Calculation of Gamma Multiplicities in a Multiplying Sample for the Assay of Nuclear Materials

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ABSTRACT

The multiplicities, or factorial moments, of the distribution of the number of neutrons emerging from a fissile sample can be used to identify and quantify fissile isotopes, in particular even-N isotopes of transuranic elements. In fact, the spontaneously emitted source neutrons can induce further fissions in the sample, thereby changing the number distributions of the neutrons leaving the sample, and therefore their multiplicities. The multiplicities increase monotonically with sample mass, hence the measurement of the multiplicities can be used to quantify the sample mass.

Analytical expressions for multiplicities that include induced fission effects have been derived for neutrons in the past. These expressions are given as functions of the probability of induced fission per neutron, and have been investigated both by Monte-Carlo methods and in experiments using thermal neutron detectors. The object of this paper is to derive analytical formulae for the multiplicities of the gamma photons emitted by both spontaneous and induced fissions, and to perform a quantitative analysis. In addition, neutron and gamma multiplicities are calculated by Monte-Carlo simulation using a modified version of the MCNP-PoliMi code. Good agreement is found between the analytical formulae and the Monte-Carlo results. The results show the potential advantage of using gamma multiplicities when compared to neutron multiplicities: their higher quantitative values may, in principle, have the effect of leading to a larger sensitivity on the sample mass when compared to the analysis based on neutrons alone.

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1. INTRODUCTION

Traditional nuclear safeguards measurements make use of helium-3 counters to detect neutrons emitted by fissile materials and to measure their multiplicities. The technique is based on the fact that fission emits multiple neutrons essentially at the same time. The characteristics of the sample, for example the sample's mass, can then be inferred by measuring the neutron multiplicity within a specified time window.

In a fissile sample, neutrons from the spontaneous fission and/or from (α,n) reactions, induce fissions and generate, in most cases, some very short-lived chains. Due to the small size of the sample, the time of neutron generation in these short chains can be taken as close to simultaneous with the time of the original source emission. It follows that the induced fissions can be interpreted as a correction to the original multiplicity distribution of the source. The extent of the correction depends on the probability of induced fission by one source neutron, and hence on the mass of the sample. This physical property leads to the possibility of estimating the sample mass from multiplicity measurements. In the past, the dependence of the multiplicities of neutrons emitted from a sample on the sample mass, and on the related probability of induced fission, was analytically derived [1-4]. This analysis shows analogies to the fast fission factor of the classical four-factor formula, which describes the increase of the mean number of primary fast neutrons from thermal fission due to neutrons emitted by fast fissions induced by the primary neutrons. The fast fission factor is thus a clear analogue of the singlets, i.e. the first factorial moment of the number of neutrons emitted from a multiplying sample per source neutron emission event.

More recently, investigations have been aimed at the use of organic scintillating detectors in a wide range of applications in nuclear nonproliferation, international safeguards, nuclear material control and accountability, and national security. One of the advantages of the use of this type of detector is that they are sensitive to fast neutrons, so there is no need to thermalize the neutrons from the spontaneous and induced fissions in the samples. Moreover, these detectors are sensitive to gamma rays, which are emitted by the fission process with a larger multiplicity than neutrons. Use of this type of detector makes possible the measurement of both neutron and gamma ray multiplicities from a fissile sample. The analysis of such measurements provides the motivation for the present work.

The present paper is aimed at the development of an analytical model for the description of the distribution of the number of neutrons and photons in a multiplying medium. In the analysis, we only consider the theoretical multiplicities due to one source neutron emission event. Hence we disregard the fact that the intensity of the source in a sample with spontaneously fissioning isotopes depends linearly on the sample mass, leading thus to a quadratic or higher dependence of the multiplicities on the sample mass. In this respect, the treatment is similar to the derivations given by Böhnel [4] and Lu and Teichmann [5-6], but is extended to include the description of the photon statistics. The formalism is fully capable to treat joint neutron-photon coincidences or multiplicities. These, however, will be treated in a later communication, together with other generalizations and extensions. It can be mentioned that the formalism, without quantitative analysis, has been used by Oberer to treat both neutron, gamma, and neutron-gamma joint distributions [7]. For the sake of comparison with photon multiplicities, the results from Ref. [4] will be quoted without derivation in this work.

2. DERIVATION OF THE MOMENTS OF THE GAMMA DISTRIBUTION

We will derive an expression for the distribution of the number of gamma rays generated in a multiplying fissile sample *per initial source neutron event.* Because no internal absorption of the gamma photons in the sample is assumed, this distribution is equivalent to the distribution of the gamma photons escaping from the sample. Internal absorption, of course, would change the statistics in the opposite direction than internal fission. This effect will be investigated in subsequent work. Also, due to the strong energy dependence of the gamma capture cross sections, the present treatment needs to be extended to allow for the energy dependence of capture, similarly to the treatment used in [4] for the spectral effects of neutron reactions.

What regards escape from the sample, the situation is different for the neutrons, because neutrons causing an internal fission are lost for emission from the sample. Hence it is worth to note that the distribution and the multiplicities of neutrons that will be cited in this work refer to neutrons generated in all the fissions, minus the neutrons causing the fissions. This tally is clearly not equal to the number of neutrons that escape from the sample, because some of the neutrons are captured in the sample. This effect will be treated in a consecutive communication. Here it suffices to say that the model and the MC calculations are compatible, and both are unaffected by the presence of absorption in the sample. In the derivations, we will use the method of probability generating functions (PGFs), which were first used by Böhnel [4] to calculate neutron multiplicities. In this treatment, the initial source neutron event can be a spontaneous fission or an (α, n) reaction, or a combination of the two [4].

From the analytical model for the gammas, we shall derive the first three factorial moments, and compare them to the known results for the neutrons. There are several different notations in use for the factorial moments, such as singlets or reals (S), doublets (D) and triplets (T), to denote the first, second, and third factorial moment, respectively. Here, however, we shall use the traditional notations of the expectations with brackets, as used in references [4] – [6].

The formalism is based on the use of the generating functions of the probability distributions concerned, and a few simple rules giving the generating functions of composite processes in terms of the generating functions of the elementary process. We need some definitions for these distributions and their generating functions, and these are listed in following sub-sections. In Section 2.1 we list some nuclear parameters of the distribution of neutrons that are independent of the sample size and geometry. In Section 2.2 we give the corresponding distributions for the gamma rays and describe how the gamma ray distributions depend on the neutron distributions in a multiplying sample. Finally, in Section 2.3 we give the first three moments of the neutron and gamma ray distributions.

2.1 Neutron Distributions

In the present model, all gamma photons in the sample are assumed to be generated by neutron induced fissions, so we give here the neutron parameters and distributions that were used in previous works [4] for the determination of neutron multiplicities. It is also implicit in the model that gamma photons are only generated in the fissile material of interest, and not in the matrix of compounds or in possible impurities. These restrictions can be readily relaxed, and this will be made in subsequent work. In the following list of definitions, we shall try to use notations for neutrons as compatible as possible with previous publications, and will introduce a full set of notations for the gamma distributions.

<u>Initial source event</u>
$p_s(n)$ is the probability of emitting *n* neutrons per source event, and

$q_s(z) = \sum_{n=0}^{\infty} p_s(n) z^n$ is the PGF of $p_s(n)$. The factorial moments of $p_s(n)$ are denoted as ν_s, $\langle \nu_s(\nu_s - 1) \rangle$, and so on.

<u>Induced fission</u>
$p(n)$ is the probability of generating *n* neutrons per induced fission event, and

$q(z) = \sum_{n=0}^{\infty} p(n) z^n$ is the PGF of $p(n)$. The factorial moments of this function are denoted as ν, $\langle \nu(\nu - 1) \rangle$, and so on.

2.2 Gamma Distributions

<u>a) Nuclear properties at individual reactions (nuclear parameters, independent of the sample size and geometry)</u>
<u>Initial source event</u>
$f_s(n)$ is the probability of emitting *n* gammas per source event, and

$r_s(z) = \sum_{n=0}^{\infty} f_s(n) z^n$ is the PGF of $f_s(n)$. The factorial moments of $f_s(n)$ are denoted as μ_s, $\langle \mu_s(\mu_s - 1) \rangle$, and so on.

<u>Gammas from induced fission</u>
$f(n)$ is the probability of emitting *n* gammas per induced fission event, and

$r(z) = \sum_{n=0}^{\infty} f(n) z^n$ is the probability generating function (PGF) of $f(n)$.
The factorial moments of $f(n)$ are denoted as μ, $\langle \mu(\mu - 1) \rangle$, and so on.

All the factorial moments mentioned above can be obtained by deriving the generating functions and evaluating them for $z = 1$.

b) Properties that depend on the multiplication of neutrons in the sample

p is the probability that a neutron born in the sample (either as a source neutron or as a neutron in the chain) will induce a fission before it leaves the sample. We will refer to this quantity as the fission probability.

$f_1(n)$ is the probability of generating a total of *n* gammas in all the fissions in the sample initiated by *a single neutron,* irrespective of whether it is a source neutron or a neutron in the chain. As mentioned before, this the same as the distribution of gammas leaving the sample, because internal absorption of the gammas is neglected.

The process is Markovian: a neutron's past cannot influence how many more gammas the neutron will generate in the future. This probability is therefore the same for all fission neutrons, irrespective of which generation they belong to in a chain.

$g(z) = \sum_{n=0}^{\infty} f_1(n) z^n$ is the PGF of $f_1(n)$.

$F(n)$ is the probability distribution of the total number of gammas generated by (and hence also emitted from the sample) one source event. This is the main quantity of interest.

$G(z) = \sum_{n=0}^{\infty} F(n) z^n$ is the PGF of $F(n)$.

The moments of *F*, which can be obtained from the derivatives of *G*, are the other quantities of interest: $\tilde{\mu}$, $\langle \tilde{\mu}(\tilde{\mu}-1) \rangle$, and so on. These will be investigated quantitatively as functions of the fission probability *p*.

The derivation of the equations for the probability distributions and their generating functions then goes like this. For the number distribution of gammas generated by one neutron already existing in the system, one has the master equation

$$f_1(n) = (1-p)\cdot\delta_{n,0} + p \sum_{\substack{k=1 \\ n_0+n_1+n_2+....+n_k=n}}^{\infty} f(n_0) p(k) \prod_{i=1}^{k} f_1(n_i)\,. \tag{1}$$

In a similar way, the distribution of gammas leaving the sample due to one source event (spontaneous fission) can be written as

$$F(n) = \sum_{\substack{k=1 \\ n_0+n_1+n_2+....+n_k=n}}^{\infty} f_s(n_0) p_s(k) \prod_{i=1}^{k} f_1(n_i)\,. \tag{2}$$

The derivation of the above formulae follows the standard procedure of deriving first collision type master equations. Hence the probability $f_1(n)$ in (1) is constructed as the sum of the probabilities of the two mutually exclusive events: the neutron either leaves the sample without any collisions (fissions) and hence generates no gammas (first term on the r.h.s. of Eq. (1)), or it will have a first fission that will possibly lead to a chain (second term on the r.h.s of Eq. (1)). In this latter case, n_0 gammas are produced with probability $f(n_0)$ by the first fission, and a number of new neutrons are generated that can in turn contribute to the gamma generation process through later fissions, with the same distribution $f_1(n)$. Here again we have to sum up for the mutually exclusive events that the fission will lead to 1, 2, etc. new neutrons, with probability $p(k)$, and we have to take into account that the fate (fissions with gamma generation) of each of these k new neutrons is independent of the fate of the others, hence the k-fold product of the $f_1(n)$-s. We also have to ensure that the sum of all later gammas in the chain, *plus the gammas generated in the initiating fission,* will equal n gammas, hence the constraint $n_0 + n_1 + n_2 + ... + n_k = n$.

This constraint is of the type of a convolution, and hence turning to the generating functions according to the definitions in subsections 2.1 and 2.2 (i.e. by multiplying both sides of the equations with z^n and summing up), will lead to a simplified form of the equations for the generating functions g and G as follows:

$$g(z) = (1-p) + pr(z)\sum_{k=0}^{\infty} p(k)\, g^k(z) = (1-p) + p\, r(z)\, q[g(z)], \qquad (3)$$

and

$$G(z) = r_s(z)\, q_s[g(z)]. \qquad (4)$$

Here, the g(z) in the square brackets is an argument of the generating functions q and q_s, hence both expressions are implicit functions of z. Thus, for example

$$q[g(z)]'\big|_{z=1} = q'\big|_{z=1}\, g'(z=1) = \nu g', \qquad (5)$$

and so on.

It should be noted here that the generation of gamma rays from interactions other than fission is not included in this formulation. Gamma rays from interactions such as inelastic scattering and absorption will be included in a further communication. The formulae for the generating functions could also be directly derived by following some simple rules for the generating functions of the distributions of the sums and products of random variables, such as those listed in Ref. [4].

Although the variable *time* does not appear explicitly in the above equations, it is clear that they are master equations for the probability generating functions of the backward type (first collision type). This is a consequence of the causality sequence taken into account in the derivation, which starts with considering one neutron and derives equations for the distributions of its progeny. The forward approach would be equivalent to considering one final neutron, and trying to derive the probability of generating such a neutron from the history of its ancestors. Such an approach is not feasible for the multiplicity problem. It can be worth mentioning that there are several other problems in which only the backward equation can be used, such as the theory of the development of random trees [8].

Eqs. (3) and (4) are implicit equations in g and G, and they cannot be solved explicitly. However, this is not necessary either, because the factorial moments can be obtained by derivation and by solving the algebraic equations that arise. The highest order derivative can always be expressed explicitly in terms of the (already known) lower order derivatives, starting with the first order ones (the singlets).

2.3 Moments of the Neutron and Gamma Ray Distributions

We have calculated the few lowest order moments, out of which the first three (singlets, doublets and triplets) will be given and analyzed here, and compared with their neutron multiplicity counterparts. As mentioned earlier, the multiplicities below refer to the neutrons and gammas leaving the sample, just as in [4] for the neutrons.

First moments (singlets)

<u>Neutrons</u> (Ref. [4])

$$\tilde{\nu} = \nu_s \frac{1-p}{1-p\nu} \tag{6}$$

<u>Gammas</u>: Derivation of (3) and (4) yields

$$\tilde{\mu} = \mu_s + \frac{\nu_s \cdot p \cdot \mu}{1-p\nu} \tag{7}$$

Second moments (doublets)

<u>Neutrons</u> (Ref. [4])

$$\left\langle \tilde{\nu}\left(\tilde{\nu}-1\right)\right\rangle = \left(\frac{1-p}{1-p\nu}\right)^2 \left\{\left\langle \nu_s\left(\nu_s-1\right)\right\rangle + \frac{p}{1-p\nu}\nu_s\left\langle \nu\left(\nu-1\right)\right\rangle\right\} \tag{8}$$

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<u>Photons</u>

$$\left\langle \tilde{\mu}\left(\tilde{\mu}-1\right)\right\rangle = \left\langle \mu_s\left(\mu_s-1\right)\right\rangle + \frac{2\mu_s\nu_s p\mu}{1-p\nu} + \left\langle \nu_s\left(\nu_s-1\right)\right\rangle\left(\frac{p\mu}{1-p\nu}\right)^2 + \frac{\nu_s p}{1-p\nu}\left\{\left\langle \mu\left(\mu-1\right)\right\rangle + 2\mu\nu\frac{p\mu}{1-p\nu} + \left\langle \nu\left(\nu-1\right)\right\rangle\left(\frac{p\mu}{1-p\nu}\right)^2\right\} \tag{9}$$

Third moments (triplets)

<u>Neutrons</u> (Ref. [4])

$$\left\langle \tilde{\nu}\left(\tilde{\nu}-1\right)\left(\tilde{\nu}-2\right)\right\rangle = \left(\frac{1-p}{1-p\nu}\right)^{3} \left\{ \left\langle \nu_s\left(\nu_s-1\right)\left(\nu_s-2\right)\right\rangle + \frac{p}{1-p\nu}[3\left\langle \nu_s(\nu_s-1)\right\rangle \left\langle \nu\left(\nu-1\right)\right\rangle + \right. \\ \left. +\nu_s\left\langle \nu\left(\nu-1\right)\left(\nu-2\right)\right\rangle\right] + 3\left(\frac{p}{1-p\nu}\right)^{2} \nu_s\left\langle \nu\left(\nu-1\right)\right\rangle^{2} \right\} \tag{10}$$

Photons

Here we need a nested formulation. Define

$$g'' = \frac{p}{1-p\nu}\left[\left\langle \mu(\mu-1)\right\rangle + 2\mu\nu\left(\frac{p\mu}{1-p\nu}\right) + \left\langle \nu(\nu-1)\right\rangle\left(\frac{p\mu}{1-p\nu}\right)^{2}\right] \tag{11}$$

$$g''' = \frac{p}{1-p\nu}\left\{ \begin{aligned} &\left\langle \mu(\mu-1)(\mu-2)\right\rangle + 3\left\langle \mu(\mu-1)\right\rangle\nu\left(\frac{p\mu}{1-p\nu}\right) + 3\mu\left[\left\langle \nu(\nu-1)\right\rangle\left(\frac{p\mu}{1-p\nu}\right)^{2} + \nu g''\right] + \\ &+\left\langle \nu(\nu-1)(\nu-2)\right\rangle\left(\frac{p\mu}{1-p\nu}\right)^{3} + 3\left\langle \nu(\nu-1)\right\rangle\left(\frac{p\mu}{1-p\nu}\right) g'' \end{aligned} \right\} \tag{12}$$

With these, one has

$$\begin{aligned} \left\langle \tilde{\mu}\left(\tilde{\mu}-1\right)\left(\tilde{\mu}-2\right)\right\rangle &= \left\langle \mu_s\left(\mu_s-1\right)\left(\mu_s-2\right)\right\rangle + 3\left\langle \mu_s\left(\mu_s-1\right)\right\rangle \nu_s\left(\frac{p\mu}{1-p\nu}\right) + \\ &+3\mu_s\left[\left\langle \nu_s\left(\nu_s-1\right)\right\rangle\left(\frac{p\mu}{1-p\nu}\right)^{2} + \nu_s g''\right] + \left\langle \nu_s\left(\nu_s-1\right)\left(\nu_s-2\right)\right\rangle\left(\frac{p\mu}{1-p\nu}\right)^{3} + \\ &+3\left\langle \nu_s\left(\nu_s-1\right)\right\rangle g'' + \nu_s g''' \end{aligned} \tag{13}$$

Higher order moments can be derived in a completely analogous manner. Their length and complication, however, grows very quickly with the moment order. They will therefore not be given here, nor will they be investigated quantitatively.

3. NUMERICAL WORK AND COMPARISON WITH MONTE CARLO

A small modification to the MCNP-PoliMi code [9] was performed to tally the number of spontaneous and induced fission neutrons and photons in a given Monte Carlo history. Each history was initiated by a Pu-240 spontaneous fission event. The tally consisted in summing the neutrons born in the spontaneous fission, and in all subsequent induced fissions in a given history. An analogous procedure was applied to the photons. Because the tally was performed at the time of fission, the subsequent fate of the neutrons or photons did not affect the value of the tally.

The simulations were performed for three plutonium metal spheres, with composition 20wt%

Pu-240 and 80wt% Pu-239 and mass equal to 0.33, 2.7, and 9 kg. The cases are denoted as 1, 2 and 3, respectively. The calculated distributions of total, source, and induced neutrons and photons per history are shown in Figs. 1.a and 1.b, respectively, for the 9 kg plutonium sample.

The first three factorial moments of the distributions of the number of neutrons and photons generated in the fissile samples were calculated from the tallies, and were compared to the moments obtained from the analytical model of Eqs. (6) through (13). In the case of the distribution of the number of neutrons, the number of induced fissions in each history was subtracted from the total number of neutrons born in the sample. This procedure is necessary to account for the loss of one neutron at each induced fission. The fission probability used in the analytical formulae, p, was estimated in the Monte Carlo calculations by taking the ratio of the total number of induced fissions in the sample to the total number of neutrons in the sample. The resulting predictions are given in Tables I - III.

In the evaluation of the analytical formulae, the moments of the number of neutrons and gamma rays, which are physical constants, were obtained from the Monte Carlo [9]. The numerical values are shown in Table IV for the first three moments of the distributions of the number of neutron and gamma rays from spontaneous and induced fission. In the 9 kg plutonium sample, 85% of the induced fissions occurred in the Pu-239, and the remaining 15% in the Pu-240, approximately.

There is reasonably good quantitative agreement between the predictions of the analytical model and the Monte Carlo calculations for the first two factorial moments in all three samples. For the third moments, the deviations between the model and the Monte Carlo values are large for the third sample, but are reasonably good for the smaller samples. The difference between the model and the Monte Carlo in the largest sample may be attributed to the simplifications of the analytical model. These simplifications lead to a greater discrepancy as the moment's order increases. The results also show that the values of all factorial moments are much greater for the gammas than for the neutrons. This is a result of the higher multiplicity of the gammas for both the source emissions and the induced fission reactions.

From the analytical formulae, the dependence of the factorial moments on the fission probability p can be easily calculated. The results are shown in Figs. 2 through 4. For sake of comparison, the values for neutrons are also shown. The figures show that the values of the first three moments of the distributions, i.e. singlet, doublet and triplet rates, increase faster with increasing p for the gamma multiplicities than for the neutron multiplicities. This means that in principle, measuring the gamma photon multiplicities is a more sensitive method for estimating the sample mass and multiplication than measuring the neutron multiplicities. It has to be mentioned here that the detection efficiency of the gammas and neutrons was not accounted for in the present study. However, in the energy range of interest, the detection efficiency of gammas is comparable to that of neutrons in the organic scintillators that will be used to perform the multiplicity measurements [10]. The effect of detection efficiency on the multiplicity will be investigated in a future communication.

CONCLUSIONS

An analytical model for the calculation of gamma multiplicities in a fissile sample was developed by using the method of probability generating functions. The proposed approach is similar to earlier works regarding neutron multiplicities. Equations were derived for the generating functions of the gamma distributions from a single neutron in the sample and from the intrinsic source, and analytical expressions were given explicitly for the first three moments of the distribution of the number of gamma rays. Understandably, these formulae are more complicated for the gamma multiplicities than for the neutron multiplicities, especially for the higher moments. The equations relate the values of the moments to a number of nuclear parameters (such as the moments of the distributions of neutrons and photons born by induced fission), and to the fission probability p, which depends on the multiplication properties of the sample. This latter quantity is in turn related to the multiplication factor (k-eff) of the sample, which is one of the primary quantities of interest in these measurements.

The results show good quantitative agreement between the analytical formulation and the Monte Carlo simulations of three plutonium metal samples of realistic size. The quantitative results show that the values of all three moments are much greater for gamma rays than for neutrons, and that they are more sensitive functions of the fission probability p, and hence of the sample characteristics, for gamma rays than for neutrons. This means a potentially higher efficiency in discovering, identifying, and quantifying unknown samples of fissile material by measuring gamma multiplicities than by measuring neutron multiplicities. To explore this potential for practical applications, the model has to be extended to include all neutron and gamma reactions including capture, the detection process, and the dependence of the source intensity on the sample mass. These extensions are underway, and will be reported on in later communications.

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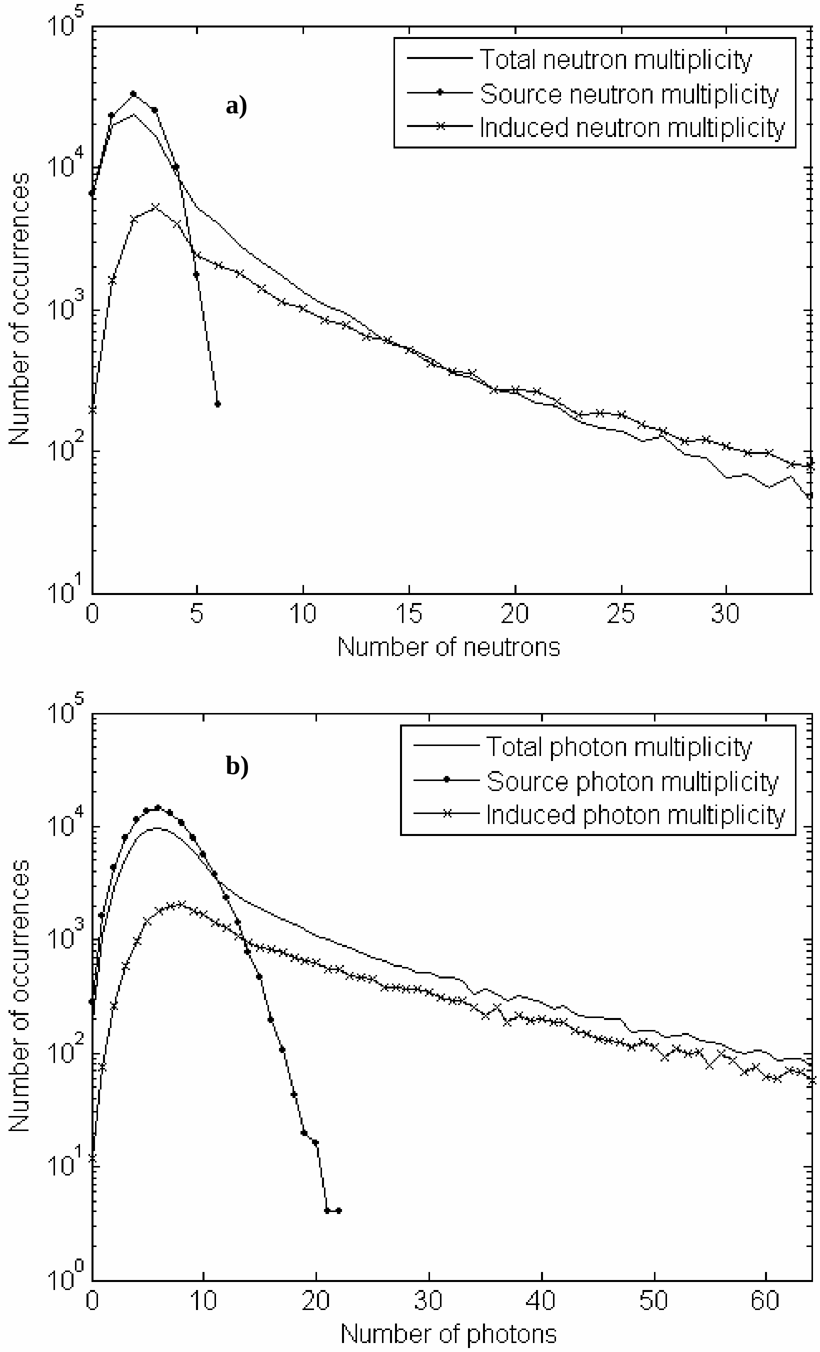
Figure 1.



a)
Total neutron multiplicity
Source neutron multiplicity
Induced neutron multiplicity
Number of occurrences
Number of neutrons
b)
Total photon multiplicity
Source photon multiplicity
Induced photon multiplicity
Number of occurrences
Number of photons

Figure 2

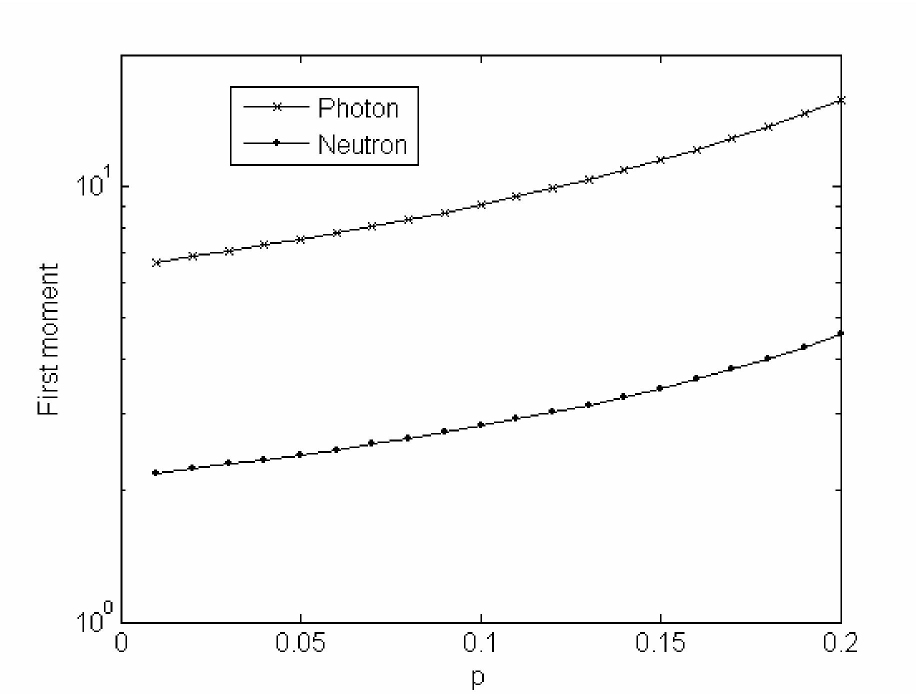


Figure 3

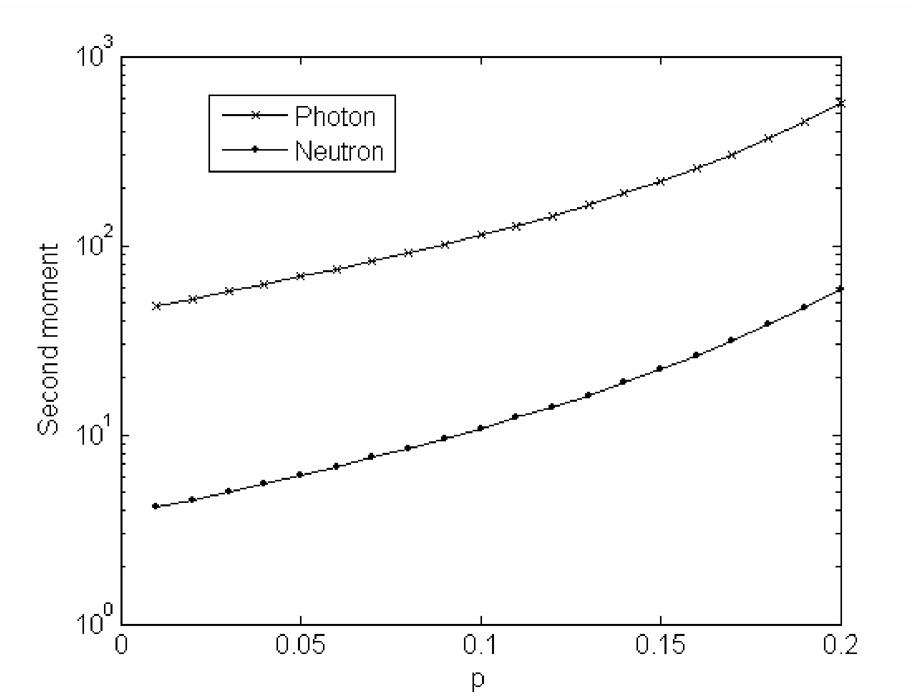


Figure 4

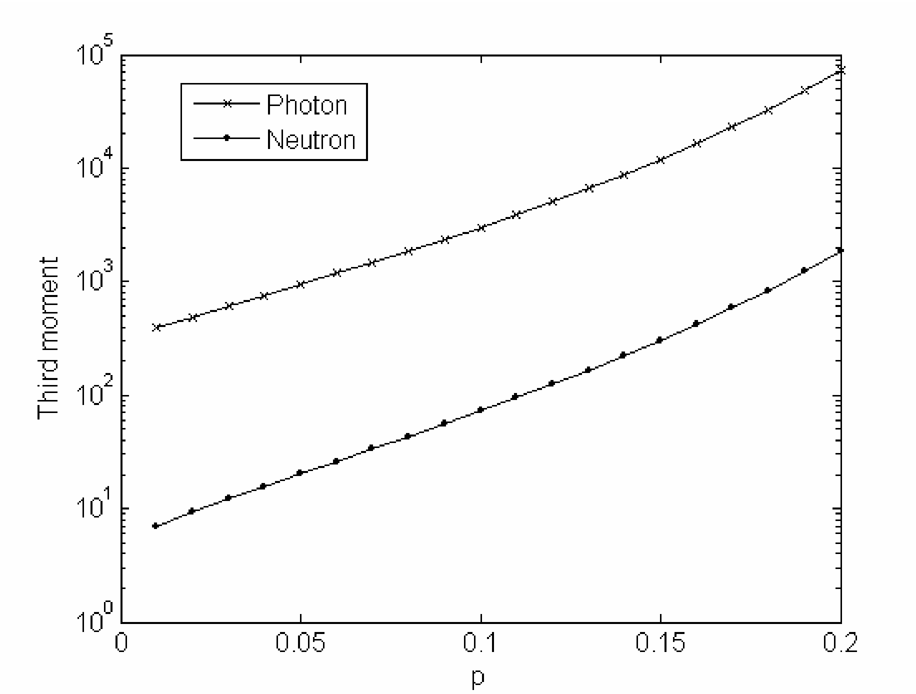

Photon
Neutron
Third moment
p
10^0
10^1
10^2
10^3
10^4
10^5
0
0.05
0.1
0.15
0.2

FIGURE CAPTIONS

Figure 1. Monte Carlo simulation of the distribution of neutrons (a) and gammas (b) in a plutonium sample (9 kg).

Figure 2. Dependence of the first moment of the neutron and the photon distribution on the fission probability *p*.

Figure 3. Dependence of the second moment of the neutron and photon distributions on the fission probability *p*.

Figure 4. Dependence of the third moment of the neutron and photon distributions on the fission probability *p*.

TABLES

Table I

Case	p	$\tilde{\nu}$ according to Eq. (6)	$\tilde{\nu}$ according to Monte Carlo	$\tilde{\mu}$ according to Eq. (7)	$\tilde{\mu}$ according to Monte Carlo
1	0.06388	2.52	2.52	7.83	7.81
2	0.12464	3.09	3.09	10.00	9.93
3	0.18383	4.13	4.06	13.96	13.06

Table II

Case	p	$\langle\tilde{\nu}(\tilde{\nu}-1)\rangle$ according to Eq. (8)	$\langle\tilde{\nu}(\tilde{\nu}-1)\rangle$ according to Monte Carlo	$\langle\tilde{\mu}(\tilde{\mu}-1)\rangle$ according to Eq. (9)	$\langle\tilde{\mu}(\tilde{\mu}-1)\rangle$ according to Monte Carlo
1	0.06388	7.14	7.11	80.6	80.14
2	0.12464	15.21	15.27	176.13	167.66
3	0.18383	41.39	37.18	503.94	342.45

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Table III

Case	p	$\langle\tilde{\nu}(\tilde{\nu}-1)(\tilde{\nu}-2)\rangle$ Eq.(11)	$\langle\tilde{\nu}(\tilde{\nu}-1)(\tilde{\nu}-2)\rangle$ Monte Carlo	$\langle\tilde{\mu}(\tilde{\mu}-1)(\tilde{\mu}-2)\rangle$ Eq.(14)	$\langle\tilde{\mu}(\tilde{\mu}-1)(\tilde{\mu}-2)\rangle$ Monte Carlo
1	0.06388	29.64	29.37	1.28e+003	1.22e+003
2	0.12464	146.50	146.91	5.70e+003	4.87e+003
3	0.18383	973.40	658.14	3.78e+004	1.43e+004

Table IV.

Spontaneous fission		Induced fission	
ν_s	2.15	ν	3.15
μ_s	6.44	μ	8.09
$\langle \nu_s (\nu_s - 1) \rangle$	3.78	$\langle \nu (\nu - 1) \rangle$	8.20
$\langle \mu_s (\mu_s - 1) \rangle$	43.01	$\langle \mu (\mu - 1) \rangle$	68.02
$\langle \nu_s (\nu_s - 1)(\nu_s - 2) \rangle$	5.20	$\langle \nu (\nu - 1)(\nu - 2) \rangle$	17.28
$\langle \mu_s (\mu_s - 1)(\mu_s - 2) \rangle$	299.78	$\langle \mu (\mu - 1)(\mu - 2) \rangle$	593.00

TABLE CAPTIONS

Table I. Comparison of analytical and Monte Carlo first moments of neutron and photon distributions for three Pu metal samples.

Table II. Comparison of analytical and Monte Carlo second moments of neutron and photon distributions for three Pu metal samples.

Table III. Comparison of analytical and Monte Carlo third moments of neutron and photon distributions for three Pu metal samples.

Table IV. First three moments of neutron and gamma ray spontaneous and induced fission distributions.