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ANALYTICAL AND NUMERICAL MODELLING OF THE DETECTION STATISTICS OF EMISSION FROM A FISSILE SAMPLE WITH ABSORPTION

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ABSTRACT

This paper investigates an analytical derivation of the distribution of the number of neutrons and photons emitted by a multiplying sample. The relationship between the statistics of the generated and detected neutrons and photons is also described. The analytical model described in this paper accounts for absorption, thus extending the model presented in previous studies. By using this new, improved model, one can investigate the relative feasibilities of measuring neutrons or gamma photons for the analysis of a specific fissile sample. In fact, larger mass will lead to larger self-shielding for gamma photons, whereas for neutrons a larger mass will lead to increased multiplicities due to an increased probability to induce fission for each neutron, with absorption playing a minor role. The results suggest that although photons have a larger initial (source) multiplication, neutrons might be more favourable to measure in the case of large samples because of the increasing self-shielding effect for gamma photons.

Key Words: nuclear safeguards, neutron and photon numbers, number distribution, master equations

1. INTRODUCTION

In non-destructive assay of nuclear materials, the statistics of the number distribution of neutrons and gamma rays emitted by fissile samples is of high importance. The multiplicities of neutrons [1] and photons [2] generated in fissile samples with internal multiplication have been investigated in the past. Investigations of the effect of including absorption in the model have also recently been made. The starting point is implicit master equations for the generating functions of the neutron and photon number distribution. These have also been used in the past for calculating factorial moments. For practical reasons, factorial moments are usually only interesting up to third or fourth order. Using multiplicity and coincidence measurements, one can deduce the sample mass and isotopic composition of a certain sample.

In contrast to the factorial moments, the probabilities $P(n)$ and $F(n)$ of emitting n neutrons or gamma photons respectively, are interesting up to large values of n. The necessary number of terms of $p(n)$ that needs to be calculated is determined from the condition that the cumulative probability should be sufficiently close to unity. This number can be over 50 for neutrons and photons in some cases.

Including absorption in the model affects the statistics of the number distribution in several ways. For neutrons, the absorption will mean that some neutrons are eliminated from the fission chain and the observable (leaked) neutrons will reduce in number. For photons the dependence is more involved, because both the absorbtion of neutrons and the gamma photons themselves will affect the gamma distribution. All of these effects will vary with the main parameter of the sample being investigated, which is the sample mass. An increase in mass affects the probability of both induced fission, as well as absorption of both neutrons and gamma photons for large samples in the few kg mass range.

The process of detection also affects the way one can observe the statistics of emission from a certain sample. There is always a certain detector efficiency involved in the process of detection. Different ways of modelling this effect were investigated earlier [3]. The model in this latter publication assumes the detectors embedded into the sample. This model does not correspond to the physical situation, hence it leads to results that could be unrealistic. In this work we will present a different way of accounting for absorption and detection, which only concerns detection of the particles that leaked out from the sample. The results of the analytical calculations are compared to those from Monte Carlo simulations.

2. THEORY

Master equations for the generating functions of the number distribution of neutrons and photons have been given in references [1, 2]. In those models, as well as in the numerical simulations to which the model results were compared, the absorption of neutrons and gamma photons was not accounted for. It was assumed that each neutron has a fixed probability p of inducing fission, or leaking out with a probability $1-p$.

The probability generating functions (PGFs) $h(z)$ and $H(z)$ of $p_1(n)$ and $P(n)$, which describe the number distribution of neutrons generated by one initial neutron and one initial neutron event (spontaneous fission) respectively, are defined as

$$h(z) = \sum_n p_1(n) z^n \quad \text{and} \quad H(z) = \sum_n P(n) z^n, \tag{1}$$

respectively. The coupled backward master equations for neutrons read as follows [1]:

$$h(z) = (1-p)z + p q_f[h(z)] \tag{2}$$

and

$$H(z) = q_s[h(z)]. \tag{3}$$

Here,

$$q_s(z) = \sum_n p_s(n) z^n \quad and \quad q_f(z) = \sum_n p_f(n) z^n \tag{4}$$

represent the generating functions of the number of neutrons generated in spontaneous and induced fission events, respectively. The number distributions are obtained by observing the fact that $p_1(n)$ and $P(n)$ are the Taylor expansion coefficients of $h(z)$ and $H(z)$ respectively:

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

The requirement to evaluate the expressions at $z=0$ gives us some complications. If the derivatives were to be evaluated at $z=1$, we would have had factorial moments of the neutrons generated in spontaneous or induced fission; ν_s, ν_f. Now we arrive at modified moments instead that are defined by:

$$\frac{d^n q_\alpha(h)}{dh^n}\bigg|_{z=0} = q_\alpha^{(n)}(h)\Big|_{z=0} = q_\alpha^{(n)}[p_1(0)] = \overline{\nu}_{\alpha n} \quad ; \quad \alpha = s, f, \tag{6}$$

where $p_1(0)$ is the probability of having zero neutrons generated from one initial neutron. This probability needs to be determined since all other terms $p_1(n)$ and $P(n)$ depend in a recursive manner on that factor, and it occurs also in the modified moments. This means that the initial probability of having zero neutrons generated when starting with one initial neutron is vitally important and needs to be calculated to be able to find the other terms in the number distribution. Note can be made also to the fact that this probability will be highly dependant on the mass of the sample, and later, whether or not absorption and detection is included in the model.

The final expressions for the individual terms of $P(n)$ show a formal equivalence to the factorial moments. By using the formulas for the number distribution we can simultaneously also obtain factorial moments to the same order as the value of n.

This same formal equivalence is found for the probability distribution and factorial moments for gamma photons. The probability distribution is given by [4]:

$$g(z) = \sum_n f_1(n) z^n \quad , \quad G(z) = \sum_n F(n) z^n. \tag{7}$$

Where $f_1(n)$ and $F(n)$ describe the number of generated gammas starting with one initial neutron or one initial source event, respectively. The generating functions are defined implicitly from the following equations [2]:

$$g(z) = (1-p) + p r_f(z) q_f[g(z)] \tag{8}$$

and

$$G(z) = r_s(z) q_s[g(z)]. \tag{9}$$

Here $q_s(z)$ and $q_f(z)$ have been defined earlier but we also see the functions $r_s(n)$ and $r_f(n)$, which are the PGFs of the number distribution of photons from one spontaneous or induced fission, respectively:

$$r_f(z) = \sum_n f_f(n) z^n \quad , \quad r_s(z) = \sum_n f_s(n) z^n. \tag{10}$$

Also these nuclear quantities give rise to modified moments for the photons that in this case are defined as follows:

$$\left. \frac{d^n r_\alpha(z)}{dz^n} \right|_{z=0} = n!\, f_\alpha(n) \equiv \overline{\mu}_{\alpha n}; \qquad \alpha = s, f. \tag{11}$$

The neutrons also have modified moments that are different from the ones in the master equations for the neutrons. This is due to the fact that the factor $p_1(0)$ in Eq. (6) will be exchanged for the factor $f_1(0)$ in the case of photons. The statistics of generated particles have been investigated earlier, and the master equations have also been used to find factorial moments. To be able to simulate actual detection statistics from fissile samples we need to include not only absorption into the model but also the detection process which takes place with a certain probability, often referred to as the detection efficiency, ϵ.

2.1. Neutron Distribution

In the neutron probability balance equation, the event of absorption can be included into the fission distribution, because it is formally the same as a fission event with zero neutrons generated. With this in mind, one can include the process of absorption by increasing the first collision probability, p, to a value p' representing the first collision probability, which hence accounts for both absorption and fission. At the same time, one has to appropriately modify (increase) the value of $p_f(0)$, and decrease the other $p_f(n)$ to maintain normalization. The formula to do this reads as follows:

$$\widetilde{p}_f(n) = \frac{p' - p}{p'}\delta_{n,0} + \frac{p}{p'}p_f(n). \tag{12}$$

The first master equation will then read as:

$$h(z) = (1 - p')z + p'\widetilde{q}_f[h(z)], \tag{13}$$

where $\widetilde{q}_f(z)$ is the generating function of the $\widetilde{p}_f(n)$ of Eq. (12). The factor $(1 - p')$ is the probability for a single neutron to leak out from the sample, as opposed to the earlier factor of $(1 - p)$ which was the probability to not induce fission. Since comparisons will be made with Monte Carlo simulations using MCNP-PoliMi [5] we take the probabilities for inducing fission or being absorbed from that code, which contains nuclear data tables.

In a similar way, the process of detection can be added by considering the neutrons that have leaked out already, since it is only these that are available for detection from a sample. By using the above formalism, but using a detection probability ϵ instead, the process of detection can be accounted for by the use of the generating function $\varepsilon(z)$ of the binary probability distribution of the number of neutrons detected per leaked neutron:

$$\varepsilon(z) = \epsilon z + (1 - \epsilon). \tag{14}$$

Here, ϵ is the detector efficiency for neutrons. The new generating functions that also include the detection process are given, from obvious considerations, by:

$$h_d(z) = h[\varepsilon(z)] \quad , \quad H_d(z) = H[\varepsilon(z)]. \tag{15}$$

The derivatives needed for finding factorial moments as well as the statistics change in a simple way

$$\frac{d^n h_d(z)}{dz^n} = \frac{d^n h(z)}{dz^n} \cdot (\epsilon)^n \quad , \quad \frac{d^n H_d(z)}{dz^n} = \frac{d^n H(z)}{dz^n} \cdot (\epsilon)^n \tag{16}$$

For the factorial moments the full change is

$$\widetilde{\nu}_{d,n} = (\epsilon)^n \cdot \widetilde{\nu}_n \tag{17}$$

The reason why these change in a very simple way is that they are evaluated at $z = 1$. For the number distribution on the other hand, we have quantities that are evaluated at $z = 0$ which leads to the modified moments such as $\overline{\nu}_{sn}, \overline{\nu}_{fn}$. These modified moments depend on the $p_1(0)$ for neutrons and $f_1(0)$ for photons. However, as absorption and detection are incorporated, the probabilities of having zero particles emitted after starting with one neutron will change, and thus the modified moments change and therefore also the final probability distribution, too.

As before, the modified neutron moments are given by

$$\left.\frac{d^n q_\alpha(h)}{dh^n}\right|_{z=0} = \left.q_\alpha^{(n)}(h)\right|_{z=0} = q_\alpha^{(n)}[p_1(0)] = \overline{\nu}_{\alpha n} \quad ; \quad \alpha = s, f, \tag{18}$$

where the factors $\widetilde{p}_f(n)$ are used. In the case of detection $p_d(0)$ will replace $p_1(0)$, which is solved from the N-th order polynomial

$$p_d(0) = (1 - p')(1 - \epsilon) + p\,\widetilde{q_f}\,[p_d(0)] = (1 - p')(1 - \epsilon) + p\sum_{n=0}^{N}\widetilde{p_f}(n)[p_d(0)]^n. \tag{19}$$

Using all these properties makes it possible to find the detection statistics from the Taylor expansions:

$$p_d(n) = \frac{1}{n!}\frac{d^n h_d(z)}{dz^n}\bigg|_{z=0} \quad \text{and} \quad P_d(n) = \frac{1}{n!}\frac{d^n H_d(z)}{dz^n}\bigg|_{z=0}. \tag{20}$$

The terms in the probability distributions can now be calculated recursively since the starting master equation is in implicit form. The analytic model now depends on the simple parameters; the probability to induce fission, which is a parameter that increases with mass, and can be calculated from the mass of the sample, provided the density is known; the absorption probability which is also depending on sample size, composition and particle affected; and finally the detection efficiency, which can be changed to reflect what type of detector is used, such as single fast scintillator detectors or large arrays of helium tubes in form of multiplicity counters.

2.2. Photon Distribution

To extend the model for photons to include absorption and detection requires more changes than for neutrons due to the fact that the generation of photons depend on neutrons. Therefore we need to consider the absorption of both the neutrons and photons. For the neutrons this was taken into account by changing the probability to induce fission, p, to p' as well as the probability distribution $\widetilde{p}_f(n)$ and hence $\widetilde{q}_f(z)$. However, for Eq. (8), the situation will be different. Since in this work we shall neglect capture gammas, leakage and absorption of a neutron will both lead to zero generated gamma photons, hence the parameter p in (8) remains that of the probability of inducing fission. The only notable change in the equations for photons when accounting for absorption of the neutrons will be that $q_f(z)$ needs to be substituted with $\widetilde{q}_f(z)$, which treats absorption as an increased probability of generating zero neutrons in a fission.

Gamma absorption will be accounted for by the probability p_L that describes the leakage probability for one single photon, likewise $(1 - p_L)$ is the probability for a created photon to be absorbed and not escape the sample. The gamma capture will be accounted for by an additional master equation that describes the two mutually exclusive events of leaking or not leaking out from the sample for a single photon:

$$l(z) = l_\gamma z + (1 - l_\gamma). \tag{21}$$

Here $l(z)$ is the generating function of the binary probability distribution of gamma photons leaving the sample per initial photon. Due to the simple form of this relationship we can note that the factorial moments for leaked photons are simply the factorial moments for generated photons times a leakage factor:

$$\widetilde{\mu}_{l,n} = (l_\gamma)^n \cdot \widetilde{\mu}_n. \tag{22}$$

Further, the master equations for the leaked out photons are given as:

$$g_l(z) = g[l(z)] \quad , \quad G_l(z) = G[l(z)]. \tag{23}$$

The next step in the simulation of the statistics obtained from measurements is to incorporate the process of detection. Detection only concerns the leaked out particles, which thus can be taken into account in the master equations by adding an extra equation that describes the probability to undergo detection or not:

$$\varepsilon_\gamma(z) = \epsilon_\gamma z + (1 - \epsilon_\gamma). \tag{24}$$

Using this equation we can obtain the detection statistics as:

$$g_d(z) = g[l\{\varepsilon(z)\}] \quad , \quad G_d(z) = G[l\{\varepsilon(z)\}]. \tag{25}$$

Since the probability distribution requires derivations to be evaluated at $z = 0$, modified moments are created once again. They have earlier depended on $f_1(0)$ which is the probability of generating zero photons when starting with one initial neutron. Now these modified moments will depend on the new statistics of $f_d(0)$, i.e., the probability of having zero photons detected when starting with one initial neutron in the sample. The quantity $f_d(0)$ is calculated as the root of a finite degree polynomial:

$$f_d(0) = (1-p) + p\, r_f[l\{\varepsilon(0)\}]\, q_f[f_d(0)] = (1-p) + p \left(\sum_{n=0}^{\sim 24} f_f(n)[l\{\varepsilon(0)\}]^n \right) \cdot \sum_{n=0}^{8} p_f(n)[f_d(0)]^n. \quad (26)$$

In the case of gamma photons, when absorption and detection are included, the modified factorial moments are defined as follows:

$$\left.\frac{d^n r_\alpha[l\{\varepsilon(z)\}]}{dz^n}\right|_{z=0} \equiv \overline{\mu}_{d,\alpha n}; \qquad \alpha = s, f, \quad (27)$$

$$\left.\frac{d^n q_\alpha(g_d)}{dg_d^n}\right|_{z=0} \equiv \overline{\nu}_{d,\alpha n}; \qquad \alpha = s, f. \quad (28)$$

These factors are straightforward to calculate but lead to expressions that contain sums, which have an increasing number of terms for higher order moments and are suitable for being calculated using computers and symbolic handling programs like Mathematica [6], which can handle the derivations easily.

3. QUANTITATIVE RESULTS

The number distributions were calculated for both neutrons and photons for plutonium metal spheres of varying mass. The model accounted for absorption. The values obtained were compared to simulations with the code MCNP-PoliMi [5]. For this reason MCNP-PoliMi had to be modified to supply the necessary tallies. The nuclear physical constants such as the values of p, and the fission-parameters $p_s(n)$, $f_f(n)$, etc. were taken from MCNP-PoliMi runs and used in the analytical model. The probability to induce fission varies with the mass of the sample. The values of p can be found in the table below for the three samples for which we have performed calculations.

Case	Mass (kg)	p
1	0.335	0.06388
2	2.680	0.12464
3	9.047	0.18383

Table I. Probability to induce fission, p, for one neutron depending on the mass of the sample. The samples have a composition of 80 wt% Pu-239 and 20 wt-% Pu-240

Figures 1-3 show examples where we have compared data from MCNP-PoliMi for samples with no absorption and the results from analytical calculations where absorption was accounted for. The comparison shows the effect of including absorption into the model, are shown in .

As can be seen, for photons there is a very evident effect of self-shielding. The probability of having high numbers of photons escaping the target are reduced significantly when absorption is taken into account. The results show that for realistic samples which might be investigated with typical non-destructive assays (NDA), the initial advantage of having high photon multiplicities is not present when using external

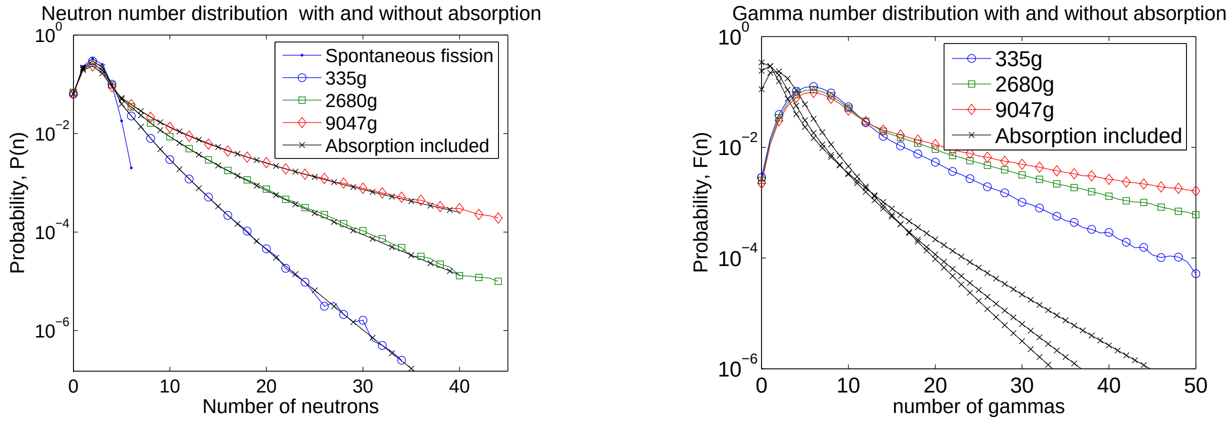

Figure 1. The number distribution of neutrons and gamma photons for plutonium samples of different masses. The Monte Carlo simulation shows the result with no absorption and the analytical data show the distributions when absorption is taken into account.

because the internal absorption of photons is much greater than that for neutrons in materials of high atomic numbers.

Photon detectors might still be useful if we consider the test-setup, if the sample is heavily screened with low Z materials, and/or materials with high neutron absorption cross sections, which do not screen gamma photons, then photon detectors might be advantageous compared to neutron detectors.

Tables II-III show the statistics of leaked neutrons and photons and the probabilities for detecting singlets, doublets, or triplets of these particles in probabilities per initial event (which is these cases are spontaneous fissions). Without internal multiplication the higher multiplicities would have very low counts, but with the possibility of having very large bursts of particles the combined likelihood of detecting for example three of 15 leaked particles grows very rapidly, and thus enables better predictions to be made for of the mass and isotopic composition of the sample.

When observing the number distribution of detected neutrons and photons in the case of 50% detection efficiency, we can see that the probabilities of detecting large numbers of particles decrease as expected, but we can also observe the fact that the singlets and doubles have probability of being detected that is higher than the amount singlets and doublets that actually escape the sample, since they can also be the result of only detecting a part of the particles of a bigger burst.

In the case of gamma photons one can note the somewhat unexpected result that it is more likely to detect rather high multiplicities such as triplets and quadruplets for a lighter sample than for a heavy one. The reason is that, although the total amount of generated photons is higher for the heavier samples, the self-shielding effect counteracts this. The fast growing self-shielding (with increasing mass) constitutes of course a disadvantage. Note that these probabilities are per source event, and in a sample of higher mass the number of spontaneous fissions will be higher.

It is still evident that a complete understanding of the statistics as well as measuring of a large number of different multiplicities will allow for faster and more accurate conclusions. The analytical models used here

Neutrons		335g	2680g	9047g
Absorption	singlets	0.21889	0.20682	0.19504
	doublets	0.29731	0.26519	0.23516
	triplets	0.2221	0.19345	0.16668
Detection efficiency, $\epsilon = 0.10$	singlets	0.19277	0.20471	0.21287
	doublets	0.024551	0.035786	0.049832
	triplets	0.0027315	0.0070163	0.015136
Detection Efficiency, $\epsilon = 0.25$	singlets	0.33452	0.32372	0.30543
	doublets	0.098325	0.1109	0.11744
	triplets	0.021883	0.035162	0.04721
Detection Efficiency, $\epsilon = 0.50$	singlets	0.37579	0.3456	0.31422
	doublets	0.21646	0.20553	0.18928
	triplets	0.079179	0.088465	0.090366

Table II. The absolute probabilities of having between one and three neutrons generated (leaked out, not internally absorbed) or detected with finite detection efficiency, from one initial source event. It can be seen that while some parameters increase with mass, some others decrease with mass for the same case. The full probability distribution makes it is easier to estimate the full trends and emission properties of a specific sample.

Photons		335g	2680g	9047g
Absorption	singlets	0.22246	0.29499	0.29221
	doublets	0.23442	0.20249	0.15579
	triplets	0.17566	0.10954	0.075544
Detection Efficiency, $\epsilon = 0.10$	singlets	0.18993	0.14105	0.1184
	doublets	0.028384	0.018158	0.016358
	triplets	0.0038585	0.0026916	0.0031021
Detection Efficiency, $\epsilon = 0.25$	singlets	0.31234	0.25007	0.20916
	doublets	0.10284	0.06464	0.051787
	triplets	0.028138	0.017295	0.015884
Detection Efficiency, $\epsilon = 0.50$	singlets	0.33685	0.31531	0.27517
	doublets	0.19744	0.13216	0.099861
	triplets	0.088085	0.050477	0.039393

Table III. The absolute probabilities of having between one and three photons generated (leaked out, not internally absorbed) or detected with finite detection efficiency, from one initial source event. Note that triplets for example can increase in one case with mass up to a certain mass, only to decrease after that, therefore a definite prediction of mass can only be made by using several detection parameters together.

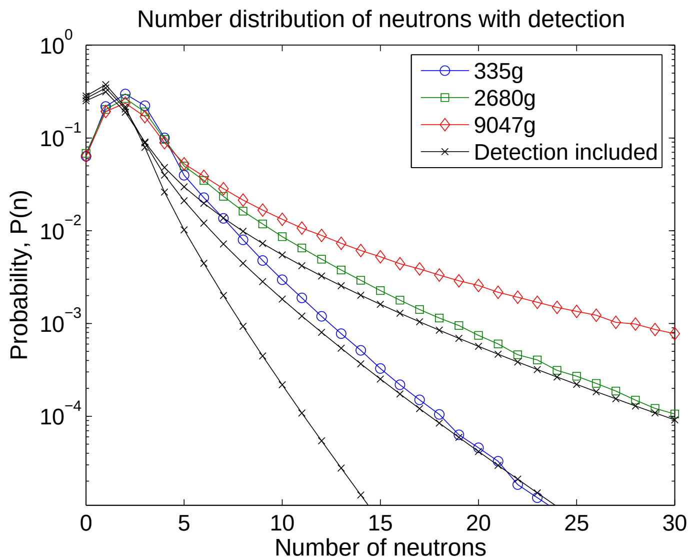

Figure 2. The statistics for neutrons when a detection efficiency of 50% is incorporated into the model. The plot shows that the statistics change and the likelihood of low detection numbers are higher compare to how many bursts there are of that multiplicity from the sample.

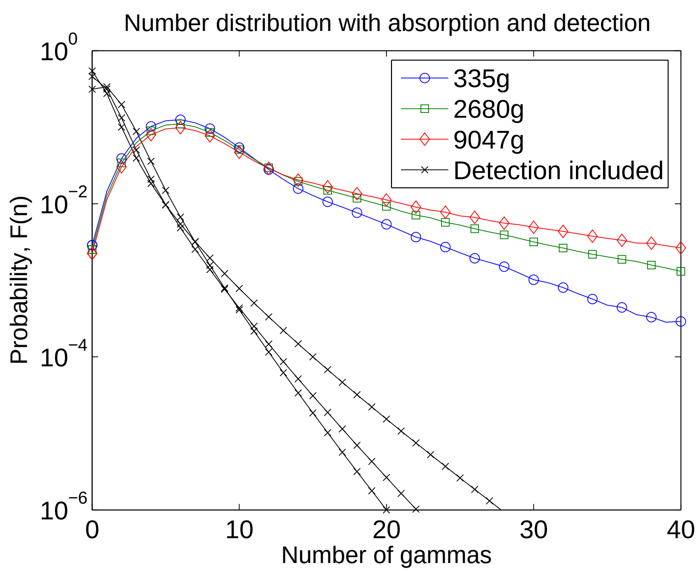

Figure 3. The statistics for neutrons when a detection efficiency of 50% is incorporated into the model. Just as when absorption is included the detection process once again lowers the probabilities for high multiplicities being observed.

could also be used as indicators of what type of detectors are best practice to employ. In small samples, the larger photon multiplicities per fission event will lead to the emission of high photon multiplicities from the sample, and the detector efficiency will be the only limiting factor what regards measurement efficiency.

For heavier samples, on the other hand, only neutrons escape the sample easily and neutron multiplicity counters might then be better to use. Finally the use of detectors capable of detecting both neutrons and photons provide more possibilities for analysis, as the joint probability distribution of both neutrons and photons can be used to obtain more information on the sample's characteristics.

4. CONCLUSIONS

We have used the symbolic computation code Mathematica to calculate high order terms of the number distribution of fissile samples with the inclusion of absorption. The results show that when absorption is accounted for, the number of photons emerging from the sample will decrease significantly, whereas the neutrons are not affected to the same extent. The multiplicities of photons leaving the sample could decrease so much that neutron multiplicities become higher. With the introduction of the detection process into the model of the leaked neutrons and photons it is possible to simulate the detector response and find the probabilities for different multiplicities of both neutrons and photons. Using this information, one could assess different sample masses with regards to what type of emission has the higher multiplicity when using detectors for non-destructive assay of the material.

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