Neutron Self Multiplication Effects in PDP Items in the Context of Coincidence Counting - 10496

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ABSTRACT

Recently two independent but complementary studies concerning self-multiplication in waste have appeared. In the first by Croft et al a general theoretical approach was presented from which the behaviour could be formulated given knowledge or assumptions about the Pu contaminated waste form. In the second by Stanfield et al an experimental approach was followed based on measurements on a set of Performance Demonstration Plan (PDP) reference items.

In the present work we revisit the first principles treatment inserting the PDP characteristics to illustrate how the two approaches are related. Because the PDP items have fixed dimensions and by design are weakly multiplying the first order theoretical approach cast in terms of the Pu density (proportional to mass in this special case) is sufficient to understand the experimental trend.

The assay of unknown items must be performed in a carefully prescribed way to ensure quality results. The understanding and treatment of uncertainty in the multiplication is somewhat subjective however since the data needed to interpret an individual drum or waste stream is incomplete or lacking. A general structured approach and understanding can suffice to bound the problem and fortunately self multiplication is typically a minor contributor to the overall uncertainty budget in most waste streams.

INTRODUCTION

The discussion presented will be of particular interest to the non-destructive assay waste measurement community concerned with using Passive Neutron Coincidence Counting (PNCC) to assay transuranic Pu contaminated material prior to shipment to the Waste Isolation Pilot Plant (WIPP) in Carlsbad New Mexico USA. However the methods are quite general and expected to have broader appeal than WIPP. The central theme is that self multiplication in the fissile material leads to an over estimate of the Pu mass by PNCC. This represents a positive bias. It constitutes an unrecognized error because the measurement data generally offers no clue that significant multiplication enhancement of the neutron signal has taken place. Current practice is to add the self multiplication effect into the estimate of the total measurement uncertainty as if it is a symmetric two-side effect. But as already noted this is not the case, multiplications results only in overestimates. The consequence is that assay result plus twice the TMU, which is used as a practical upper bound, leads to an over reporting, sometimes dramatically high so if worse case assumptions are made on the potential effect based on the fissile mass being present as a single 'lump', because in effect the upper mass estimate now includes the physical effect and twice the bounding effect when really the apparent value should be reduced to account for the physical effect. The implications for waste operations of using this flawed logic are severe: overly conservative operational limits must be set which means more drums are needed to dispose of a given amount of waste. This is wasteful of the repository volume. Drums which do not exceed permitted limits may need to be unpacked unnecessarily which incurs cost and dose exposure risk. Finally in sending drums to the repository with artificially high assay values the activity limit of the repository is being underused.

When using the PNCC technique self-multiplication in lumps (chucks or aggregates) of Pu can bias waste assay results high and lead to unrecognized error. This can lead to, amongst other things, under utilization of repository limits. Bounding and quantifying the potential magnitude of the effect is important to building a complete Total Measurement Uncertainty (TMU). Constructing a plausible and justifiable TMU is crucial in setting operational limits designed to ensure that waste package criteria such as ^{239}Pu fissile gram equivalent (FGE), of concern for post closure criticality assessment, ^{239}Pu equivalent activity (PE-Ci), which factors into the repository safety case as an inventory limit based on pathways back to the environment, and thermal wattage, which controls local heating and ultimately geological integrity of the repository, are not exceeded [1]. The temptation to build overly conservative TMU budgets is bad science and may negatively impact best practice clean-up efforts through the setting of operational limits which are lower than necessary.

The contribution to the TMU for PNCC resulting from self-multiplication effects has received attention recently because it is an example where if one is unnecessarily pessimistic the impact can be quite restrictive. For instance if all the Pu is assumed to be present in the form of a single worst case (spherical) impure (high alpha-n to spontaneous fission neutron ratio) lump of Pu the enhancement to the Reals rate means that an operational or working limit several times below the threshold must be applied. Often Acceptable Knowledge (AK) or other information (real time radiography images, segmented gamma scanner profiles, Reals coincidence to Totals neutron

count rate ratio etc.) can be used to show that such pessimistic assumptions are not appropriate so that the blind application of an overly conservative algorithm is not good measurement practice. Croft et al [2] have discussed the dilemma facing the metrologist from a somewhat theoretical perspective in an attempt to provide the human Expert Analyst (EA) with the understanding and numerical framework to make informed assessments. Stanfield et al took a direct experimental approach to demonstrating that self-multiplication effects are modest by performing a series of measurements as a function of Pu mass using a High Efficiency Neutron Counter (HENC) [3]. In this paper we show how these apparently conflicting approaches to treating multiplication effects in coincidence counting may be unified. We show how an experimental approach can be represented by basic theory and how the theory may be used in other measurement scenarios.

Totals (gross or Singles) counting is simply the word we use in PNCC to refer to the total detected neutron event rate. Reals counting is the way in PNCC we identify time correlated events associated with neutrons liberated in bursts from spontaneous fission from neutrons that occur randomly in time and are characteristic of alpha-n reactions which in waste are less quantitatively related to the mass of Pu present. In essence every detected event opens a coincidence gate and the number of events falling into it forms a pair associated with the triggering event. Once corrected for accidental coincidences (which we do by opening a similar gate later in time when the time correlation in the counter has vanished) we are left with the true pairs coincidence rate or real coincidence rate net of accidentals a quantity that is commonly referred to as the Reals rate for short.

REVIEW OF THE THEORETICAL TREATMENT

According to the one-energy group, prompt-fission, point-geometry model, the Reals coincidence rate expected from a single lump is increased by a factor, C, given by [2]:

$$C = M^2 \left(1 + \left(M - 1\right) . \kappa . \left(1 + \alpha\right)\right) \qquad \text{(Eq. 1)}$$

where, in the approximation that the ratio of the probability, p_c, that 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 random to spontaneous fission neutrons generated in the lump; and κ is a function of basic nuclear data which is rather insensitive to the Pu composition for the present purposes and may be treated as being roughly constant with a value of roughly 2.18 [2].

For pure Pu metal α is close to zero since (α, n) production is forbidden and the delayed neutron contribution is relatively small. In reality there is very little Pu-metal in waste (except as entrained Pu in salts) because in aged waste metallic Pu exposed to mist in air will (readily) oxidize. For pure Weapons Grade PuO_2 α is of the order of unity. Actual wastes are routinely observed to have higher α-values reflecting the presence of low atomic number impurities (such as C and 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 resulting from refining operations and the α-value can be driven higher still (e.g. for aged Pu) subject to the ^{241}Am:Pu ratio. It can be difficult to

accurately estimate the α value of an item and even harder to ascribe the α-value to individual Pu lumps that might 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 the Reals-to-Totals ratio to be (much) lower than normal and the associated precision on the Reals rate to be correspondingly poorer (to the extent of being worthless) 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). During human expert review assessments may be made on a case by case basis by drawing on all available additional information available, for example alpha induced gamma ray lines might indicate the presence of certain impurities [4]. In this way the reality may be introduced into the assay process in an intelligent fashion.

It seems reasonable to assume that for a lump to be of concern it needs to have a shape which is compact. A sphere would be the most multiplying shape but it would be an unlikely shape to find in waste. Therefore if we assume that a squat cylinder (diameter = height) has the characteristics we are seeking it was shown in [2] that for weapons grade (WG) Pu a reasonable approximation for a single lump is:

$$(M-1) = a_1 \left(\rho^{2/3} \,.\, m^{1/3}\right) + a_2 \left(\rho^{2/3} \,.\, m^{1/3}\right)^2 \qquad \text{(Eq. 2)}$$

where m is the mass of WG Pu present in the lump expressed in grams, ρ is the density of the Pu expressed in $g.cm^{-3}$, and the numerical values of the free parameters, based on fits to Monte Carlo calculations are a_1=(4.187±0.073)x10^{-3} and a_2=(39.0±2.1)x10^{-6} for m<300 g.

Armed with Equations (1) and (2) and item specific estimates for ρ, m and α one can derive estimates for C. For dilute (non-multiplying, M=1) material C would equal unity and this is one bounding case. Typically for waste the assay value is generated on the assumption that the Pu is dilutely dispersed. The factor C can then be treated as a reasonable upper bounding case. Alternatively one could make a nominal correction for self multiplication and spread the uncertainty symmetrically. This is a matter of procedural choice to be taken in discussion with the regulators based on knowledge of the waste stream.

The value of m to be used is the mass of the individual lump(s) assuming that the individual lumps are spread throughout the waste container so that they are neutronically isolated from each other. When treated waste is being assayed it is not expected that large single lumps will be present. The nature of the waste generation process and recovery operations prevents manufactured items being present. Also note that packing records or radiometric measurement information may also be on hand to support this. Thus, in common with the estimation of the uncertainty associated with the spatial distribution of Pu within the item in most cases it is credible to assert that a number, n, (such as 3-7 for a 55 US gal drum, say) of lumps are present or that the largest lump can not exceed a certain mass, m_L, (e.g. 15 g, say, and for reference a slug of Pu delta-phase metal 1 cm diameter x 1 cm long would have a mass of about 12.4 g) based on waste generation and sorting criteria. Thus:

$$m = \frac{m_{assay}}{n} \quad if\ m < m_L \qquad otherwise\ m = m_L \tag{Eq.3}$$

where m_{assay} is the measured Pu mass. To give a concrete example which for most applications could be considered reasonably extreme, suppose we are assaying oxidised waste so that we take the density of Pu present in the power to be of the order of 3 g.cm^{-3}, we take α=3 and m=15 g. Inserting Eq.2 into Eq.1 and evaluating returns a value of C of 1.25.

CONSIDERATION OF THE DIRECT EXPERIMENTAL APPROACH

Stanfield et al [3] opted to look at the self-multiplication issue in an entirely different way. They measured the Reals response as a function of WG Pu mass in a HENC (LANL HENC#2) loaded with various combinations of NIST traceable calibration sources of the PDP type. The system had previously been calibrated in Neutron Coincidence Counter mode using PDP sources and calibration verified with a different set of similar sources. Multiplicity analysis using factory parameters is also available. An integral pillar of the Waste Isolation Pilot Plant (WIPP) certification program is testing with Performance Demonstration Plan (PDP) sources. The PDP program is a DOE administered scheme whereby certified working reference materials are used to check the performance of an assay system used to sentence waste destined for WIPP before it enters service and periodically thereafter. Based on data taken from [5] the sources containing small particle size WG PuO_2 (Phase I and IIA) are doubly encapsulated in stainless steel. The PuO_2 within the sources is dispersed in a diatomaceous earth substrate which for the present discussion we take to be benign from the perspective of prompt (fission neutron) induced fission. The radioactive material/diatomaceous earth substrate is contained in a volume of approximately 306 cm^3. The Internal diameter of the inner capsule is 1.71" (4.3 cm) and the height between the base and the graphite felt frit cap is about 8.13" (20.7 cm). The mean geometrical escape path length $\langle l \rangle$ [6] is estimated at approximately 0.983" (2.5cm). The PDP sources contain between 0.02 and 75 g of WG Pu. This means that the highest WG Pu density is approximately 75/306 = 0.25 g.cm^{-3}.

Because the PDP sources are not squat cylinders we should strictly not apply Equations (1) and (2) blindly – since those expressions were intended as general guidance for ill-defined concentrations of Pu in waste rather than for specific manufactured items. To proceed we therefore draw on the work of Bourva and Croft [6] in which it is was shown that for weakly multiplying items we can approximate the self-multiplication by:

$$(M-1) = k.\rho.\langle l \rangle \tag{Eq.4}$$

where as before ρ is the density of Pu in g.cm^{-3}, $\langle l \rangle$ is the mean geometrical flight path length of particles born uniformly and isotropically within the active region to the outside, and k is a constant of proportionality. Using the table of multiplication values given in [2] for squat cylinders of WG Pu metal it is easy to extract a suitable value of k for WG Pu, it is 0.0105 cm^2/g.

Using Equations (4) & (1) we can now estimate the factor *C* for PDP style sources. We take κ=2.18 [2] and α=2.10. This value of α is based on a Reals-to-Totals analysis (corrected for multiplication iteratively using multiplicity analysis) of the measurement data taken by Stanfield et al [3] and also weekly Quality Assurance (QA) check data taken using some of the same sources. It is difficult to estimate a value for α by a method other than analysis of PNCC or multiplicity data because the contribution from (α,n) production in the diatomaceous earth is unknown. We expect some of the Pu alpha-particles to emerge from the grains of PuO_2 and into the substrate [roughly SiO_2 86wt%; Na_2O 5wt%; MgO 2.5 wt%; Al_2O_3 2.5 wt%; Fe_2O_3 2.7 wt%; CaO 1.3 wt%)] but the energy spectrum of the emergent alpha-particles is dependent on knowing the particle size distribution of the powder and their effectiveness for generating neutrons depends on also knowing the impurity content (e.g. B, Be, F … concentrations) of the diatomaceous earth, which is a naturally occurring material and as such presumably subject to variability in composition. In generating the C-values we also show the impact of varying α by ±0.15 about the adopted value of 2.10. We see that the influence of even this quite large change is not very important.

The study by Stanfield et al [3] spanned the WG Pu mass range from 3g to 242g but in practice this was achieved by using combinations of a few smaller mass items containing 3, 26.1, 50 and 67g of WG Pu. The combinations used are shown under the comments column of Table II. Also shown in Table II is the true Pu mass loading and the apparent mass loading which is the sum of the mass of the individual items each multiplied by the respective Reals self-multiplication enhancement factor. The ratio of the two values is also given and this is the factor by which the Reals signal is expected to be increased over the same amount of Pu present in non-multiplying (dilute) form. The ratio is also plotted in Fig 1. It is important to note that the relatively minor impact of self-multiplication with mass observed by Stanfield et al [3] is reflected in the results reported in Table I and Fig 1 also. So we can appreciate the reasons behind the absence of a strong experimental effect. We should also note that at the few percent level, which is the domain in which we are operating, it is difficult to draw precise conclusions from the experimental results because the response will also vary a little depending on how the combination of sources is distributed with the simulated waste drum. Bear in mind the calibration is also based on PDP sources and so one does not expect to see strong deviations. This was understood by Stanfield et al but what they wished to show directly was that the use of two sided symmetric uncertainty estimators, which is currently a procedural requirement, based on an algorithm which is a function of Pu mass and uses default parameters is inappropriate and unduly pessimistic at the upper mass range. McElroy et al have also made this point [7].

Table I. Estimated multiplication deviation from unity, (*M*-1), and Reals enhancement factor, *C*, due to self-induced fissions, as a function of Pu mass in PDP style sources. For these calculations an α-value of 2.10 has been adopted. The uncertainty listed, σ_C, shows the impact of varying α by ±0.15 which is the order of variation we see in extracting α from the weekly check data.

Pu mass, m (g)	Pu density, ρ ($g.cm^{-3}$)	(M-1)	C	σ_C
0.02	6.537E-05	1.720E-06	1.0000	0.0000
0.03	9.805E-05	2.580E-06	1.0000	0.0000
0.3	9.805E-04	2.580E-05	1.0002	0.0000
0.55	1.798E-03	4.730E-05	1.0004	0.0000
3	9.805E-03	2.580E-04	1.0023	0.0001
3.6	1.177E-02	3.096E-04	1.0027	0.0001
7.6	2.484E-02	6.536E-04	1.0057	0.0002
15	4.902E-02	1.290E-03	1.0113	0.0004
26.1	8.530E-02	2.245E-03	1.0197	0.0007
30	9.805E-02	2.580E-03	1.0227	0.0008
50	1.634E-01	4.300E-03	1.0379	0.0014
65	2.124E-01	5.590E-03	1.0494	0.0018
67	2.190E-01	5.762E-03	1.0509	0.0019
75	2.451E-01	6.450E-03	1.0571	0.0021

Table II. True mass, apparent Reals mass, enhancement factor (ratio) and source combination information estimated here in an effort to explain from first principles the results expected for the study of Stanfield et al [3].

Pu mass, m (g)	Apparent mass (g)	Ratio	Comment
3	3.01	1.002	1x3
26.1	26.61	1.020	1x26.1
100	103.79	1.038	2x50
176.1	182.30	1.035	3x50 + 1x26.1
217	226.10	1.042	1x67 + 3x50
242	252.72	1.044	1x67 + 3x50 + 1x26.1

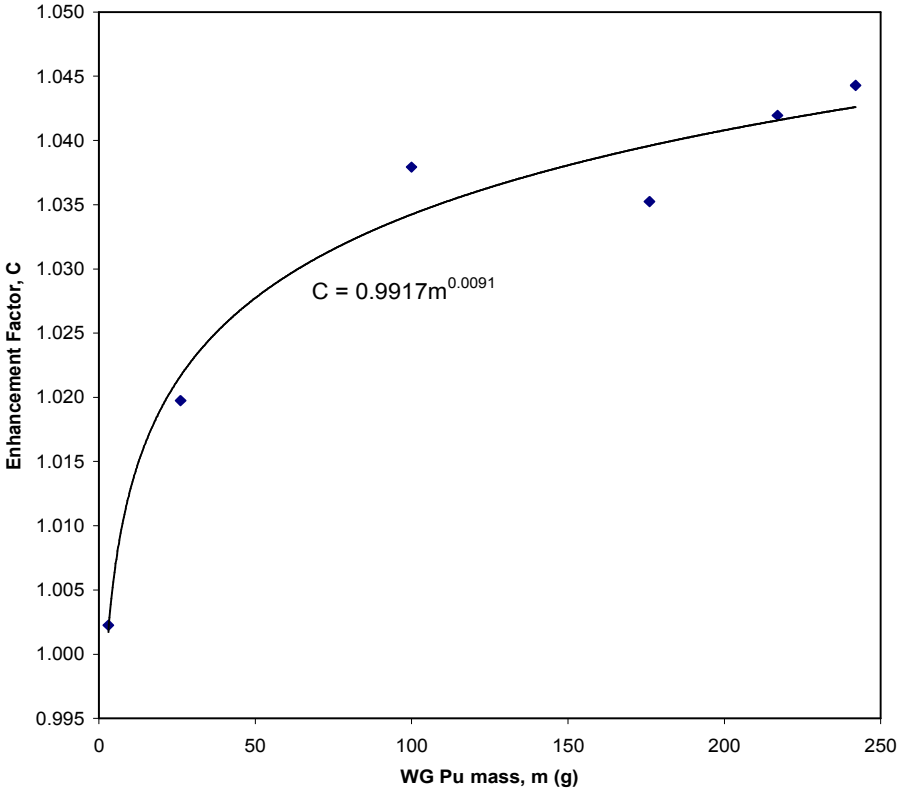

Fig1. Plot of the calculated Reals enhancement factor over the mass range 3 g to 242 g of WG Pu for the PDP source combinations used in the study of Stanfield et al [3]. We note from the data and trend line that the effect and mass dependence is rather weak for this combination of sources and is therefore difficult to compare directly against experiment given the additional (e.g. spatial variation) uncertainties associated with the experiments at the few percent level.

DISCUSSION

Earlier work has highlighted that self-multiplication in lumps of Pu can result in a one sided positive bias to assay results based on PNCCs calibrated under the assumption that Pu is dilutely dispersed throughout the matrix.

Quantifying the magnitude of this effect is not a trivial exercise and the blind application of software engines developed in other contexts may be misleading. It is important therefore for the expert analysis to bring to bear as much secondary information as possible [2] in order for a reasonable and credible item specific estimate to be made. In turn this allows a suitable uncertainty contribution to be propagated. We feel this is good measurement practice and preferable to treating all items as worst practical cases which can result in applying a large uncertainty no matter what the actual situation may be. Historically it has been waste drums that did not show much multiplication and have high mass that cause artificial issues since if a large

uncertainty allowance is made for potential multiplication effects the assay value plus the coverage factor can, for example, exceed the FGE limit.

The primary intent of the paper was therefore to provide the human expert analyst with the tools and physics based guidance on how to establish credible multiplication error limits based on measurement and other acceptable knowledge about the item.

The assumption that Pu in waste is contained in a few squat lumps remains conservative. The broader question is how to best balance conservatism and realism in trying to estimate the uncertainty and it is here that experience and knowledge of the individual waste streams comes into play.

We wish to emphasize that it is normal practice to calibrate a PNCC assay system at the factory using sources with little or no self-multiplication. Pu present in the form of lumps will therefore tend to result in assays that are too high. That is to say the assay value will already be biassed high so that an additional allowance for multiplication does not need to be added. Rather the correction factor should be in the direction of reducing the reported value. However, the final calibration used to WIPP certify an instrument across the operational mass range may well be performed using a set of PDP style sources and the factory calibration will be certified using a different set of PDP sources. In this case the calibration may well have a built-in contribution from self-multiplication present in these working reference materials. For this reason we have also shown how to estimate the magnitude of the Real coincidence enhancement factor for WG PDP sources and shown that the effect is rather modest since the PuO_2 within them is not highly concentrated since it is uniformly distributed within a diatomaceous earth substrate. We find that even for the highest mass (75 g WG Pu) PDP capsule the Reals rate is elevated by only about 6% over the dilute case. Therefore supposing such an item alone was used to create a calibration, the worst case underestimation (of genuinely dilute Pu in a waste drum) would be of the order of 6%, while if there were concentrations present similar to the concentration present in the PDP source the assay would be unbiased, furthermore if genuine massive lumps were present (high density and high (α,n) self-interrogation) the assay value would be biassed higher than the true loading. Therefore as generally practiced there is no reason why an assay value should require a coverage factor of more than a few percent.

CONCLUSIONS AND FUTURE WORK

We have created a coherent framework within which the issue of self-multiplication, as it affects passive neutron coincidence counting of Pu contaminated waste, can be discussed and quantified. We have shown how to extend the treatment to include self-multiplication present in the familiar PDP-style working reference materials. We conclude that in the worst case a system calibrated with such sources will under-report by at most a few percent while when lumps are genuinely present the reported assay value will tend to be higher that the true mass present so that a coverage factor is not appropriate.

In passing we note that it might also be useful to exercise these concepts further by: (1) systematically comparing the neutron and gamma data in waste and standards measurements and (2) systematically comparing PNCC with neutron multiplicity results in such a study. Finally, we can also compare HENC data with waste-generators' estimates (e.g. packing list and Real Time Radigrapghy data) which for new arisings prepared under modern best practice may well be rather reliable in some situations. Although this is done rountinely today to reconcile individual item assays a systematic analysis across a broad body of data and focused on the multiplication issues would be beneficial in developing improved operational practice.

An obvious extension of the work presented here is to non Weapons Grade Pu-compositions and other elements. Occasionally, for example, an item containing non-WG Pu or mixed actinides will present with what seems like a large amount of multiplication. Some processes such as (n,2n) can in some situations can mimic such effects. But consideration of Heat Source Pu (HSPu rich in ^{238}Pu) amongst other common situations can also be treated in a similar way to that applied here.

From a metrology perspective however we hope that we have demonstrated that self-multiplication in plutonium contaminated waste can be understood and quantified by the subject matter expert rather simply. It is important to realise that it tends to gives rise to a one sided bias (over estimate) so that current practice of inflating the assay result by adding an additional coverage factor for a hypothetical self-multiplication based on the mass of Pu present makes no sense. From a procedural point of view this could be handled through the introduction of more detailed uncertainty reporting structures that accommodates one sided and asymmetric confidence limits.

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