materials in the measured sample, the outcome of the measurement is bound to be biased.

While the assumption that the sample does not affect the detection efficiency is, in general, a reasonable assumption, it is definitely not a certainty. For instance, the risk of a decrease in the detection efficiency was recently described in [7], reporting the difficulties arising in implementation of NMC methods in the Fukushima–Daiichi decommissioning. In particular, the presence of neutron absorbers, originating from the damaged reactor's regulation system, cause a reduction in the effective detection efficiency.

In current high efficiency detection systems, the sample cavity is separated from the detection system by a thin Cd layer, aimed to prevent neutrons moderated in the moderating media surrounding the detectors to re-enter the sample and induce additional fissions. Therefore, a

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reduction in the neutron count may be caused by the mere presence of a moderator inside the sample cavity, such as polyethylene (PE) or even lead. In the present study, any decrease in the detection efficiency caused by the sample itself is referred to as a *shielding effect*.

Currently, the most well known method for detecting (and correcting) a bias in the detection efficiency due to neutron shielding is the “add a source” method [8]. The AAS methods, involves, in general lines, adding an additional (calibrated) neutron source to the measurement, allowing the user to estimate (and quantify) neutron absorption (in the sample or the Cd lining) due to additional materials in the sample.

The outline of the present study is to introduce, through both theory and implementation, a new method for detecting a reduction in the detection efficiency. The implementation of the method involves sampling the fourth moment of the count distribution, but does not require any additional operations nor extension of the measurement system, and may be easily applied to any standard NMC counter.

The paper is arranged in the following manner: the remainder of the present section gives some general background on NMC. Section 2 gives the theoretical background for the method. In Section 3 we analytically derive explicit formula for the fourth moment of the count distribution. Section 4 gives a detailed description of the method. Section 5 is devoted for experimental results and in Section 6 conclusions are given.

## 2. Scientific background

### 2.1. Neutron multiplicity counting

In neutron multiplicity counting, the different statistical attributes of the three neutron sources is used to quantify each source by sampling not only the average count rate, but also the second and third factorial (or central) moments of the count distribution. For that aim, the first three moments of the count distribution must be expressed in terms of three sources. Due to the high complexity of the mathematical modeling, the expressions are derived under the assumption that all the reaction rates can be averaged in a reasonable manner (over space and energy), and the dynamics of the count distribution (often referred to as the *stochastic transport*) is modeled using the point model.

In the point model, which is at force in all NMC methods, the sample is defined by three attributes [9]:

1.  $S$ , the total rate of spontaneous source events (both the spontaneous fissions and  $(\alpha, n)$  reactions).
2.  $S_f$ , the rate of the spontaneous fission source only. For convenience, this parameter is expressed using  $U$ , the ratio between fission neutrons' rate to the total neutron rate ( $U = S_f/S$ ).
3.  $M_L$ , the leakage multiplication factor, which quantifies the contribution of the induced fissions to the total count distribution [10].

Since the mass of spontaneously fissile material is proportional to  $S_f$  in a known proportion, the purpose of the modeling is to obtain an inversion between the sampled moments of the count distribution and the sample parameters.

The term “inversion formula”, in the present context, refers to a well defined set of equations connecting the three unknowns  $S$ ,  $U$ , and  $M_L$  and the sampled moments [1,9].

In the inversion formula, as we would obviously expect, the detection efficiency is assumed to be a system parameter. The detection efficiency can be pre-calibrated using well defined neutron source, but for most commercial systems (such as the PSMC and AWCC), the detection efficiency is well known [11,12].

### 2.2. The SVM method

The SVM method, introduced by the authors in [9], is a fairly new NMC technique, based on a correlation between the first three central

moments (the Mean, Variance, and Skewness) of the number of detections in a given time interval and the measurement parameters: sample parameters ( $S$ ,  $U$ , and  $M_L$ ), the measurement system parameters (the neutron die-away time  $\frac{1}{\lambda}$  and the detection efficiency  $P_d$ ), and the  $k_{th}$  factorial moments of the spontaneous and induced fission multiplicities  $D_{if,k}$ ,  $D_{sf,k}$ ,  $k = 1, 2, 3$  (respectively), which depend on the material and may be found in the literature.

The first three central moments of the number of detections are sampled in the following manner: the measurement is divided into  $N$  consecutive gates of duration  $T$  ( $T$  is, typically, 2 or 3 die-away times [9]) and the number of detection in the  $n$ th gate is denoted by  $X_n$ ,  $n = 1, 2, \dots, N$ . Then, the mean ( $E$ ), variance ( $V$ ) and skewness ( $Sk$ ) are sampled by:

$$E = \frac{1}{N} \sum_{j=1}^N X_j, \quad V = \frac{1}{N} \sum_{j=1}^N (X_j - E)^2, \quad Sk = \frac{1}{N} \sum_{j=1}^N (X_j - E)^3. \quad (1)$$

The correlation between the system parameters and the observables is done through the following set of equations [9]:

$$\begin{aligned} SD_{g,1}(U, M_L) &= \frac{E}{P_d T} \\ SD_{g,2}(U, M_L) &= \frac{(V - E)\lambda}{P_d^2 (e^{-\lambda T} - 1 + \lambda T)} \\ SD_{g,3}(U, M_L) &= \frac{(Sk - 3V + 2E)2\lambda}{P_d^3 (4e^{\lambda T} - e^{-2\lambda T} - 3 + 2\lambda T)}, \end{aligned} \quad (2)$$

where  $T$  is the duration of the time interval in which the detections are counted,  $D_{g,j}(U, M_L)$  ( $j = 1, 3$ ) are the generalized factorial moments written in terms of the source to noise ratio  $U$  and the leakage multiplication factor  $M_L$ , given explicitly by (see [10] for full details):

$$\begin{aligned} D_{g,1}(U, M_L) &= (U(D_{sf,1} - 1) + 1)M_L \\ D_{g,2}(U, M_L) &= M_L^2 \left( U D_{sf,2} + \frac{M_L - 1}{1 - D_{if,1}} (U(D_{sf,1} - 1) + 1) D_{if,2} \right) \\ D_{g,3}(U, M_L) &= M_L^3 \left( U D_{sf,3} + \frac{M_L - 1}{1 - D_{if,1}} (3U D_{sf,2} U_{if,2} \right. \\ &\quad \left. + D_{if,3} (U(D_{sf,1} - 1) + 1)) \right. \\ &\quad \left. + 3 \left( \frac{M_L - 1}{1 - D_{if,1}} \right)^2 D_{if,2}^2 (U(D_{sf,1} - 1) + 1) \right). \end{aligned} \quad (3)$$

Once  $S$  and  $U$  are known, then the spontaneous fission rate is nothing more than  $S_f = U \times S$ , and the mass is estimated by  $\frac{S \times U}{473.5}$  [1].

The SVM formalism and the Multiplicity formalism may be correlated by the following formulas:

$$\begin{aligned} Singles &= \frac{E}{T} \\ Doubles &= \frac{V - E}{e^{-\lambda T} - 1 + \lambda T} \lambda \\ Triples &= \frac{Sk - 3V + 2E}{4e^{-\lambda T} - e^{-2\lambda T} - 3 + 2\lambda T} 2\lambda \end{aligned} \quad (4)$$

## 3. Analytic derivation of the fourth central moment of the count distribution

The idea presented in this study is to add one more equation of the fourth central moment to our set of equations, which will be later used as an “indicator” to verify that the value used for the detection efficiency is indeed correct.

The fourth central moment, here denoted by  $K$ , is defined as

$$K = E [(X - E(X))^4], \quad (5)$$

where the notation  $K$  stands for “Kurtosis”.<sup>1</sup>

<sup>1</sup> In most literature, the Kurtosis is not defined exactly as in Eq. (5), rather it is normalized such that Kurtosis of a Poisson distribution is equal to the mean.

The first goal is to write an explicit formula for the fourth central moment of the number of detections in terms of the system parameters. From a technical point of view, obtaining an analytic expression for the fourth central moment is done in two steps:

1. Derivation of the fourth moment in terms of the generalized factorial moments: In [9], this is done for the first three central moments, and the expansion to the fourth central moment may be achieved using the exact same argumentation.
2. Derivation of the fourth generalized factorial moments  $D_{g,4}(U, M_L)$ : For the first three factorial moments this was done in [10], and derivation of the fourth moment can be done in a very similar manner (see also [13]).

### STEP 1: Derivation of the fourth moment in terms of the generalized factorial moments

Following the exact same argumentation presented in [9], the fourth central moment of the count distribution may be written as:

$$K = I_1 + 7I_2 + 3I_1^2 + 6I_1I_2 + 6I_3 + 3I_2^2 + I_4, \quad (6)$$

where

$$I_k = P_d^k D_{g,k} \int_0^\infty (1 - e^{-\lambda T} - U_0(t-T)(1 - e^{-\lambda(t-T)}))^k dt; \quad k = 1, \dots, 4 \quad (7)$$

The first 3 integrals were computed in [9], resulting with:

$$\begin{aligned} I_1 &= P_d D_{g,1} T \\ I_2 &= P_d^2 D_{g,2} \frac{e^{-\lambda T} - 1 + \lambda T}{\lambda} \\ I_3 &= P_d^3 D_{g,3} \frac{4e^{-\lambda T} - e^{-2\lambda T} - 3 + 2\lambda T}{2\lambda}, \end{aligned} \quad (8)$$

and through direct calculations we obtain:

$$\begin{aligned} I_4 &= P_d^4 D_{g,4} \int_0^\infty (1 - e^{-\lambda T} - U_0(t-T)(1 - e^{-\lambda(t-T)}))^4 dt \\ &= P_d^4 D_{g,4} \frac{e^{-3\lambda T} (2 - 9e^{\lambda T} + 18e^{2\lambda T} - 11e^{3\lambda T} + 6\lambda T e^{3\lambda T})}{6\lambda}, \end{aligned} \quad (9)$$

which completes the first step.

### STEP 2: Derivation of the fourth generalized factorial moment

For a multiplying system, the generalized factorial moments are defined as the factorial moments of the number of neutrons emitted in the entire neutron chain, initiated by a source event. The analytic expression for the generalized factorial moments are obtained in the following manner: define  $h(x) = \sum_{n=0}^\infty x^n a_n$  as the probability generating function of the number of neutrons emitted due to a *single fission event*, and then evaluate the first four factorial moments of  $\{a_n\}_{n=0}^\infty$  as follows [13]:

$$\begin{aligned} d_1 &= h'(1) = \frac{1 - p_f}{1 - p_f D_{if,1}} \\ d_2 &= h''(1) = \frac{p_f D_{if,2} (1 - p_f)^2}{(1 - p_f D_{if,1})^3} \\ d_3 &= h^{(3)}(1) = \frac{p_f (1 - p_f)^3}{(1 - p_f D_{if,1})^4} \left( D_{if,3} + 3D_{if,2}^2 \frac{p_f}{1 - p_f D_{if,1}} \right) \\ d_4 &= h^{(4)}(1) = \frac{p_f}{1 - p_f D_{if,1}} \left\{ D_{if,4} \left( \frac{1 - p_f}{1 - p_f D_{if,1}} \right)^4 \right. \\ &\quad + 6D_{if,3} \left( \frac{1 - p_f}{1 - p_f D_{if,1}} \right)^2 \frac{p_f D_{if,2} (1 - p_f)^2}{(1 - p_f D_{if,1})^3} \\ &\quad + 4D_{if,2} \frac{p_f (1 - p_f)^4}{(1 - p_f D_{if,1})^5} \times \\ &\quad \left. \left( D_{if,3} + 3D_{if,2}^2 \frac{p_f}{1 - p_f D_{if,1}} \right) \right\} \end{aligned}$$

$$\left. + 3D_{if,2} \left( \frac{p_f D_{if,2} (1 - p_f)^2}{(1 - p_f D_{if,1})^3} \right)^2 \right\}. \quad (10)$$

Notice,  $d_j$ ,  $j = 1, \dots, 4$  depend only on the induced fission probability and the induced fission factorial moments.

Next, define  $H(x) = \sum_{n=0}^\infty x^n b_n$  as the probability generating function of a number of neutrons emitted in the entire neutron chain, initiated by a single source event. Following the exact same argumentation as in [10], the generalized 4<sup>th</sup> factorial moment is given by  $D_g(4) = H^{(4)}(x)|_{x=1}$ . Or, explicitly:

$$\begin{aligned} D_{g,4}(U, M_L) &= U D_{sf,4} d_1^4 + 6U D_{sf,3} d_1^2 d_2 + 3U D_{sf,2} d_2^2 \\ &\quad + 4U D_{sf,2} d_1 d_3 + (U D_{sf,1} + 1 - U) d_4 \end{aligned} \quad (11)$$

Implementation of Eq. (10) in Eq. (11) gives an explicit formula for  $D_{g,4} = D_{g,4}(U, M_L)$ , which concludes the second step. Notice, Eq. (10) is written in terms of the fission probability  $p_f$  rather than  $M_L$ , but the transformation is trivial using the equality  $M_L = \frac{p_f}{1 - D_{f,1} p_f}$ .

Finally, through algebraic considerations, we obtain that:

$$K(X) = 6E(X) - 11Var(X) + 6Sk(X) + 3Var^2(X) + Q, \quad (12)$$

where

$$Q = P_d^4 S D_{g,4}(U, M_L) \times \frac{e^{-3\lambda T} (2 - 9e^{\lambda T} + 18e^{2\lambda T} - 11e^{3\lambda T} + 6\lambda T e^{3\lambda T})}{6\lambda} \quad (13)$$

These formula form the theoretical basis of the method introduced in this study, which will be described in details in the next section.

## 4. Detecting neutron shielding using the quadruples rate

### 4.1. Theory

Once the first four central moment are sampled,  $Q$  may be realized in two different ways. First, it can be sampled directly using the equality

$$Q = K(X) - (6E(X) - 11Var(X) + 6Sk(X) + 3Var^2(X)) \quad (14)$$

The value of  $Q$  obtained via Eq. (14) is denoted by  $Q_m$  (“*m*” for “measured”).

A second realization may be obtained in the following manner: Once the first three central moments are sampled, Eqs. (2) and (3) can be solved, followed by substitution of the values of  $S$ ,  $U$ , and  $M_L$  (together with the system parameters  $P_d$  and  $\lambda$ ) into Eq. (13). The value of  $Q$  obtained in this form is denoted by as  $Q_c$  (“*c*” for “calculated”).

Theoretically,  $Q_c$  and  $Q_m$  should be equal (up to measurement uncertainties). However, if the effective efficiency of the system differs from the declared value, then the values are expected not to be equal. Therefore, a strong discrepancy between  $Q_c$  and  $Q_m$  can serve as an indication for a deviation from the declared detection efficiency, or, equivalently, an indication of neutron shielding.

Theoretically, Eq. (14) might be an algebraic combination of Eq. (3). In such case, this method will not work. While we did not prove analytically that this is not the case, the conditions for this to happen are unlikely to occur. As shown later, the experimental results clearly indicate that Eq. (14) is not an algebraic combination of Eq. (3).

### 4.2. How good is good enough?

As stated earlier, the main idea is to compare two different realizations of  $Q$ :  $Q_m$  and  $Q_c$ . Even if the values would theoretically be equal, the actual numeric values will always be subjected to experimental uncertainties, creating a discrepancy between the values. Thus, the “go/no go” condition will not be of the form “ $Q_m = Q_c$ ”, but rather a condition of the form  $D = |Q_m - Q_c|/Q_m < \epsilon$ : If  $D < \epsilon$ , then it is assumed that the sample is *not* shielded, and if  $D > \epsilon$ , then it is assumed that the

**Table 1**  
Experimental setting for the un-shielded measurements.

Sample no.	Total Pu mass [g]	<sup>240</sup> Pu effective mass [g]	Counter type	Detection efficiency $P_d$	Measurement duration [min]
1	4.79	1.108	PSMC	54%	100
2	20.57	5.41	AWCC	33%	60
3	49.7	6.51	AWCC	33%	60
4	6.7	1.4	JCC31	16%	50

sample is shielded. The parameter  $\epsilon$  is referred to as the *tolerance* of the procedure. As in any “go/no go” classification, choosing the tolerance balances between two types of false result: “false negative”, i.e., the procedure indicates that the sample is not shielded when it is, and “false positive”, i.e., when it is decided that the sample is shielded, when in fact it is not. Clearly, there is always a trade-off between the false positives and the false negatives: As  $\epsilon$  increases, the probability of a false positive is reduced at the expense of increased probability of false negative (and vice versa). Thus, one of the goals of this study, is to choose proper values for the tolerance  $\epsilon$ , and quantify the trade-off between the false positive and the false negative.

## 5. Experimental verification of the method

The present section is devoted to experimental implementation of the method introduced, followed by a discussion on the optimal value of the tolerance  $\epsilon$ . The experimental results are divided into two different settings. In Section 5.1, the difference between  $Q_c$  and  $Q_m$  is sampled on a set of 4 standard measurements, in two manners: First,  $Q_c$  and  $Q_m$  are sampled without any additional manipulation. Second,  $Q_c$  and  $Q_m$  are sampled after an artificial “shielding” is inflicted, by randomly removing a certain fraction of the detection events. In Section 5.2, the method is implemented on a set of 18 measurements, where in some of the measurements the sample is covered by a PE or Lead cup, creating an observable reduction in the count rate.

The uncertainty indicated in all the tables and figures refers to the statistical uncertainty, obtained using Statistical Analysis of Data Cycles (SADC) as introduced in [14].

### 5.1. Experiment 1: non-shielded measurements

#### 5.1.1. Experimental setting 1

The initial validation of the method is performed using a set of 4 measurements, measured using 3 standard neutron coincidence counters: JCC31 [15], AWCC [1], and the PSMC [11]. The measurements were taken at the PERLA facility in the JRC laboratory, Ispra, Italy. The characteristics of each system and measured samples are listed in Table 1. All samples, except sample number 1, consist of pure Plutonium with varying isotopic composition. Sample number 1 also contains a small fraction (3.6%) of Gallium. The detector die-away time was estimated to be 50  $\mu$ s for all measurement systems.

For each of the following measurements a shielding effect is emulated by randomly deleting a fraction  $F$  of the counts. This is done for  $F = 0.1, 0.2, 0.3, 0.4, 0.5$ . For each value of  $F$ , the effective detection efficiency is given by  $P_d \times (1 - F)$ .

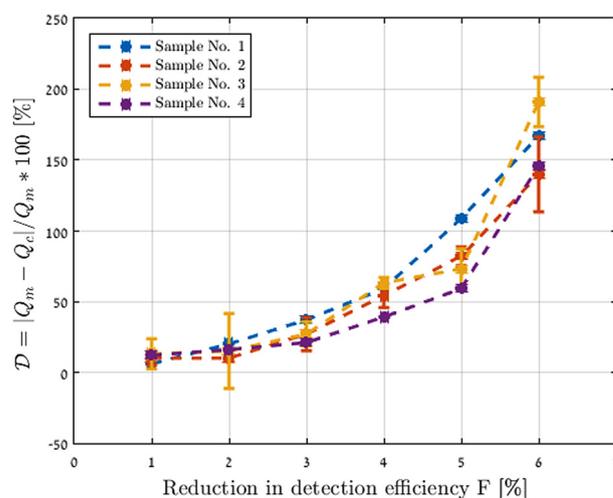
#### 5.1.2. Experimental results 1

For each value of  $F$ , the quantity  $D$  is calculated according to Section 4.2. Theoretically, for  $F = 0$ ,  $D = 0$  as well. In practice, this is never the case, because the measurement is always subjected to numeric, systematic, and statistical uncertainties. On the other hand, one should expect that as  $F$  grows, the discrepancy (hence  $D$ ) grows as well. The experimental results (in terms of  $D$ ) for all samples are shown in Table 2 and Fig. 1.

As can be seen, the prediction that  $D$  increases with  $F$  is met in all the samples. A second thing to notice is that the results obtained using the

**Table 2**  
Experimental results in terms of  $D$  for the non-shielded measurements.

Sample no.	$F$					
	0	0.1	0.2	0.3	0.4	0.5
1	$6 \pm 0.3$	$20 \pm 0.2$	$37 \pm 0.2$	$59 \pm 0.1$	$109 \pm 0.5$	$167 \pm 1$
2	$10 \pm 4$	$10 \pm 2.5$	$27 \pm 12$	$54 \pm 8$	$82 \pm 7$	$140 \pm 26$
3	$13 \pm 10$	$15 \pm 23$	$27 \pm 8$	$63 \pm 4$	$73 \pm 15$	$191 \pm 17$
4	$12 \pm 4$	$16 \pm 4$	$21 \pm 4$	$39 \pm 3$	$59 \pm 5$	$146 \pm 13$



**Fig. 1.** The measured value of  $D$  with respect to the fraction of detections removed  $F$ .

PSMC seem to be more consistent and reliable than the results obtained by other systems. First, for  $F = 0$  the discrepancy (in terms of  $D$ ) is minimal. Second, for  $F > 0$ , the discrepancy is largest (with a single exception). Finally, the uncertainty is very small. This can be attributed to the fact that the measurement duration in the PSMC was a bit longer, but more significantly, to the high detection efficiency characterizing the PSMC.

Still, the true question is not whether or not  $D$  increases with  $F$ , but rather, is there a value of  $\epsilon$  that allows for distinct determination whether the sample is shielded or not. The answer, naturally, depends on  $F$ . For small values of  $F$  (10%), the shielded and non-shielded signals cannot be fully separated using a single value of  $\epsilon$ . But for  $F \geq 0.2$ , by choosing  $\epsilon = 0.15$  (and  $D \geq 0.15$  as the shielding criterion), a 100% success rate can be achieved both in terms of false positive and false negative, and the results are statistically significant (in the sense that the statistical uncertainty does not cover the threshold value). Finally, it should be noted that these results are true for several different systems with various values of  $P_d$ , as shown in Table 2.

### 5.2. Experiment 2: shielded measurements

#### 5.2.1. Experimental setting 2

Based on the satisfactory results of the preliminary validation done using non-shielded samples, the proposed method is implemented on a

**Table 3**  
Effective  $^{240}\text{Pu}$  mass of the measured samples.

Sample no.	1	2	3	4	5
Effective $^{240}\text{Pu}$ mass [g]	1.58	1.18	0.98	0.39	1.1

**Table 4**  
Measurement's duration (in minutes) for all 18 experiments.

Sample no.	Date	Non-shielded	PE cup		Lead cup
			Small	Large	
1	May 2012	190	–	220	–
1	Nov. 2014	20	20	150	100
2	May 2012	300	180	266	–
3	May 2012	110	380	930	–
3	Nov. 2014	40	–	70	–
4	May 2012	970	990	–	–
5	Nov. 2014	20	20	–	–

set of 18 measurements, from which in 11 measurements the sample was shielded. All measurements were done using a standard PSMC at the JRC laboratory, in Ispra, Italy. The measurements were conducted on a set of 5 plutonium samples, four of which (No. 1–4) have approximately the same plutonium mass of 6.6 g (differing only in the isotopic composition), and one sample (No. 5) containing a total 4.73 g of plutonium. The effective  $^{240}\text{Pu}$  mass of each sample is given in Table 3.

The samples were measured in 4 different configurations:

1. plain NMC measurements of the samples.
2. the samples were placed inside a small polyethylene (PE) cup.
3. the samples were placed inside a large PE cup.
4. the samples were placed inside a lead cup.

The small PE cup had a diameter of 10 cm, height of 8 cm, and a total weight of 0.5 kg. The large cup had a diameter of 15 cm, height of 13 cm, and a total weight of 1.6 kg. The lead cup had a diameter of 9.5 cm, height of 14 cm, and a total weight of 9.5 kg. As mentioned, although no absorber is added to the samples, the moderator around the sample is enough to create a shielding effect due to the Cd lining between the sample cavity and the detector rings.

The small and large PE cups reduce the count rate by approximately 9% and 38%, respectively, whereas the lead cup reduce the count rate by 5%. Measurements were taken in two separate campaigns, held on May 2012 and Nov. 2014. Some of the measurements were repeated to check the robustness of the method.

The measurements' duration are listed in Table 4. As can be seen, the measurements taken during the first campaign, which served as the feasibility test of the method, are significantly longer to avoid substantial statistical uncertainties. In the second campaign the measurements' duration are typical for actual NMC measurements.

### 5.2.2. Experimental results 2

The differences between  $Q_m$  and  $Q_c$  (in percentage) for all 18 measurements are summarized in Table 5. The full results of the measurements, in terms of the count rate and all four central moments, are given in Appendix. As can be see, the results are even more distinct than in the previous section, i.e., using a 13% tolerance ( $\epsilon = 0.13$ ) yields perfect distinction between the shielded and non-shielded measurements, with zero false positive/negative.

### 5.3. Quantification of additional effects due to moderation

The purpose of covering the sample with cups made of neutron moderating materials is to reduce the count rate by increasing neutron absorption in the Cd lining. However, covering the sample with a

**Table 5**  
Measured values of  $\mathcal{D}$  (%) for all 18 measurements.

Sample no.	Date	Non-shielded	PE cup		Lead cup
			Small	Large	
1	May 2012	11 ± 2%	–	108 ± 1%	–
1	Nov. 2014	5 ± 2.5%	17 ± 3%	91 ± 4%	15 ± 1%
2	May 2012	2 ± 2%	24 ± 2%	87 ± 3%	–
3	May 2012	7 ± 1%	28 ± 2%	85 ± 2%	–
3	Nov. 2014	3 ± 2%	–	87 ± 4%	–
4	May 2012	1 ± 3%	18 ± 3%	–	–
5	Nov. 2014	6 ± 2.5%	16 ± 1.5	–	–

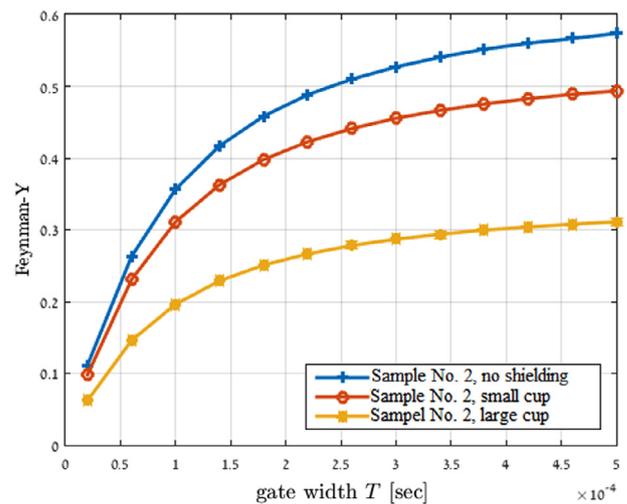


Fig. 2. The Feynman-Y curves as a function of the gate width  $T$ .

moderating medium may create two additional side-effects: increase of the neutron die-away time, and change of the neutron multiplication factor  $M_L$ . In the present section these effects are studied as well as their impact on the validity of the presented results.

#### 5.3.1. Effect on the neutron die-away time

Similarly to the detection efficiency, the neutron die-away time is a system parameter, which is pre-calibrated and used as an input when solving the inverse problem. One way to calibrate the die-away time is via the so-called Feynman-Y plot. Through algebraic manipulations on the first two moments in Eq. (2), it can be shown that

$$\frac{\text{Var}(T)}{E(T)} - 1 = Y_\infty \left( 1 - \frac{1 - e^{-\lambda T}}{\lambda T} \right). \quad (15)$$

The term  $\frac{\text{Var}(T)}{E(T)} - 1$  is often referred to as the Feynman-Y plot (see [16] and the reference there within). Fig. 2 shows the Feynman-Y plot as sampled for sample No. 2 in three configurations: no shielding, small and large cups. As shown, the plots are very different, since  $Y_\infty$  is proportional to the detection efficiency and depends also on the detector's dead time [17,18]. However, when fitting to the exponential model in (15), all plots give the same value of  $1/\lambda = 50 \pm 2$  microseconds. The same results are observed for all samples. Thus, it can be determined that the system die-away time does not change throughout the different experiments.

#### 5.3.2. Effect on the leakage multiplication factor

The fact that the leakage multiplication factor  $M_L$  might change, by itself, does not pose any problem. When solving the equations,  $M_L$  is a property of the sample that is calculated. A problem, however, might occur if the moderator in the sample causes a deviation from two basic model assumptions: First, the single energy group model, and second,

**Table A.1**

The full results of the shielded measurements — Mean, Variance, Skewness, Kurtosis,  $S$ ,  $U$ ,  $M_L$ ,  $Q_m$ ,  $Q_c$  and  $\mathcal{D}$ , for the 18 measurements described in Section 5.

Measured Sample	Mean	Variance	Skewness	Kurtosis	$S$	$U$	$M_L$	$Q_m$	$Q_c$	$\mathcal{D}$ (%)
Sample No.1, plain (May 2012)	0.2218	0.2851	0.4514	1.179	3010	0.2911	1.021	0.0321	0.0283	11
Sample No.1, large cup (May 2012)	0.1359	0.1571	0.2072	0.4084	2103	0.1932	0.979	0.0039	-0.0003	108
Sample No.1, plain (Nov. 2014)	0.2291	0.2971	0.4777	1.273	3076	0.2984	1.026	0.0359	0.0345	5
Sample No.1, small cup (Nov. 2014)	0.2034	0.2559	0.3915	0.9736	2839	0.2675	1.014	0.0225	0.0186	17
Sample No.1, large cup (Nov. 2014)	0.1386	0.1612	0.2150	0.4306	2161	0.201	0.982	0.0041	0.0003	91
Sample No.1, lead cup (Nov. 2014)	0.2287	0.2966	0.4771	1.2785	3075	0.295	1.027	0.0418	0.0355	15
Sample No.2, plain (May 2012)	0.1641	0.2080	0.3252	0.7984	2283	0.2584	1.025	0.0227	0.0222	2
Sample No.2, small cup (May 2012)	0.1479	0.1824	0.2412	0.6232	2127	0.2360	1.012	0.0152	0.0115	24
Sample No.2, large cup (May 2012)	0.1012	0.1159	0.1510	0.2808	1590	0.1724	0.982	0.0022	0.0003	87
Sample No.3, plain (May 2012)	0.0760	0.1031	0.1745	0.4145	939	0.402	1.023	0.0137	0.0127	7
Sample No.3, small cup (May 2012)	0.0683	0.0896	0.1441	0.3220	884	0.360	1.010	0.0091	0.0065	28
Sample No.3, large cup (May 2012)	0.0462	0.0552	0.0767	0.1404	682	0.2409	0.981	0.0011	0.0001	85
Sample No.3, plain (Nov. 2014)	0.0764	0.1043	0.1779	0.4248	944	0.4235	1.026	0.0139	0.0134	3
Sample No.3, large cup (Nov. 2014)	0.0465	0.0562	0.0795	0.1491	684	0.2646	0.983	0.0022	0.0003	87
Sample No.4, plain (May 2012)	0.0393	0.0511	0.0824	0.1820	525	0.3036	1.026	0.0061	0.0059	1
Sample No.4, small cup (May 2012)	0.0351	0.0443	0.0680	0.1410	487	0.2756	1.012	0.0038	0.0031	18
Sample No.5, plain (Nov. 2014)	0.2107	0.3251	0.6255	1.813	1916	0.8582	1.023	0.0555	0.0521	6
Sample No.5, small cup (Nov. 2014)	0.1874	0.2815	0.5272	1.4745	1826	0.7481	1.020	0.0458	0.039	16

the assumption that the duration of the fission chains is sufficiently small (allowing the use of the Bohnel formula for the generalized factorial moments). To verify that in the present study both assumptions are still valid, all the shielded measurements are re-analyzed but with the real efficiency (which, as stated, can be estimated by comparing the count rates). In all the experiments, the estimated leakage multiplication in the shielded experiments is similar to that in the non-shielded experiments within less than 0.5% difference (and in all samples,  $M_L$  is between 1.022 and 1.029).

## 6. Conclusions

A new method for detecting a shielding of the sample in NMC is introduced. The detection is done by detection of a decrease in the detection efficiency, translated into a discrepancy between two different realizations of  $Q$ .

## References

The method is implemented and validated in two settings. First, the samples are not shielded, but the count rate is reduced artificially. Second, the samples are covered by a polyethylene or lead cup, creating a reduction in the count rate. In the first setting, results indicate, that a 15% discrepancy between  $Q_c$  and  $Q_m$  is a clear indicator of a neutron shielding, and a clear distinction between a shielded and a non-shielded measurement appears once the decrease in the count rate is 20% and more. In the second setting, which is more similar to an actual shielding, results indicate that even a 5% shielding can be detected. One possible reason for the difference between the results is that the second setting was only measured using a PCMS counter, which has a very high efficiency to begin with (54%).

One fact that should be mentioned is that all the samples analyzed in this study are fairly small, containing up to 6.6 g of plutonium. Thus, further examination of the method on larger masses is necessary to complete the validation of the method.

Since the method does not require any change of the system configuration and can be easily implemented on any existing facility (assuming that the data acquisition is done in LIST mode), it can serve as an effective tool to determine whether additional measurements are required using the “Add-A-Source” method.

## Appendix. Full results for all experiments

See Table A.1.

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