

# AN ALTERNATIVE PERSPECTIVE ON THE WEIGHTED POINT MODEL FOR PASSIVE NEUTRON MULTIPLICITY COUNTING

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## ABSTRACT

The failure of the point model for highly multiplying Pu metal items is largely a result of the fact that at larger masses the leakage multiplication is not constant throughout the item. This represents a violation of the underlying assumptions of the point model. One way to tackle this dilemma is to introduce finite extent parameters into the model by integrating the multiplication terms present in the basic point model equations over the spatial dimension(s). Here we consider simple cases of weapons grade Pu spheres and squat cylinders in both a non-reflective geometry and surrounded by a basic packing material reflector. The reduced spatial moments,  $g_n = \langle M^n \rangle / \langle M \rangle^n$ , as a function of Pu mass are evaluated and the prospect of using these functions as weighting factors for terms in the point model is considered.

## INTRODUCTION

It is well established that application of multiplicity counting to the assay of Pu metal in the kg range results in an assay bias [1,2]. This is partially attributed to a violation of the spatial invariance assumption in the point model expressions used for the detector response function during the analysis step. A weighted point model approach has been developed by Krick and colleagues at Los Alamos National Laboratory [3] in which some of the parameters in the point model are replaced by more complex forms chosen semi-empirically to match the results of analog Monte Carlo simulations of the multiplicity counting experiment. In this way the point model equations are treated as physically based guides to the shape of the calibration curve with empirically determined, problem dependent, coefficients.

In applying the point model to a spatially extended problem, several of the assumptions upon which it is built may be challenged – for example the assertion that the neutron detection efficiency is single valued. For the assay of compact metallic items perhaps the largest influence comes from the spatial variation in the multiplication throughout the object. This is the only aspect of the problem we shall consider. Our aim here is to delineate this effect in isolation to better understand the physics at work.

The Singles, Doubles and Triples rates are enhanced over the corresponding rates in the absence of induced fission by factors of  $\Phi_1$ ,  $\Phi_2$  and  $\Phi_3$  respectively. These factors, in general, depend on the self-multiplication taking place (for the present discussion we shall neglect any distinction between total and leakage self-multiplication and present only self-multiplication values), the  $\alpha$ -value and combinations of basic nuclear data (which to first order can be taken as constants for our limited purposes). In the case of metallic items the random-to-(SF,n) production ratio,  $\alpha$ , is expected to be close to zero apart from for the inevitable presence of low atomic number contaminants (this is very object specific and also depends on the relative Pu and  $^{241}\text{Am}$  composition but  $\alpha$  could amount to several percent of the nominal pure dioxide value for real metallic items). The problem then becomes how to evaluate the volume (which is equivalent to *mass* for a uniform item) weighted average (VWA)  $\Phi$ -values (e.g.  $\langle\Phi_i\rangle$ ,  $i=1$  to 3). We see from the form of the point model equations that  $\Phi_3$  is of order 5 in the (leakage) self-multiplication factor,  $M$ , and so we need a means to evaluate  $\langle M^5 \rangle$ , the VWA value of  $M(\mathbf{r})$  raised to the fifth power, the explicit spatial dependence being indicated by the introduction of the spatial parameter  $\mathbf{r}$ . For convenience we have introduced  $g_n = \langle M^n \rangle / \langle M \rangle^n$  which varies over a smaller dynamic range and gives a direct indication of the deviation from using a spatially invariant average value.

We have generated  $g_n$  functions for a few simple shapes in free space and under a simplified reflective condition in order to discover how important the effect is and how, for a particular class of problems, a simple parametric correction might be applied. If so then the analysis can remain as a single variable problem;  $\langle M \rangle$  being the unknown to be extracted from the assay, but with approximate functions for the  $g_n$  derived from it as a function of mass and shape which allow the higher orders terms to be factored into the data interpretation more in an iterative way as needed.

## DESCRIPTION

Our objective was to carry out a pure physics study, broadly linked to real life problems but, not specific to classified shapes or conditions. To this end we decided to study a nominal weapons grade Pu/Ga delta phase alloy of density  $15.30\text{g.cm}^{-3}$  in which the Ga to Pu mass ratio,  $x$ , is 0.01. The relative isotopic composition of the plutonium was taken to be: 0.012, 93.694, 5.920, 0.341 and 0.033 wt% with respect to  $^{241}\text{Pu}$  for the isotopes  $^{238-242}\text{Pu}$  respectively. No  $^{241}\text{Am}$  or other minority impurities were included in the transport calculations. This set of isotopes was chosen because the high  $^{239}\text{Pu}$  content leads to high multiplication effects with high spatial variability, thereby leading to a severe test of the ability to parameterize the effect. Calculations were performed over the mass range 1g to 10kg for two simple shapes with stainless steel cladding. Results for the items located in free space and also in a moderator similar to the packaging material within a storage drum were generated. The two shapes considered were the sphere and the squat right circular cylinder (diameter equal to the height). The stainless steel cladding was specified as AISI 316 S12 SS EN58J of density  $7.96\text{g.cm}^{-3}$  with an elemental composition of Fe(71), Cr(18), Ni(8) and Mo(3.0), respectively, where the values in parenthesis are the weight fractions. The cladding thickness was fixed at 0.4cm for all cases. The packing material was represented by cellulose ( $\text{C}_6\text{H}_{10}\text{O}_5$ ) of density  $0.25\text{g.cm}^{-3}$ . It was in the

form of a hollow cylinder of radius 29cm and length 84cm with a central cavity of radius 9cm with a length of 50cm. For the purposes of modeling the Pu items were placed with axial and radial symmetry at the geometrical centre of the cavity in the cellulose.

Multiplication factors were calculated using the Monte Carlo code MCNPX [4]. Spontaneous fission neutrons with a  $^{240}\text{Pu}(\text{SF}, n)$  spectrum approximated by the Watt shape [with spectral parameters  $a= 0.799 \text{ MeV}$  ,  $b=4.903 \text{ MeV}^{-1}$ ] were launched from spherical surfaces as a function of radius in the case of the sphere and from rings as a function of both radius and height in the case of the squat cylinders.

## RESULTS

The spontaneous fission sources were located at 10 radial steps for the case of the spherical items while for the squat cylinders rings were located at 10 radial steps and 5 axial steps to cover the half height of the squat cylinders. Figure 1 shows the net multiplication variation for the spherical samples in air for the masses modeled against the source radial position over the sample total radius.

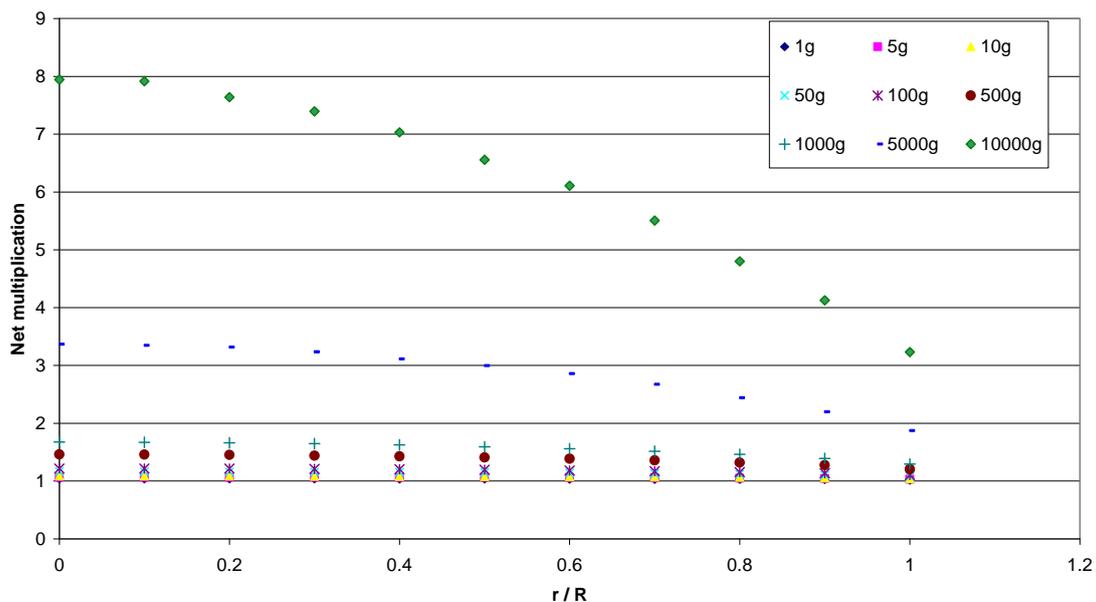


Figure 1. Net multiplication variation with the source radial position within Pu/Ga alloy spheres of different masses

Figure 2 shows the net multiplication variation for the cylindrical items in air as a function of source ring position, results are presented for the 10 radial positions (from the centre to the outside radii) and 5 axial positions (from centre to top of the squat cylinder) for each of the masses.

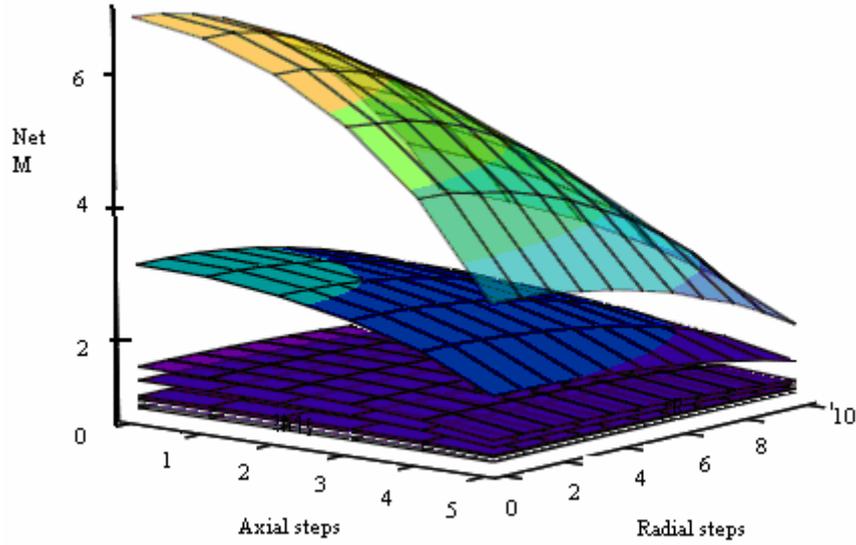


Figure 2. Net multiplication variation with the source radial and position within Pu/Ga alloy cylinders of different masses

Similarly the net multiplication was modeled for items placed in the cellulose packing material. Once the data was calculated polynomial functions  $M(r)$  and  $M(r,z)$  were fitted to the data obtained for the spherical and cylindrical shapes respectively, in order to represent the geometrical variation of the net multiplication. In these expressions the scalar  $r$  denotes the radial variable and the scalar  $z$  the axial variable. The volume weighted averages were calculated as follows:

$$\langle M^n \rangle_{sphere} = \frac{4\pi \int_0^R M^n(r) r^2 dr}{V} \quad \langle M^n \rangle_{cylinder} = \frac{2\pi \int_0^{H/2} \int_0^R M^n(r,z) r dr dz}{0.5V}$$

The reduced spatial moments,  $g_n = \langle M^n \rangle / \langle M \rangle^n$  with  $n=2 \dots 5$  for each of the shapes and reflective conditions studied,  $g_2$ ,  $g_3$ ,  $g_4$  and  $g_5$ , are shown in Figures 3, 4, 5 and 6 respectively. The value of  $g_1$  is equal to unity by definition and so is not plotted.

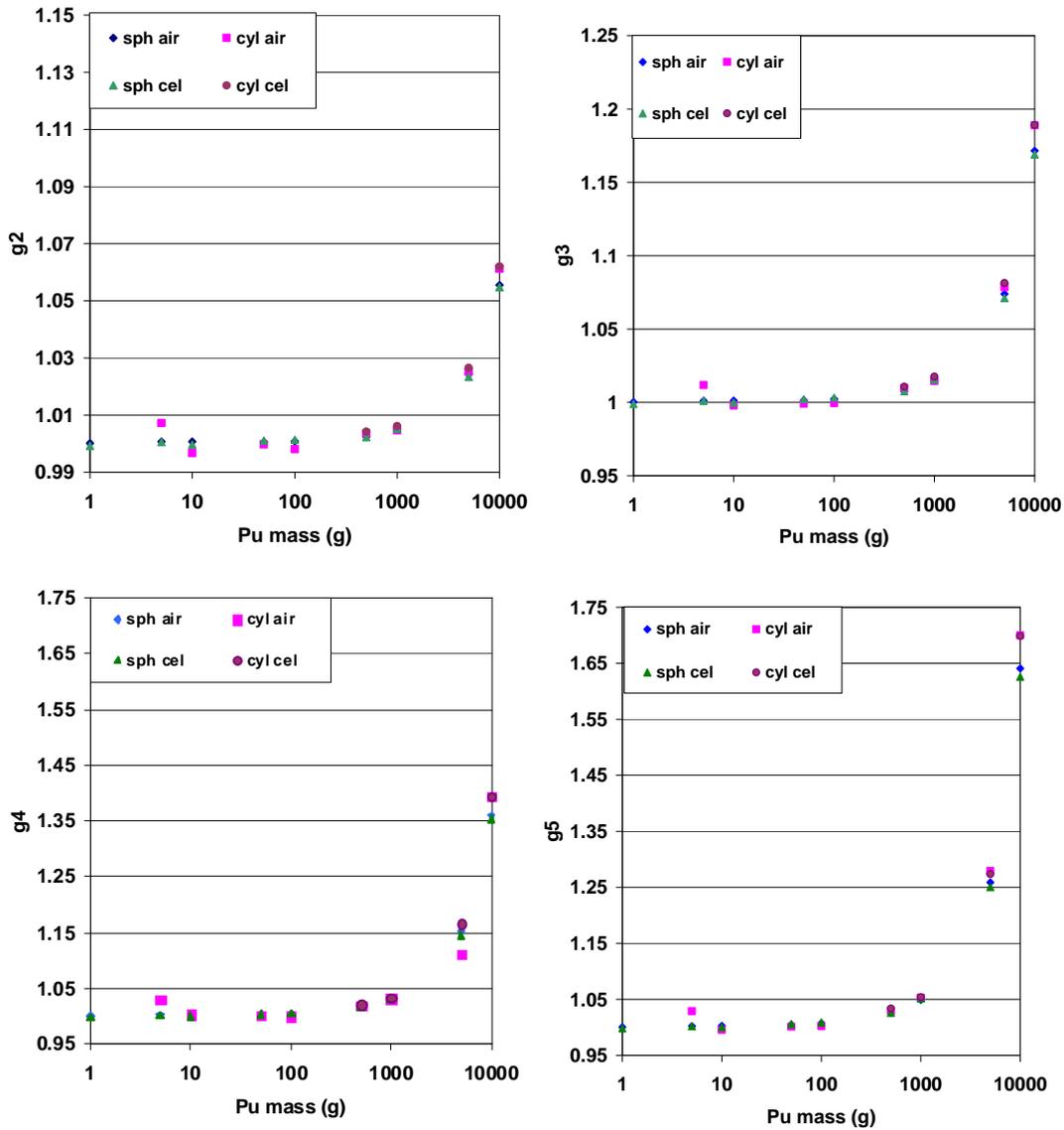


Figure 3, 4, 5 and 6. Reduced spatial moments ( $g_2$ ,  $g_3$ ,  $g_4$  and  $g_5$  respectively) vs mass for spheres and right squat cylinders in air and cellulose.

The behaviour of the reduced spatial moments has been parameterized in terms of Pu mass assuming a simple exponential trend; the coefficients for the different shapes and reflective conditions are presented in Table 1. The coefficients are the result of free fits. In practice we would expect all of the  $g_n$  values to approach unity from above as the mass is drops to zero. That the  $a$ -coefficient close to unity is therefore as expected and suggests it could be clamped to unity without penalty in application.

Table 1 Fitting coefficients for spheres (Sph) and right squat cylinders (Cyl) for the multiplication reduced spatial moments  $g_n$  express in terms of mass for the two reflective cases studied.

$g_n = a.exp[b.mass(g)]$								
	Sph in air		Sph in cellulose		Cyl in air		Cyl in cellulose	
	a	b	a	b	a	b	a	b
$g_2$	0.9999	5.E-06	0.9999	5.E-06	0.9998	6.E-06	1	6.E-06
$g_3$	1	2.E-05	0.9998	2.E-05	0.9998	2.E-05	0.9995	2.E-05
$g_4$	1	3.E-05	0.9999	3.E-05	1.001	3.E-05	0.9991	3.E-05
$g_5$	1.0004	5.E-05	1.0005	5.E-05	1.0023	5.E-05	1.0023	5.E-05

From the plots it is clear that the reduced spatial moments for both shapes in both air and in cellulose show similar behaviour. The reflecting condition of the packing material was expected to have a larger effect on the functions due to a higher induced fission but in fact it is not a dramatic effect. In the same way differences were expected for the different geometrical shapes; since the sphere has the largest volume to surface area ratio of any non-reentrant uniform body and so we expect the multiplication to be the highest of any shape. The squat cylinder has an aspect ratio that makes it resemble a spherical somewhat. The difference between the two results is not dramatic which suggests that  $g_n$ -factors for a class of problems could be extracted and applied without having to know the exact shape of each item perfectly.

## DISCUSSION

In the present analysis we shall neglect the small difference between the total multiplication and the leakage self-multiplication so that the point model expressions can be written solely in terms of the single parameter leakage self-multiplication which we shall denote by  $M$ .

If we evaluate the Singles (S), Doubles (D) and Triples (T) rates according to the point model [5] and form the ratio with the corresponding rate calculated without self-multiplication then we obtain the enhancement effect which stems from the possibility of induced fission. For S, D and T we call these ratios  $\Phi_1$ ,  $\Phi_2$  and  $\Phi_3$  respectively. The theoretical evaluation of these terms is reliant on knowing relationships between some basic nuclear data [5] which are encapsulated in what we refer to here as the K-coefficients. The ratios  $\Phi_1$ ,  $\Phi_2$  and  $\Phi_3$  are given by:

$$\Phi_1 = M$$

$$\Phi_2 = M^2 \cdot \{1 + (M-1) \cdot K \cdot (1 + \alpha)\}$$

$$\Phi_3 = M^3 \cdot \{1 + (M-1) \cdot (1 + \alpha) \cdot K_{31} + 3 \cdot (M-1) \cdot K_{32} + 3 \cdot (M-1)^2 \cdot (1 + \alpha) \cdot K_{33}\}$$

For detailed, high accuracy work one may seek to calculate neutron source production weighted K-values. Because of their differences we often consider the prompt (SF, n), delayed fission neutron, ( $\alpha$ , n) neutron and prompt induced fission neutron spectra individually. However, for the present discussion our aim is to not to provide a

refined analysis for a particular case but rather to create a general picture of the magnitude of the spatial variation of  $M$  when applying the point model expressions. Thus, to first order, we take in the case of multiplying metallic items:

$$\begin{aligned} K &\approx 2.2 \\ K_{31} &\approx 3.4 \\ K_{32} &\approx 2.8 \\ K_{33} &\approx 6.1 \end{aligned}$$

The VWA values of  $\Phi_1$ ,  $\Phi_2$  and  $\Phi_3$  may be calculated as a function of the  $\langle M \rangle$  and the spatial moments using the following expressions:

$$\Phi_1 = \langle M \rangle$$

$$\Phi_2 = \langle M \rangle^2 \cdot \{g_2 + (g_3 \langle M \rangle - g_2) \cdot K \cdot (1 + \alpha)\}$$

$$\Phi_3 = \langle M \rangle^3 \cdot \{g_3 + (g_4 \langle M \rangle - g_3) \cdot (1 + \alpha) \cdot K_{31} + 3 \cdot (g_4 \langle M \rangle - g_3) \cdot K_{32} + 3 \cdot (g_5 \langle M \rangle^2 - 2 g_4 \langle M \rangle + g_3) \cdot (1 + \alpha) \cdot K_{33}\}$$

Figures 7 and 8 illustrate the variation of  $\Phi_2$  and  $\Phi_3$  respectively for the range of items modeled. These values have been calculated using the discrete values of  $g_n$  calculated directly and not in their parameterized form. As previously noted, for metallic Pu the expectation is that the  $\alpha$ -value in the material will be low because the  $\alpha$ -particle interaction rates with high atomic number elements are strongly suppressed by the coulomb potential barrier. However inevitably light element impurities will be present albeit at a low level. For illustrative purposes we have assumed a random-to-(SF,n) value of 0.06 which is based loosely on our experiences with reference materials used to manufacture low mass calibration sources.

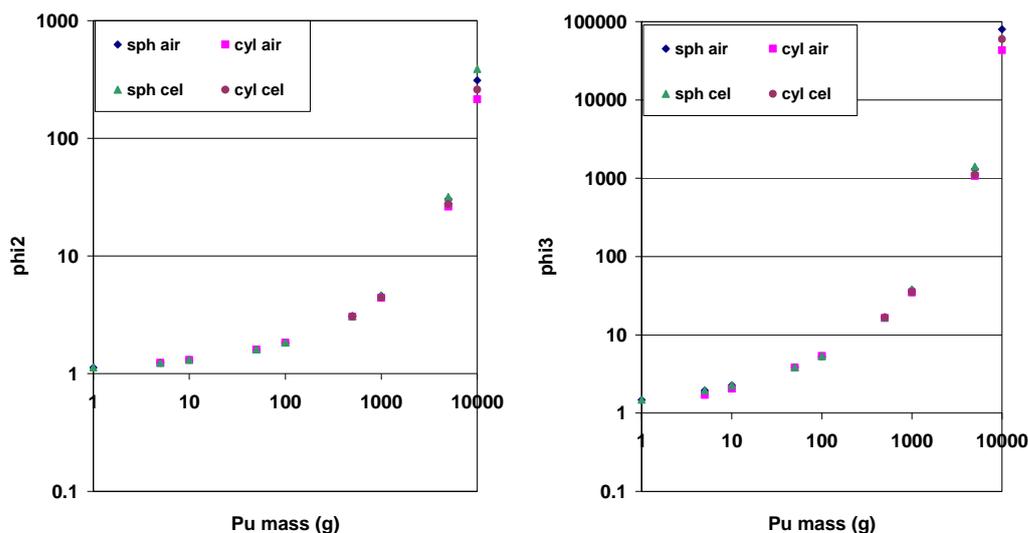


Figure 7 and 8. Variation of  $\Phi_2$  and  $\Phi_3$  respectively for spheres and right squat cylinders in air and cellulose with mass.

## CONCLUSIONS

We have performed Monte Carlo simulations of the multiplication as a function of position within simple Pu items located both in free space and surrounded by a cellulosic moderator. The results have been used to extract volume weighted moments of the multiplication factor as a function of Pu mass. The deviation of the geometrical parameter  $g_n = \langle M^n \rangle / \langle M \rangle^n$  is an indication of the breakdown of the point model. To the extent that  $g_n$  can be simply, generically and accurately approximated as a function of e.g. the Pu mass and shape of the item for a given assay problem, it can be inserted into the familiar point interpretational model and applied iteratively. The results here presented show that it is fairly independent of shape, at least for the case of the spheres and right squat cylinders studied in this work, which suggests that  $g_n$ -factors for a class of problem could be extracted and applied without having to know the exact shape of the item perfectly.

The lower mass results are also of direct interest in the assay of Pu bearing waste and scrap where it is important to establish bounding cases for the potential over estimation of mass introduced by multiplication in any lumps that may be present.

## REFERENCES

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