The Contribution of and Uncertainty Associated with Self-Multiplication when Assaying Plutonium in Waste by Passive Neutron Coincidence Counting - 8386

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ABSTRACT

When Pu is present in waste items as lumps, the Reals neutron coincidence signal may be bolstered by virtue of self induced fission events. This gives rise to a positive bias to the assay result and leads to a one sided contribution to the total measurement uncertainty. In the general it is not feasible to determine to magnitude of this effect experimentally and allowance for it must therefore be estimated using separate ad hoc rules.

In this paper we to develop a simple model for the self-multiplication enhancement in small lumps allowing the importance of the effect in waste assay to be quantified. In addition, an approach is suggested for how to propagate an uncertainty contribution in to the final result.

INTRODUCTION

Passive Neutron Coincidence Counting (PNCC) of spontaneous fission neutrons for the assay of bulk Pu is a well-established technique [1]. The measurement yields an estimate of the effective Pu-240 mass, m_{eff}, which when combined with the relative isotopic composition from high-resolution gamma spectroscopy or acceptable knowledge enables the total Pu mass, fissile gram equivalent, α-activity and heat output to be obtained.

The approach is also routinely applied to the nondestructive assay of waste containers. Compared to the measurement of bulk product materials the isotopic composition, chemical form and physical structure of the Pu in waste may be poorly known. This means that the (α,n) production rate can be highly variable and is difficult to estimate from presumed knowledge of the Pu material. As a consequence, the Totals counting rate is often not amenable to quantitative analysis. Determination of m_{eff} must therefore be based solely on the net Reals rate corrected for deadtime, background and matrix effects. Matrix perturbation of the response can have a significant impact in waste measurements and special techniques involving interrogating the matrix with an external correlated neutron source are commonly used to derive a correction factor. Residual uncertainties associated with the inhomogeneity of the waste matrix and the spatial distribution of the Pu throughout the container remain and are propagated into the Total Measurement Uncertainty (TMU). These considerations again set waste assay measurements apart from bulk product measurements, since bulk items are generally quite uniform.

Another distinguishing feature is that whereas in the assay of bulk materials self-multiplication is expected, in the measurement of waste it is commonly assumed that the Pu will be dilutely intermingled with the matrix throughout the container so that self-multiplication can be neglected to first order. [For the present discussion we shall not consider the usual and rare special

situation of Pu or α–contaminated low atomic number materials with high (n,2n) cross-sections, or Pu contaminated U pieces, which might mimic multiplication in larger quantities of Pu].

However, if the fissile material is in the form of lumps or aggregates (e.g. metal chips, foundry spills, filled crevices, discarded parts, cracked pellets, etc.) self-multiplication may indeed take place in waste and scrap. In this circumstance the PNCC assay will tend to over-report the Pu because the induced fission signal will be wrongly assigned to the effective spontaneous fission mass, there being no way for the PNCC to distinguish the two sources of correlated neutrons in the waste arena (in which the Totals rates are not so quantitative, the detection efficiency is typically too low to permit Triples counting and residual uncertainties can mask theoretical relationships between the Totals, Reals and Triples signals). The degree of the over reporting increases with the mass of Pu in the lump and is dependent on the concentration of Pu and also on the ratio α, the ratio of the random (α,n) to spontaneous fission neutron production rate, since the (α,n) neutrons also serve to induce fission events.

A false high Pu mass is conservative in that subsequent safety and handling conditions applied to the item will be more stringent than perhaps is necessary. But on the other hand unnecessary restrictions may be invoked incurring time and cost penalties and also repository capacity may be unnecessarily allotted. It is clear, therefore, that bounding of the self-multiplication effect is required for the purpose of deriving reasonable and justifiable TMU's. The objective of this paper is to develop a simple logical framework by which the contribution to the TMU due to multiplication effects can be made.

BACKGROUND

It is well known that neutron multiplication affects the assay of bulk Pu product material during routine nuclear materials safeguards activities. When the isotopic and chemical composition is known reliable methods to correct for it have been developed [2, 3]. If the form of the material is known the effect may be calculated using radiation transport codes [4, 5]. In field measurements the geometry of bulk items can also be used to good effect in difficult to measure cases (e.g. impure materials of high and unknown (α,n) proportion) to derive an estimate of the multiplication [6, 7]. A variation of the known-M method has been developed by Thomason et al [8] for use with the multiplicity counting of Pu scrap. In this approach the leakage self-multiplication is approximated by a quadratic polynomial in terms of the effective Pu-239 (Pu-240eff) mass. Fill height variations of the cans can also be accommodated. The advantages of the known-M method applied to conventional PNCC of bulk items have also been reviewed and highlighted recently by Nizhnik et al [9].

There are many instances of measurement campaigns which report, for a given class of materials (most often PuO_2 which is commonly the target of safeguards measurements), measured or estimated multiplication effects [e.g. 10-15]. The literature is vast and it is a daunting task to consolidate it. Fortunately, Ming-Shih Lu and his collaborators [16] developed a useful and simple empirical correlation between the leakage multiplication factor M_L and the fissile mass, m_{fis}, which goes some way to bringing coherence to this field. Their correlation equation, for reactor grade oxides, was based on a statistical analysis of the measurements of some 123 items.

Based on the foregoing short review we are led to consider two empirical forms for M_L. From the known-M camp we have:

$$M_L = 1 + a.m_{fis} + b.m_{fis}^2 \quad \text{(Eq. 1)}$$

where *a* and *b* are coefficients to be estimated for a class of materials over a pertinent dynamic range. While from the work of M-S Lu et al we have:

$$M_L = 1 + m_{fis} / (A + B.m_{fis} + C.m_{fis}^2) \quad \text{(Eq. 2)}$$

where *A*, *B* and *C* are also parameters to be fit to the problem. From [16], for m_{fis} in g, *A* = (1.5261±0.202), *B* = (8.0656±0.242), *C* = (-0.92594±0.0474) giving a regression coefficient R^2 = 0.967 over the 123 RG PuO_2 items considered.

An alternative first principles approach, and one we shall follow, is based on a simple physical model of the multiplication model [17-19] is developed in the next section. Our choice of this route is founded on our objective to create a simple yet generic approach applicable to the diversity of Pu contaminated waste in the Department of Energy complex. Individual, dense, lumps are not expected to exceed a few 10's of grams at most and the principle material type of concern is Weapons Grade Pu. We note that in moist air Pu rapidly oxidizes and so that aged waste drums will invariable contain friable oxide even if metal was present initially.

MULTIPLICATION ENHANCEMENT

Within the familiar one-energy group, prompt-fission, point-geometry model, the true Reals rate expected from a single lump is increased by a factor C given by [17]:

$$C = M^2.[1+(M-1).\kappa.(1+\alpha)] \quad \text{(Eq. 3)}$$

where, in the approximation that the probability, p_c, of a neutron born in the lump is parasitically captured to the probability p_f that it will induce fission is small (i.e. $p_c/p_f << 1$), M is the prompt leakage multiplication factor which is also therefore numerically equal to the total multiplication.

α is the ratio of (α,n) to spontaneous fission neutrons produced in the lump

and

κ is a function of basic nuclear data [8,17] which for the present purposes may be treated as being constant with a value of roughly 2.2.

This is our master equation for C. We shall develop a linearized form for simplicity but note we can always apply the full form as needed. It is evident from the form of C that the mass per unit Reals calibration becomes non-linear due to the multiplication dependence, which in turn depends on the (fissionable) mass of the body. For a well-behaved size progression of bodies (e.g. fixed composition, density and shape but incrementing dimensions) it would be possible to

construct an empirical non-linear calibration based on this expected trend (smooth and monotonic). This is a suitable approach for certain bulk items.

For small bodies it is straightforward to show that to first order [17]:

$$\delta = (M-1) \approx \psi \cdot \Sigma_f \cdot x \qquad \text{(Eq. 4)}$$

where, ψ is a shape dependent constant of proportionality (essentially independent of the characteristics of the material), Σ_f is the macroscopic fission cross section of the body and *x* is the mean escape path length for neutrons. For a given material type (i.e. Pu grade and chemical form) Σ_f is directly proportional to the material density, ρ.

If we take the lumps to have fairly regular shapes then *x* is proportional to the characteristic dimension of the body and we may go on to write:

$$x \propto (m/\rho)^{1/3} \qquad \text{(Eq. 5)}$$

where m is the fissionable mass and ρ is the density of the multiplying body which is proportional to the Pu density.

Combining results we have:

$$\delta \approx \theta \cdot \rho^{2/3} \cdot m^{1/3} \qquad \text{(Eq. 6)}$$

where θ is a constant for the material type and bodystyle.

Note that if we need extra accuracy in representing δ, perhaps to extend to higher masses, we may go to higher orders in $(\rho^{2/3} \cdot m^{1/3})$. Also, for a given grade of material we may replace the fissionable mass by the total mass without loss of generality, provided the coefficients are chosen appropriately.

For small bodies $M \approx 1$, or in other words $\delta << 1$, and we can use this fact to linearize the expression for the enhancement factor C. Thus, we have from our master expression:

$$C = 1 + [2 + \kappa \cdot (1 + \alpha)] \cdot \delta + [1 + 2 \cdot \kappa \cdot (1 + \alpha)] \cdot \delta^2 + \kappa \cdot (1 + \alpha) \cdot \delta^3 \qquad \text{(Eq. 7)}$$

which can be simplified, for small bodies, such as would be expected in waste, to:

$$C \approx 1 + [2 + \kappa \cdot (1 + \alpha)] \cdot \delta \qquad \text{(Eq. 8)}$$

in cases where $(1 + \alpha)$ is not excessive (e.g. of the order of a few).

Substituting for δ we arrive at the following first order expression:

$$(C - 1) \approx [2 + \kappa \cdot (1 + \alpha)] \cdot \theta \cdot (\rho/\rho_{ref})^{2/3} \cdot m^{1/3} \qquad \text{(Eq. 9)}$$

where θ is a (new) constant for a given material type (e.g. Weapons Grade Pu), ρ is the (fissionable) Pu density of the material and ρ_{ref} is the (fissionable) Pu density of the reference material used to derive θ. *m* is the fissionable mass of the lump, $\kappa \approx 2.2$ as previously intimated and α is the $(\alpha,n)/(SF,n)$ ratio as defined earlier.

The result of this simple one collision physical model is that $(C - 1)$ is directly proportional to $\delta = (M - 1)$, which in turn is directly proportional to the mean escape path length of the body. We shall see later that the first order approximation works well, for the present needs, which we consider to be up to about 30g of Weapons Grade Pu (WG Pu) metal (as an extreme) in the form of a squat cylinder (which is similar to the sphere in having a high volume to surface ratio and therefore a high self-multiplication effect).

To apply this expression a value of θ for a reference material is required. This can be readily estimated from calculations already available in the literature [17-19]. The Pu density of the lump is needed but for practical purposes might be taken as the reference value depending on what is known about the waste. The fissile mass of the lump must be estimated and this can be done (iteratively if need be) from the assay value. Note that in waste the normal expectation is that the Pu will not be present as a single lump. For example, when assaying 55 US gal. drums it is common accepted practice, when estimating the contribution to the TMU due to unknown source distribution, to assume at least three equal localized sources of activity are present. Thus, in the application of the present expression *m* should be taken as the assay value divided by, n, where n is the minimum number of lumps that can reasonably be assumed to be present in the container. Under expert review High Resolution Gamma Spectroscopy (HRGS) scan data, for instance, may be able to confirm a spatially distributed Pu source.

The final requirement for the evaluation of an upper limit of C is the value of α. For pure metallic items α should be close to zero. However, in aged waste metallic Pu will invariably (partially) oxidize. For pure Weapons Grade PuO_2 α is of the order of unity. However, real wastes are routinely observed to have higher α-values reflecting the presence of low atomic number impurities (such as F) with a high (α,n) cross section in intimate contact with the α-emitter. High α-values are also characteristic of lean Pu salts and the presence of relatively high Am-241 abundance can drive the value higher. As a general rule it is difficult to accurately estimate the (α,n)–to–(SF,n) ratio of an item and further to ascribe the α-value to individual Pu lump that may be present. This is problematic because when α is large (>10 say, and for PuF_4 it can exceed 100) the self-interrogation of the lump by (α,n) neutrons can result in the induced fission signal completely swamping the SF signal. The potential for over reporting is therefore very high (although in such extreme cases one may expect Reals-to-Totals ratio to be (much) lower than normal and the precision on the Reals to be poorer than expected if the signal was coming from clean material). A reasonable range for α in typical WG Pu waste based on our operational experience is (3 ± 2).

For dilute Pu we would expect the enhancement factor $C \approx 1$. For Pu distributed into *n* roughly equal concentrations we would anticipate C being given by the formula developed. As an estimate of the associated uncertainty we propose to take half of the spread as a simple pragmatic

approach i.e. ±(C-1)/2 would be the relative uncertainty (compared to an assay value based on C=1). A more refined approach is generally not justified given the level of prior information available on the contents of historic waste containers.

For larger lumps the first order expression can, of course, be replaced by the full expression if needed and as discussed in the next section higher order expressions for δ can be used.

APPLICATION

Of any non-reentrant homogeneous body the sphere has the largest self-multiplication factor owing to its maximal volume-to-surface area ratio. But a spherical lump is an unrealistic thing to expect in nature. In the interests of being somewhat conservative however a fairly squat body (all characteristic dimensions about equal) is prudent. On this basis we consider the squat cylinder (diameter, D = length, H) to be a justifiable yet extreme lump shape model. Multiplication factors for a pure WG Pu metal are summarized in Table I for this geometry. The results are based on the Monte Carlo calculations described in [17] for bodies in free space. The impact of the surrounding matrix is secondary [18, 19], as well as being difficult to estimate since it will vary case by case.

Table I Calculated multiplication properties of squat WG Pu metal spheres located in free space.

Pu mass (g)	Size D = H (cm)	Leakage Multiplication deviation, $\delta = (M - 1)$
0.01	0.09306	0.0066
0.03	0.1342	0.0096
0.1	0.2005	0.0150
0.3	0.2892	0.0198
1.0	0.4319	0.0285
3.0	0.6230	0.0413
10	0.9306	0.0643
30	1.342	0.0958
100	2.005	0.1544
300	2.892	0.2465
1000	4.319	0.4459
3000	6.230	0.9001

The relative isotopic composition of the Pu, expressed in weight percent was taken to be 0.012, 93.81, 5.81, 0.349 and 0.022 for Pu-238 – Pu-242), respectively. A density of 15.80g.cm^{-3} was adopted. No Am-241 or alloying agent (such as Ga) was considered. Cross-sections were taken from the NJOY (JEF) ’87, continuous energy, evaluation and were mixed at a temperature of 300K. The initiating source spectrum was that of Pu-240 spontaneous fission. The values of $\delta = (M - 1)$ presented in Table I are expected to be accurate in an absolute sense to better than 5%. This is more than adequate for our present purposes because at low masses the consequences of multiplication are modest and (in any event) other factors contribute to the overall uncertainty in the assay value to a greater extent (e.g. matrix corrections; α-value, etc.). We also feel justified in using the same values for all fast neutron initiating spectra, since, for example the O(α,n)

reaction has a mean energy similar to that of fission spectra. In the other extreme, namely for the largest masses (e.g. in the kg range), M is not constant throughout the cylinder and in the strictest sense the point-model is violated. This regime is not the focus of our attentions but even if it were the treatment would still probably be sufficient for estimating TMUs given the other sources of possible assay uncertainty. But in this extreme one may also question whether the cylindrical model is appropriate. Lumps of such extreme mass would likely be manufactured items.

To proceed we introduce the approximation:

$$\delta = a_1 \cdot [(\rho/\rho_{ref})^{2/3} \cdot m^{1/3}] + a_2 \cdot [(\rho/\rho_{ref})^{2/3} \cdot m^{1/3}]^2 + a_3 \cdot [(\rho/\rho_{ref})^{2/3} \cdot m^{1/3}]^3 \qquad \text{(Eq. 10)}$$

Taking *m* to be the total mass of WG Pu, for convenience, values of the coefficients (a_1, a_2, a_3) may be extracted from the data presented in Table I. For this special case (ρ/ρ_{ref}) is unity since we are treating the WG Pu metal as the reference case. Best-fit values were obtained using the Deming code [20] with equal weighting in δ. Depending on the mass range of interest the order of the fit and the value of the coefficient will change. For small lumps and modest α-values we only need the linear term and may use the linearized version of (C – 1). For other cases we can use higher order expressions for δ and the master equation for C. Table II gives a summary of working values. We give uncertainties in the principle parameters from Deming to show they are well defined by the data in Table I, but stop short of presenting covariance information because in application model assumptions will also be challenged and contribute to bias. In other words the uncertainty contribution does not make use of the uncertainty in the fitting parameters since they are determined with good accuracy for our purposes.

Table II Parameters for the δ expression for squat cylinders of WG Pu.

WG Pu mass range (g)	Fitted coefficients		
	a_1	a_2	a_3
≤ 10	$(2.9457 \pm 0.031) \times 10^{-2}$		
≤ 30	$(3.0187 \pm 0.033) \times 10^{-2}$		
≤ 300	$(2.6363 \pm 0.046) \times 10^{-2}$	$(1.5456 \pm 0.083) \times 10^{-3}$	
≤ 3000	$(3.0337 \pm 0.074) \times 10^{-2}$	$(-2.6824 \pm 1.7) \times 10^{-4}$	$(1.7257 \pm 0.085) \times 10^{-4}$

Nizhnik et al [9] discuss how to calculate the fissionable mass weight fraction given the relative isotopic composition. (We use the term fissionable here because the reaction is induced by fast neutrons rather than thermalised neutrons). WG Pu within the DOE complex is dominated by Pu-239 and the composition does not deviate much – the Pu-240eff weight fraction is normally within a band of ±5% (relative). For the composition used in this paper the Pu-240eff weight fraction is 5.88wt%. Thus the model parameters presented in this work should therefore be widely applicable as is.

To be clear on how to apply the proposed scheme to WG Pu, in the polynomial expressions for δ, for WG Pu, m is the total Pu mass of the individual lump(s). This is estimated from the total Pu mass, m_{tot}, present according to $m = (m_{tot}/n)$ where n is the minimum number of lumps assumed to be present. In turn m_{tot} is related to the Pu-240eff mass, m_{eff}, through the relative isotopic composition so that $m_{tot} = (m_{eff}/w)$ where w is the Pu-240eff weight fraction obtained from gamma spectroscopy or Acceptable knowledge.

Scaling to different Pu densities, where it is known, is straightforward. For instance, suppose we have a WG Pu powder of density $3.5 g.cm^{-3}$. The weight fraction of Pu in PuO_2 is 0.882 and so the Pu density, ρ, to be used in the formulae is $3.09 g.cm^{-3}$ compared to the reference value of $15.8009 g.cm^{-3}$.

OTHER SOURCES OF IINFORMATION AVAILABLE TO THE EXPERT REVIEWER

The present analysis should not be applied blindly in isolation from other information such as:

- Multiplicity data when available
- Reals-to-Totals ratio
- HRGS data (which can flag self attenuation, quantify Pu composition and indicate impurity reaction γ–rays from e.g. C, F and Be)
- Records
- Radiography and other data.

Taken in the round these sources allow the expert analyst to formulate a clearer picture of the character of the waste and to apply the multiplication TMU accordingly.

FUTURE WORK

This is a new proposal and it remains to be seen how expert analysts might use and adapt the concepts for routine work. The key is how to go from the apparent assay value to a plausible maximum lump size and then how we choose a corresponding α-value. The Reals enhancement factor then lies in the interval zero to C. A TMU contribution of $\pm(C-1)/2$ (relative uncertainty) is suggested in the text but when sufficient operational experience has been garnered perhaps it will be found that the range can be treated as being more akin to $\pm 3\sigma$ such that $\pm(C-1)/6$ (relative uncertainty) would be a suitable estimate of the standard deviation. These are discussion for the future.

CONCLUSIONS

By introducing a generic scheme we have taken the first pragmatic steps to accounting for multiplication effects in the TMU in the determination of Pu-240eff –mass by PNCC in waste. We have used a simple physical model to derive the underlying relationships needed. A first order expression suitable for small, dense lumps was set out, primarily for instructive reasons, but this is not a restriction of the method. The method is readily scaleable to material of different isotopic composition, density and α. Numerical parameters suitable for practical applications

have been established and we feel the approach is fit for the intended purpose reflecting operational experiences and the general state of knowledge of waste items.

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