Nuclear Inst. and Methods in Physics Research, A 902 (2018) 83–87

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Mass uncertainty in neutron multiplicity counting associated with the uncertainty on the fission multiplicity factorial moments

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ARTICLE INFO

Keywords:
Neutron multiplicity counting
Nondestructive special nuclear materials assay
Uncertainty propagation

ABSTRACT

Passive neutron multiplicity counting relies on measurement of the spontaneous fission neutron yield, to estimate the amount of ^{240}Pu in the tested sample. To account for additional neutron sources in the sample, typically (α, n) reactions and induced fissions in the odd fissionable isotopes, the first three sampled moments of the neutron count distribution are used in an inversion formula that quantifies the amplitude of all three neutron sources. When solving the set of equations corresponding to the inversion formula, the first three factorial moments of the fission multiplicity distribution (both the spontaneous and induced fission) are used. Thus, any uncertainty on the nuclear data and the numeric values of the neutron multiplicity moments, is bound to create a parametric uncertainty on the estimated mass. So far, most studies on the uncertainty associated with the nuclear data are experimental by nature, often focusing on a better estimation of the factorial moments and a viable uncertainty estimation on the reported values.

Since the inversion formula is non-linear, the error propagation from the multiplicity moments to the mass is also non-linear, and might have a very strong dependence on the sample parameters. In the present study, we formulate mathematical formulas that describe the error propagation from the factorial moments of the fission multiplicity to the mass, and implement the formulas to quantify the uncertainty in terms of the sample characteristics. For validation, the computational results are then compared with experimental results.

1. Introduction

Passive Neutron Multiplicity Counting (NMC) relies on measurement of the spontaneous fission neutron yield, to estimate the amount of ^{240}Pu in the tested sample. Due to the relative transparency of structure materials to neutron flux, NMC methods have been proven to be a very efficient for nondestructive analysis of poorly characterized impure samples. To account for additional neutron sources in the sample, typically (α, n) reactions and induced fissions in the odd fissionable isotopes, the first three sampled moments of the neutron count distribution are used in an inversion formula that quantifies the amplitude of all three neutron sources [1]. There are several ways to implement NMC methods, differing in the type of the moments sampled [2]: the most common implementation is multiplicity method, where the so called Singles, Doubles and Triples (together, referred to as the *Multiplicity Moment*) are sampled [1], but equivalent results may be obtained by sampling the central moments of the count distribution [3,4].

Using the Multiplicity method, the sampled moments (Singles-S, Doubles-D and Triples-T) are connected with sample parameters through the following set of equations:

$$\begin{cases} S = F \times P_d \times M_L \left(U(D_{S,1} - 1) + 1 \right) M_L \\ D = F \times P_d^2 \times f_d M_L^2 \left(U D_{S,2} + \dfrac{M_L - 1}{1 - D_{I,1}} (U(D_{S,1} - 1) + 1) D_{I,2} \right) \\ T = F \times P_d^3 \times f_t M_L^3 (U D_{S,3} + \dfrac{M_L - 1}{1 - D_{I,1}} (3U D_{S,2} D_{I,2} \\ \quad + D_{I,3}(U(D_{I,1} - 1) + 1)) \\ \quad + \left(\dfrac{M_L - 1}{1 - D_{I,1}} \right)^2 D_{I,2}^2 (U(D_{S,1} - 1) + 1)) \end{cases} \tag{1.1}$$

The parameters in Eq. (1.1) are as follows: the experimentally measured observables are S, D and T. The three unknowns are the total source rate F, the spontaneous fission fraction U and the leakage multiplication factor M_L. The set of equations is defined by several external parameters: the detection efficiency P_d, the Doubles and Triples

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https://doi.org/10.1016/j.nima.2018.06.008
Received 22 February 2018; Received in revised form 3 June 2018; Accepted 4 June 2018

gate utilization factors f_d and f_t and finally the first three factorial moments of the spontaneous fission multiplicity $D_{S,i}, i = 1,2,3$, and the first three factorial moments of the induced fission multiplicity $D_{I,i}, i = 1,2,3$. Once the set of equations is solved, the spontaneous fission rate is given by $F \times U$, and the mass of ^{240}Pu effective is estimated by dividing it with the spontaneous fission rate per gram of ^{240}Pu, 473.5 [fissions/sec] [1].

The leakage multiplication factor, which quantifies the contribution of the induced fissions, is defined by $M_L = \frac{1-P_f}{1-D_{I,i}P_f}$, where P_f is the fission probability of a neutron in the sample [5]. Since P_f it is a more natural quantity, we will hereon refer to P_f rather than M_L.

Since they appear in the set of Eq. (1.1), any biasing on the numeric values of the neutron multiplicity moments is bound to bias the estimated mass of ^{240}Pu. This observation is obviously not new, and the mass uncertainty was treated before. However, most studies on the uncertainty associated with the nuclear data are experimental by nature, often focusing on a better estimation of the factorial moments and a viable uncertainty estimation on the reported values [6,7]. Since the inversion formula is non-linear, the error propagation from the multiplicity moments to the mass is also non-linear, and might have a very strong dependence on the sample parameters. In [8,9] a more integrated approach is considered, by numerical and experimental sensitivity analysis of NMC measurement. While both studies offer a good overview on the related uncertainties, we believe that the error propagation related to nuclear data was not treated in a complete fashion: both offer a short discussion on the effect of uncertainty of the nuclear data (see section 3.3 [8]), but the conclusion is fairly obscure, claiming that the uncertainty can be included in the uncertainty on the detection efficiency.

The outline of the present study is to develop explicit formulas for the uncertainty propagation from the nuclear data, manifested in the factorial moments of the fission neutron multiplicity distribution, on to the final outcome of the measurement, the estimated mass of ^{240}Pu.

2. Mathematical formulas for the error propagation of the factorial moments

2.1. The uncertainty associated with each factorial moment

The set of Eqs. (1.1) can be formally written as a set of three non linear equation of the form:

$$\begin{cases} F_1(S,U,P_f|D_{S,i},D_{I,i}, i=1,2,3) = S \\ F_2(S,U,P_f|D_{S,i},D_{I,i}, i=1,2,3) = D \\ F_3(S,U,P_f|D_{S,i},D_{I,i}, i=1,2,3) = T \end{cases} \tag{2.2}$$

For fixed values of S, T and D, the solution of the set of equation defines an implicit inverse function

$$\begin{cases} U = U(D_{S,i},D_{I,i}, i=1,2,3) \\ F = F(D_{S,i},D_{I,i}, i=1,2,3) \\ D = D(D_{S,i},D_{I,i}, i=1,2,3) \end{cases} \tag{2.3}$$

If we denote by $\Delta D_{S,i}, (\Delta D_{I,i}), i = 1,2,3$ the uncertainty related to the spontaneous (induced) fission multiplicity factorial moments, than the uncertainty on each of the parameters F, U and P_f associated with each factorial moment can be estimated by:

$$\Delta_{D_{X,i}} F = \left| \frac{\partial F}{\partial D_{X,i}} \Delta D_{X,i} \right|, \quad \Delta_{D_{X,i}} U = \left| \frac{\partial U}{\partial D_{X,i}} \Delta D_{X,i} \right|,$$
$$\Delta_{D_{X,i}} P_f = \left| \frac{\partial P_f}{\partial D_{X,i}} \Delta D_{X,i} \right|, \tag{2.4}$$

for $X = S, I$ and $i = 1,2,3$.

The estimations in (2.4) are of little use in their present form, since F, U and P_f are implicit functions of factorial moments, and the derivatives are not known. To obtain the derivatives in an explicit form, there is a standard methodology, which we will introduce shortly. We will only demonstrate it for one factorial moment, $D_{S,1}$, as the implementation is exactly the same for the rest.

By derivation of all three equation in (2.2) with respect to $D_{S,1}$, assuming the S, U and P_f are functions of $D_{S,1}$, we obtain

$$\frac{\partial F_i}{\partial F}\frac{\partial F}{\partial D_{S,1}} + \frac{\partial F_i}{\partial U}\frac{\partial U}{\partial D_{S,1}} + \frac{\partial F_i}{\partial P_f}\frac{\partial P_f}{\partial D_{S,1}} + \frac{\partial F_i}{\partial D_{S,1}} = 0 \quad i = 1,2,3$$

And hence the partial derivatives of S, U and P_f are determined by:

$$\begin{pmatrix} \frac{\partial F}{\partial D_{S,1}} \\ \frac{\partial U}{\partial D_{S,1}} \\ \frac{\partial P_f}{\partial D_{S,1}} \end{pmatrix} = (-1) \begin{pmatrix} \frac{\partial F_1}{\partial F} & \frac{\partial F_1}{\partial U} & \frac{\partial F_1}{\partial P_f} \\ \frac{\partial F_2}{\partial F} & \frac{\partial F_2}{\partial U} & \frac{\partial F_2}{\partial P_f} \\ \frac{\partial F_3}{\partial F} & \frac{\partial F_3}{\partial U} & \frac{\partial F_3}{\partial P_f} \end{pmatrix}^{-1} \begin{pmatrix} \frac{\partial F_1}{\partial D_{S,1}} \\ \frac{\partial F_2}{\partial D_{S,1}} \\ \frac{\partial F_3}{\partial D_{S,1}} \end{pmatrix} \tag{2.5}$$

Equality (2.5) describes the relation between the implicit derivatives of U, F and P_f with respect to $D_{S,1}$ and the explicit derivatives of $F_i (i = 1,2,3)$ with respect to F, U, P_f and $D_{S,1}$. Before we continue, some remarks are due:

1. The coefficient matrix $A = \begin{pmatrix} \frac{\partial F_1}{\partial F} & \frac{\partial F_1}{\partial U} & \frac{\partial F_1}{\partial P_f} \\ \frac{\partial F_2}{\partial F} & \frac{\partial F_2}{\partial U} & \frac{\partial F_2}{\partial P_f} \\ \frac{\partial F_3}{\partial F} & \frac{\partial F_3}{\partial U} & \frac{\partial F_3}{\partial P_f} \end{pmatrix}$, which defines the set of equations, is independent (explicitly) of $D_{S,1}$. Therefore, when writing the set of equations for any other factorial moment, the coefficient matrix A will not change.
2. When solving a set of equations, there is always the question whether or not the coefficient matrix is invertible. While we did not prove that it is always invertible, from a practical point view point, it is fair to assume that it is, for three reasons: first, the conditions for A to be invertible are exactly the conditions for a unique solution to the set of equations, which is always assumed. Second, the conditions for A to be invertible are extremely generic (for instance, the determinant must not be 0). Third, practice shows that it is, eventually, invertible.
3. Once $\Delta_{D_{X,i}} F$ and $\Delta_{D_{X,i}} U$ are estimated ($X = S, I$ and $i = 1,2,3$), we estimate the uncertainty on the mass by
$$\Delta_{D_{X,i}}(Mass) = \frac{\Delta_{D_{X,i}} F \times U + \Delta_{D_{X,i}} U \times F}{472.5} \tag{2.6}$$
4. At a first glance, it might seem that the uncertainty does not depend on the sampled moments S, T and D. The uncertainty has an implicit dependence on S, T and D, since the values of F, U and P_f are determined by S, T and D.

2.2. How to sum uncertainties?

Once the uncertainty associated with each factorial moment in quantified, we encounter a question that often arises in uncertainty quantification: how do we accumulate all the uncertainties into a single error bar?

One of the most common approaches for accumulation of errors is through a simple sum of squares. That is, for each variable V ($V = F, U$ or P_f), we estimate:

$$\Delta V = \sqrt{\sum_{j=1}^{3} \left(\Delta_{D_{S,j}} V\right)^2 + \sum_{j=1}^{3} \left(\Delta_{D_{I,j}} V\right)^2} \tag{2.7}$$

But the use of Eq. (2.7) is questionable for two reasons. First, the uncertainty on the factorial moments is not a statistical uncertainty, but rather a constant biasing from the true value. Second, Eq. (2.7) assumes that the error in all factorial moments is not correlated, which is clearly not the case. To account for the correlation between the moments we need information on the covariance, which (to the best of our knowledge) is unavailable.