The number distribution of neutrons and gammas generated in a multiplying sample

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Abstract

The subject of this paper is an analytical derivation of the full probability distribution of the number of neutrons and photons generated in a sample with internal multiplication by one internal source emission event, and its comparison with Monte Carlo calculations. We derive recursive analytic expressions for the probability distributions $P(n)$ of neutrons and photons up to values of n for which $P(n)$ is significant, as functions of the first collision probability p of the source neutrons. The derivation was performed by using the symbolic algebra code MATHEMATICA. With the introduction of a modified factorial moment of the number of fission neutrons and photons, the resulting expressions were brought to a formally equivalent form with those for the factorial moments of the searched probability distributions. The results were compared with Monte Carlo calculations, and excellent agreement was found between the analytical results and the simulations. The results show that the probability distributions change with increasing sample mass such that the "bulk" of the distribution changes only slightly, but a tail develops for higher n values, which is the main reason for the increase of the factorial moments with increasing sample mass.

KEYWORDS: Nuclear safeguards, materials control and accounting, neutron and photon numbers, number distributions, generating functions, master equations, multiplicity

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1. Introduction

The statistics of the number of neutrons and photons generated in a finite sample with a spontaneous internal source and multiplication has been studied in the past both with analytical methods and by Monte-Carlo simulations. These studies are motivated by the fact that the knowledge of the statistics of the generation of neutrons and photons makes it possible to determine, by coincidence and multiplicity measurements, the mass and isotopic composition of an

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unknown sample. The number distribution of generated or emitted neutrons and photons will be different from that of the elementary spontaneous fission events if the sample is multiplying, since the source neutrons can create short chains through induced fission events, thus affecting the distribution of both the neutron and photon numbers.

Regarding the theory, closed form implicit equations were derived for the probability generating functions (PGFs) of the distributions of neutrons [1–3], and later that of the photons [4]. A full explicit solution of these equations for the PGFs is not possible, due to their strongly non-linear character. However equations can be derived for the factorial moments of different orders which are always linear in the highest order moment. These equations can be solved recursively. In principle, these calculations only require calculations of the derivatives of an implicit function, and algebraic rearrangements, hence they can be performed explicitly up to arbitrarily high orders. However, the complexity of the algebraic equations grow very fast. So far explicit solutions were given only for the first three factorial moments of the neutrons and photons. Higher moments are of no practical interest since they are usually not measured, so there was no motivation for the involved calculation of the higher order moments.

The probabilities $P(n)$ and $F(n)$ of emitting n neutrons or gamma photons, respectively, can be calculated with a recursive procedure similar to that of deriving the factorial moments. However, unlike the latter, it is of interest to determine the $P(n)$ and $F(n)$ for all n values for which they are significant so that the cumulative probability is close to unity, that is

$$\sum_{n=0}^{N} P(n) \geq 0.99$$

and similarly for $F(n)$. The values of N fulfilling the above can be up to 100 or larger for fissile samples of considerable size (kg amounts).

In this paper we present a methodology to determine the probability distributions from the corresponding master equation, in a recursive manner for increasing n, up to large n values. Even if the procedure of generating algebraic equations for the $P(n)$ and $F(n)$ is rather straightforward, since these include nth order derivatives of an implicit nested equation, the corresponding equations soon become exceedingly complicated. We have therefore used the symbolic algebra code MATHEMATICA [5] in the calculations. However, the non-trivial character of the calculations is true even when calculating the derivatives by symbolic algebra codes.

The methodology we used in this work was to organise the symbolic calculations such that in the recursive calculations of the higher order moments, the expressions of the (already determined) lower order moments were kept symbolic. This way the fast inflation of the formulae by the order n did not paralize the symbolic manipulations. Likewise, in the numerical work, the evaluation of the expressions was made in an upwind manner, so that the lower order moments entered the highest one, to be calculated in the actual step, only by their quantitative value. By this procedure a full analytic and numerical determination of the full probability distributions was possible.

The work had the unexpected side effect that by solving for the probability distributions, it turned out that we also have solved the problem of determining high order factorial moments as well in the analytical work (and hence could do the job of numerical evaluations as well). Namely, with the introduction of some modified factorial moments of the number of neutrons and photons generated in spontaneous and induced fission, the expressions for the $P(n)$ or $F(n)$ (i.e., the distribution of the neutrons and photons generated in the sample), for all n

values can be written in a form that is formally equivalent with the nth factorial moment of the same distributions. By replacing the above-mentioned modified moments with the real factorial moments of the fission neutron and gamma distributions, and including some factorials, $P(n)$ and $F(n)$ can be converted into factorial moments of the total number of neutrons and photons respectively. This means that with trivial changes, if the distributions can be calculated, their factorial moments can also be calculated with the same MATHEMATICA algorithm up to large orders.

2. General principles

Master equations for the generating functions of the number of neutrons and photons in a sample with both spontaneous and induced fission have been derived in references [1, 4]. In both cases, it is assumed that the probability p of a neutron's first collision before escaping from the sample is known. In the present work, similarly to the previous works, absorption will be neglected; hence, p will be equal to the probability of induced fission per neutron. For this reason, the quantity we calculate corresponds to the distribution of the neutrons (or photons) *generated* in the sample and we will avoid talking about the neutrons *emitted* from the sample. Correspondingly, this means that the probability for a neutron to escape from the system without inducing fission is $(1-p)$. Taking into account absorption and the detection process in the analytical model is relatively easy and will be addressed in future communications. In the following, we will describe first the procedure to determine the distribution of neutrons in the present (absorption-free) model and then determine the same distribution for the gamma photons.

2.1 Neutron distributions

Let $p_s(n)$ and $p_f(n)$ denote the probability distributions of generating n neutrons in a spontaneous fission event (=source event) and in an induced fission event, respectively. Their generating functions $q_s(z)$ and $q_f(z)$ are defined as

$$q_s(z) = \sum_n p_s(n) z^n \quad , \quad q_f(z) = \sum_n p_f(n) z^n. \tag{1}$$

Further, let $p_1(n)$ denote the probability distribution of the number of neutrons generated in the sample by *one* initial neutron. In the same manner, let $P(n)$ denote the probability distribution of neutrons generated in the sample from one initial *source* event. The source event can be either spontaneous fission, or an (α, n) process, or a combination of the two; all these can be treated with a formally completely identical formalism.

The generating functions $h(z)$ and $H(z)$ of $p_1(n)$ and $P(n)$, respectively, are defined as

$$h(z) = \sum_n p_1(n) z^n \quad \text{and} \quad H(z) = \sum_n P(n) z^n. \tag{2}$$

As shown in [1] and [4], the following coupled master equations can be obtained for the generating functions $h(z)$ and $H(z)$:

$$h(z) = (1-p)z + p q_f[h(z)] \tag{3}$$

and

$$H(z) = q_s[h(z)]. \tag{4}$$

From the above we shall derive the distribution $P(n)$. It can be obtained from Eqs. (3) and (4) by noting that $p_1(n)$ and $P(n)$ are the Taylor expansion coefficients of $h(z)$ and $H(z)$, respectively; that is,

$$p_1(n) = \frac{1}{n!}\frac{d^n h(z)}{dz^n}\bigg|_{z=0} \quad \text{and} \quad P(n) = \frac{1}{n!}\frac{d^n H(z)}{dz^n}\bigg|_{z=0}. \tag{5}$$

As shown in Eqs. (3) and (4), calculation of the derivatives of $H(z)$ requires derivatives of the generating function $h(z)$, that is derivations of the implicitly given function $q_f[h(z)]$ on the r.h.s. of Eq. (3). It can be seen that higher-order derivatives will contain algebraic combinations of the lower-order ones; hence, they need to be calculated recursively by starting from the lower-order ones. For the case $n = 0$, calculation of $p_1(0)$ requires just inserting $z = 0$ into Eqs. (3) and (2). This leads to an Nth-order algebraic equation, where N is the maximum number of neutrons generated in induced fission; that is,

$$p_f(n) = 0 \quad \text{for} \ n > N.$$

Hence one has

$$p_1(0) = (1-p)z + pq_f[h(z)]\big|_{z=0} = pq_f[p_1(0)] = p\sum_{n=0}^{N} p_f(n)[p_1(0)]^n \tag{6}$$

The value of N is given by the probability distributions of fission neutron numbers, which are nuclear physics constants. In the present work we will limit N to 8, based on the data available for the isotope Pu-240 that we will use in the quantitative work. Hence we end up with an 8th-degree polynomial in $p_1(0)$ where the probabilities $p_f(n)$, $n = 0, 1...8$ for the numbers of neutrons emitted from an induced fission event appear as coefficients. Incidentally, these data are also implemented in some Monte Carlo codes, which are capable of calculating higher-order moments of the distribution of the neutrons from numerical simulation. The code MCNP-PoliMi [6], which was used in the quantitative investigations in this paper, has the capacity of handling the full statistics of neutron and gamma transport. We used the fission neutron data from this code for the evaluation of the analytical model.

The first step in the calculations is thus to solve the polynomial equation (6) for $p_1(0)$, which can be done numerically. In addition to the distribution $p_f(n)$, the solution will depend also on the value p of the probability of inducing fission. Once $p_1(0)$ is known, the source-induced probability, $P(0)$, can be obtained from Eq. (4) as

$$P(0) = \sum_{n=0}^{8} p_s(n) \cdot p_1(0)^n. \tag{7}$$

The higher-order terms, in contrast to the determination of $p_1(0)$, do not require the solution of higher-order algebraic equations, only linear ones. For the quantity $P(1)$, from Eq. (5), we obtain

$$P(1) = \frac{1}{1}\frac{dH(z)}{dz}\bigg|_{z=0} = \frac{dq_s(h)}{dh}\frac{dh(z)}{dz}\bigg|_{z=0}. \tag{8}$$

From Eq. (3) one obtains,

$$\frac{dh(z)}{dz} = (1-p) + p\,\frac{dq_f(h)}{dh}\frac{dh(z)}{dz},$$

with the solution

$$\frac{dh(z)}{dz} = \frac{(1-p)}{\left(1 - p\frac{dq_f(h)}{dh}\right)}. \tag{9}$$

For the quantity $\frac{dq_f(h)}{dh}$ we will have

$$\left.\frac{dq_f(h)}{dh}\right|_{z=0} = \left.\sum_n n p_f(n)[h(z)]^{n-1}\right|_{z=0} = \sum_{n=0}^{8} n \cdot p_f(n)[p_1(0)]^{n-1}. \tag{10}$$

If Eq. (10) were to be evaluated at $z = 1$, such as in the case of the moments, then instead of $p_1(0)$, one would have unity, and the r.h.s. of (10) would reduce to the first factorial moment $\langle \nu_f \rangle$ of the number of neutrons generated in induced fission. In our case, the expression is different, and instead of an expected value, we have an expectation "weighted" by $[p_1(0)]^{n-1}$. To simplify notations, as an analogue to $\langle \nu_f \rangle$, which is a shorthand notation for a weighted sum (the expectation value), we shall introduce a similar shorthand notation for the type of weighted sum in Eq. (10) with an overbar notation (i.e. as $\overline{\nu}_f$). Then, in the general case, we will have

$$\left.\frac{d^n q_f(h)}{dh^n}\right|_{z=0} = \left. q_f^{(n)}(h)\right|_{z=0} = \overline{\nu_f(\nu_f - 1)\ldots(\nu_f - n + 1)}. \tag{11}$$

Similar notations will be used for spontaneous fission, that is the weighted moments of $q_s^{(n)}(z)$. Using this notation, we obtain for the term $P(1)$ the explicit expression

$$P(1) = \left.\frac{dq_s(h)}{dh}\frac{dh(z)}{dz}\right|_{z=0} =$$

$$= \left[\sum_{n=0}^{8} n \cdot p_s(n)[p_1(0)]^n\right] \left(\frac{(1-p)}{1 - p\sum_{n=0}^{8} n \cdot p_f(n)[p_1(0)]^n}\right) = \overline{\nu_s}\frac{(1-p)}{1 - p\overline{\nu_f}} \tag{12}$$

One can see that with the introduction of the "modified" expected values $\overline{\nu_f}$ and $\overline{\nu_s}$, the expression for $P(1)$ is formally equivalent with the expected value $\widetilde{\nu}$ of the number of neutrons (singlets) generated in one source emission event [1,4], the difference being that $\overline{\nu_f}$ and $\overline{\nu_s}$ replace $\langle \nu_f \rangle$ and $\langle \nu_s \rangle$. The dependence of $P(1)$ on the non-leakage probability p is though more complicated than that of $\widetilde{\nu}$ since, unlike $\langle \nu_f \rangle$ and $\langle \nu_s \rangle$, which are nuclear physics constants, $\overline{\nu_f}$ and $\overline{\nu_s}$ also depend on p.

The calculation of the higher-order terms proceeds in a similar manner. For $P(2)$, after some algebra, one obtains the expression

$$P(2) = \frac{1}{2}\left(\frac{1-p}{1-p\overline{\nu_f}}\right)^2 \left[\overline{\nu_s(\nu_s - 1)} + \frac{p}{1-p\overline{\nu_f}}\overline{\nu_s}\overline{\nu_f(\nu_f-1)}\right]. \tag{13}$$

Again, with the exception of the coefficient $1/2!$ and the difference in the definition of the expectations of ν_s and ν_f, the expression for $P(2)$ is formally identical to that of the second factorial moment $\widetilde{\nu}(\widetilde{\nu} - 1)$ of the neutrons generated in the sample by one source emission event, that is, the doublets [1,4]. This equivalence can be easily shown to be generally true from the definition of the factorial moments and the corresponding terms of the probability distribution; that is, one has

$$P(n)|_{\overline{\nu}_f,\overline{\nu}_s} = \frac{1}{n!}\left.\langle \widetilde{\nu}(\widetilde{\nu}-1)...(\widetilde{\nu}-n+1)\rangle\right|_{\langle \nu_f \rangle, \langle \nu_s \rangle} \qquad n = 1, 2... \tag{14}$$

This means that, with certain straightforward changes, the solution obtained for the distribution $P(n)$ contains also the solution for the factorial moments.

The problem of calculating the higher-order terms up to large n values remains to be solved. The general nested higher-order derivatives needed to calculate $H(z)$ and $h(z)$ will become more and more cumbersome for increasing n, hence these were calculated by the symbolic manipulation code MATHEMATICA [5]. In this case the use of a recursive formula that can be symbolically handled by MATHEMATICA becomes very helpful. Namely, the nth derivative contains all lower-order derivatives, so to be able to evaluate it for a certain n, which is needed to find $p_1(n)$, we need to calculate and evaluate all lower-order derivatives. Those are, on the other hand, already calculated in previous steps of the calculation and need not be calculated again.

A significant advantage of this methodology is that it serves an explicit expression with all parameters free (unspecified). The parameters can then easily be assigned numerical values later. This procedure proved to be fully feasible, and no problems were encountered with calculating explicit expressions (in a nested form) up to $n = 50$ for neutrons, and even higher values for gamma photons.

2.2 Gamma distributions

For gammas one can derive a similar set of coupled master equations for the PGFs based on arguments similar to those for the neutrons. For reasons mentioned earlier, here again we have to deal with coupled equations for the distribution of the gammas emitted from (generated in) the sample, induced by one initial neutron, and the distribution of gammas induced by one initial source emission event. This leads to [4]

$$g(z) = (1-p) + pr_f(z)q_f[g(z)] \tag{15}$$

and

$$G(z) = r_s(z)q_s[g(z)], \tag{16}$$

with $q_f(z)$ and $q_s(z)$ given earlier. Here we also introduced $f_s(n)$ and $f_f(n)$, the number distribution of the number of gammas produced in one spontaneous and one induced fission event by source neutrons, respectively, and their PGFs as

$$r_f(z) = \sum_n f_f(n)z^n \quad , \quad r_s(z) = \sum_n f_s(n)z^n. \tag{17}$$

These are again known nuclear parameters, and for the quantitative work in the present paper, we obtained their values from MCNP-PoliMi. Further, we used the obvious notations for the generating functions of the number distributions that are the main subject of interest as

$$g(z) = \sum_n f_1(n)z^n \quad , \quad G(z) = \sum_n F(n)z^n. \tag{18}$$

Again we can identify the $f_1(n)$ and $F(n)$ as Taylor expansion coefficients of $g(z)$ and $G(z)$, respectively:

$$f_1(n) = \frac{1}{n!}\frac{d^n g(z)}{dz^n}\bigg|_{z=0}, \quad \text{and} \quad F(n) = \frac{1}{n!}\frac{d^n G(z)}{dz^n}\bigg|_{z=0}. \tag{19}$$

The procedure is similar to that for neutrons, and we only summarize the main steps here. For the probability $f_1(0)$ we obtain from Eq. (15) an algebraic equation for $f_1(0)$ as

$$f_1(0) = (1-p) + p\, r_f(0)\, q_f[f_1(0)] = (1-p) + p\, r_f(0) \sum_{n=0}^{8} p_f(n)[f_1(0)]^n. \tag{20}$$

It is seen that for the probability $f_1(0)$, we end up with an 8th-degree polynomial, as in the case for neutrons, despite the fact that the gamma multiplicity is greater than the neutron multiplicity. However, the order of the equation is determined by the branching (multiplication), which is only associated with the fission process, since the gammas do not multiply.

Having determined $f_1(0)$ from Eq. (16), one readily obtains $F(0)$ as

$$F(0) = f_s(0)\, q_s[f_1(0)] = f_s(0) \sum_{n=0}^{8} p_s(n)[f_1(0)]^n. \tag{21}$$

Continuing to higher-order terms, we need to calculate the derivatives of $g(z)$ and $G(z)$. To this end, as Eqs. (15) and (16) show, we need to calculate the derivatives of the functions $q_\alpha(g)$ and $r_\alpha(z)$, $\alpha = \{s, f\}$ and evaluate them at $z = 0$. Again, it is practical to introduce shorthand notations for the occurring weighted sum expressions, as in the case of the neutrons, and to bring out the formal equivalence between the $F(n)$ and the corresponding factorial moments of order n, denoted as $\langle\tilde{\mu}(\tilde{\mu}-1)...(\tilde{\mu}-m+1)\rangle$ in [4]. Following the nomenclature there, statistical quantities (moments) of gammas induced by a single neutron will be denoted by μ, and those for a source emission effect by $\tilde{\mu}$.

According to the above, we introduce the notations

$$\left.\frac{d^n r_\alpha(z)}{dz^n}\right|_{z=0} = n!\, f_\alpha(n) \equiv \widetilde{\mu}_\alpha(n); \qquad \alpha = s, f; \tag{22}$$

and

$$\left.\frac{d^n q_\alpha(g)}{dg^n}\right|_{z=0} = \sum_m m(m-1)\dots(m-n+1)\cdot\, p_\alpha(n)[f_1(0)]^{m-n} \equiv$$

$$\widetilde{\nu_\alpha(\nu_\alpha-1)\dots(\nu_\alpha-n+1)} \equiv \widetilde{\nu_\alpha}(n); \qquad \alpha = s, f. \tag{23}$$

In Eq. (23) we have introduced a further condensation of the notations as compared to Eq. (11). We also note that despite the formal similarities between the definitions (11) and (23), the quantities $\overline{\nu_f(\nu_f-1)\dots(\nu_f-n+1)}$ and $\widetilde{\nu_f}(n)$ are not equal, since in the former the weight function is $p_1(0)$, whereas in the latter it is $f_1(0)$.

With the help of the definitions $\widetilde{\nu}_f(n)$, $\widetilde{\nu}_s(n)$, $\widetilde{\mu}_f(n)$, and $\widetilde{\mu}_s(n)$, we can start calculating the $F(n)$ such that they become formally equivalent to the corresponding factorial moments, just as we did for the neutron distributions. For illustration, we give below results for the terms $n = 1$ and $n = 2$. These read as

$$F(1) = r_s'(z)\, q_s[g(z)] + r_s(z)\, q_s'[g(z)]g'(z)|_{z=0} =$$

$$= \widetilde{\mu_s}(1)\,\widetilde{\nu_s}(0) + \widetilde{\mu_s}(0)\,\widetilde{\nu_s}(1)\,\frac{p\,\widetilde{\mu_f}(1)\,\widetilde{\nu_f}(0)}{1-p\,\widetilde{\mu_f}(0)\,\widetilde{\nu_f}(1)}. \tag{24}$$

and

$$F(2) = \frac{1}{2!}\Big[\ \ \widetilde{\mu}_s(2)\,\widetilde{\nu}_s(0) + 2\,\widetilde{\mu}_s(1)\,\widetilde{\nu}_s(1)\,f_1(1) + \widetilde{\mu}_s(0)\,\widetilde{\nu}_s(2)\ [f_1(1)]^2 + \\ +\widetilde{\mu}_s(0)\,\widetilde{\nu}_s(1)\ \cdot 2!f_1(2)\Big] \quad (25)$$

It can be shown that with the substitutions described earlier, the r.h.s. of Eqs. (24) and (25)are equivalent to the first and second factorial moments (singlets and doublets) of gamma photons, given in Eq. (9) of Ref. [4].

The higher-order terms $F(n)$ can be calculated according to this scheme by using symbolic computation. Due to the higher multiplicity of fission gamma photons compared to neutrons, for the sample masses considered in this paper (they are the same as those used in [4]), one has to calculate up to about 100 terms in order to cover 99% of the probability distribution. If higher accountancy of the total probability is desired, the number of required terms increases. The procedure of analytical calculations works well up to this order, and the obtained formulae are also stable for quantitative evaluation. This will be illustrated in the next section.

3. Numerical work

The distributions derived analytically for the neutrons and gamma photons were evaluated numerically and compared to the results of Monte Carlo simulations using MCNP-PoliMi. MCNP-PoliMi can tally the number of spontaneous and induced neutrons and gamma photons in a given Monte Carlo history. The use of PoliMi to calculate the factorial moments, coincidences, and number distributions is described in [4, 6, 7].

3.1 Neutron distributions

The recursive analytical formulae were evaluated quantitatively by using numerical values of the spontaneous and induced fission and the non-escape probability p. The nuclear constants were taken from the code MCNP-PoliMi, and the probability p from the calculations with the same code, for three different masses of a spherical plutonium metal sample having composition 80wt% Pu-239 and 20 wt% Pu-240.

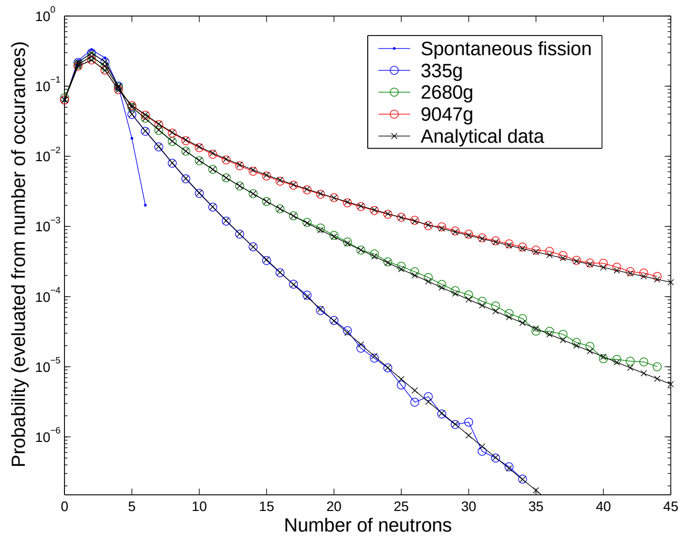
Figure 1 shows a comparison between the numerical data from Monte Carlo simulations and the results from the analytical calculations for three different sample masses and corresponding first-collision probabilities. It can be seen that there is a very good quantitative agreement between the Monte Carlo and the analytical data. The figure shows that with increasing sample mass, and hence increasing non-leakage probability p, the number distributions change in such a way that the bulk of the distribution for low n values remains the same with some decrease of the amplitude, whereas a tail develops for large n values, which is the main reason for the increase of the factorial moments with increasing p.

The phenomenon of a tail developing in the probability distribution for higher numbers of n is a general characteristic of the statistics of Markov chains with renewal and regeneration. Higher p only means that the Markov chain has a “longer life”.

3.2 Gamma distributions

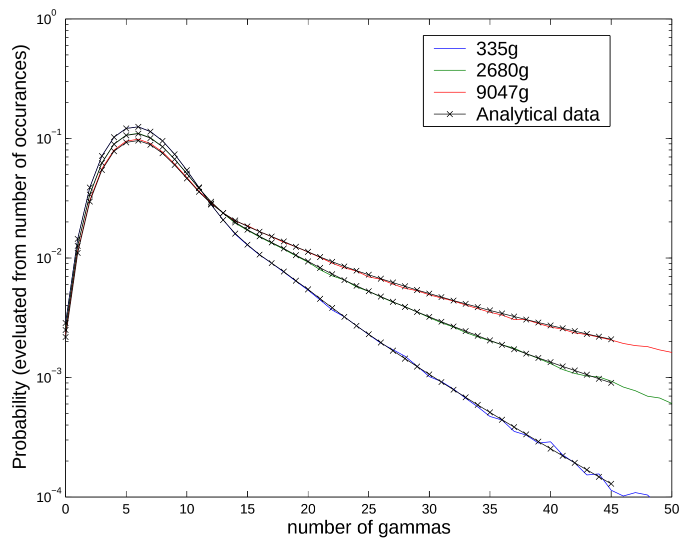
As mentioned in [4], the number distributions of gamma photons is interesting due to their higher fission multiplicities, which hence promises an increased sensitivity in measurements. This potential advantage might be enhanced by the higher penetration power of gamma photons in certain types of shielding material.

Figure 1: Comparison of the analytical results with Monte Carlo calculations for three different sample masses. Spontaneous fission is shown for reference.



The calculations of the statistics of gammas are more involved because they require considerably longer running times, and depend on more kinds of nuclear data. Spontaneous and induced fission gamma distributions, as nuclear input parameters, need to be considered up to $n = 23$.

Figure 2: Comparison between the analytical model and Monte Carlo simulations for the gamma photons.



The results from analytical calculations for photons were compared to those from Monte Carlo simulations, as shown in Fig. 2. The findings are qualitatively very similar to those for neutrons. The analytical and Monte Carlo results agree very well, and the distributions develop a tail for high photon numbers with increasing p.

4. Conclusions

It was demonstrated that by proper application of symbolic computation, high-order terms of the probability distributions of the number of neutrons and photons emitted in fission can be obtained. This technique supplies numerically stable data for very high-orders of these distributions. The calculations were performed in a recursive way such that the lower order distributions are handled symbolically while computing the current highest order. In addition, by introducing certain modified moments of the fission neutron and gamma distributions, we could bring the resulting expressions for the number distributions and the factorial moments into an equivalent form, whereby the solutions obtained for the distributions contain also those for the factorial moments.

The quantitative results show a good agreement with Monte Carlo simulations performed with the code MCNP-PoliMi [4]. The results confirm the general tendency that can be expected intuitively and which was seen in Monte Carlo simulations earlier, namely, that a tail develops with increasing first-collision probability for higher neutron or photon numbers.

This model and the applied analytical technique can be extended to the case of including absorption of both the neutrons and the gammas, as well as the statistics of detection. These will be included in the model and the calculations in the next stage of this work.

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