

# Calibrating Passive Neutron Multiplicity and Waste Counters Using Calibrated $^{252}\text{Cf}$ Sources: Estimating $a_{\text{Pu}}$ -Values Without Pu

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## *Abstract*

$^{252}\text{Cf}$  sources are especially interesting for calibrating passive multiplicity counters without the need for Pu because:

- they are genuinely point-like and being lightly encapsulated offer a nearly isotropic unperturbed fission spectrum of neutrons
- for all practical purposes they can be considered as a pure source of spontaneous fission neutrons which considerably simplifies interpolation. That is the leakage self multiplication,  $M_L$ , can be taken as unity and the ratio  $\alpha$  of  $(\alpha, n)$ -to- $(\text{SF}, n)$  neutrons can be taken as zero.
- it is a well studied multiplicity system with the factorial moments  $v_i$  of the distribution  $P(v)$  being well established.
- the energy spectrum is similar to that of the spontaneous spectra of the Pu isotopes
- it is readily available with well known outputs,  $Y$ , determined absolutely by reference to Mn-baths operated by National Standards Laboratories

In this work we exploit these features in the framework of the point-model interpretational equations discussed in ASTM C 1500 'Standard Test Method for Nondestructive Assay of Plutonium by Passive Neutron Multiplicity Counting'. In particular we: extend the equations to include the delayed neutron contribution showing how to extract the gate utilization factors (GUFs) accurately from the data available; extend the solutions given in the Appendices to cases where the Triples GUF is not equal to the square of the Doubles GUF; and, illustrate how to project  $^{240}\text{Pu}_{\text{eff}}$  performance from measured  $^{252}\text{Cf}$ .

## *Introduction*

The field of international nuclear safeguards demands the highest accuracy from non destructive radiometric assay in order to minimize the amount of unaccounted special nuclear material. The reason is that over many assays random uncertainties average out while systematic errors do not. Therefore, when the flow of materials is high even a small unrecognized systematic bias can result in a significant quantity of material being unaccounted for over a short period of time. Increasingly Monte Carlo calculations are being applied to develop the calibration of waste and safeguards passive neutron assay systems. Modern Monte Carlo particle interaction and transport codes, for example MCNPX [1], are very powerful and within the limits of our knowledge of basic nuclear data can faithfully model all of the important physical processes taking place in the item and detector. They provide a means to interpolate or extend the calibration when representative reference materials are unavailable or impractical to apply. Although in principle they may be used to estimate the response absolutely, given that the details of construction are adequately specified in practice, this is rarely done. It is considered better

practice to normalize the model calculations to a carefully performed and highly controlled experiment(s). In this way the overall accuracy can often be improved because certain sources of systematic error, largely common to all calculations (e.g. moderator density and as built dimensions, effective volume of  $^3\text{He}$  proportional counter gas etc.) cancel out when one deals in ratios.  $^{252}\text{Cf}$  spontaneous fission sources provide a readily available and convenient surrogate for Pu and other neutron emitters in many cases. The emission spectrum is reasonably well known and they are available with very small dimensions lightly encapsulated. For many practical applications they can therefore be regarded as point-like. This considerably simplifies the interpretation of experimental results and often satisfies the assumptions of simple analytical models so that a means exists to gain added confidence with the Monte Carlo results. Based on the results of an intercomparison exercise [2] we conclude that the current state of the practice of several National Laboratories around the world for the absolute determination of the total neutron output of commercially available Cf sources is 0.3 – 0.4 % relative at the 68% confidence level. Pu items containing masses known to greater accuracy than this are possible but for benchmarking purposes remain limiting because of large relative uncertainties in spontaneous fission half-lives and multiplicity yield per fission. Given that Cf is readily available and can be neutron calibrated to a fraction of a percent we are therefore motivated to develop methods that take full advantage of this fact. Also we recognized that the  $^{240}\text{Pu}$ -effective worth of an item could easily be re-expressed in terms of a  $^{252}\text{Cf}$ -effective mass with the conversion being amenable to simple cross calibration. With the application of suitable detector specific response characteristics then the relationship would be a basic nuclear parameter.

It is worth noting explicitly that Cf reference sources of known emission rate are commonplace whereas Cf sources whose mass is accurately known are rare. In contrast Pu reference materials are traditionally available as oxides of well known weight, well known Pu composition and well known Pu:oxide weight fraction. This strongly influences our thinking. In principle however there is no technical reason why an isotopically pure, non-multiplying metallic  $^{240}\text{Pu}$  sample with negligible ( $\alpha, n$ ) contribution cannot be manufactured with a mass known to comparable accuracy (certainly  $<0.1\%$ , say) of the current best reference materials. The availability of such materials for basic research would be highly desirable in our endeavors to reduce the uncertainties in basic nuclear data parameters of interest in neutron counting applied to safeguards and waste assay.

In this paper we consider the application of point model neutron multiplicity equations (see [3], [4] and references given there for details) and in particular how  $^{252}\text{Cf}$  of known emission rate can be used to characterize a passive neutron counter operating with shift register neutron correlation analyzer electronics. In particular we revisit the approximate form of the point model equations present in Appendix X1 (“Other Multiplicity Solutions”) of ASTM C1500-02 [5].

### *$^{252}\text{Cf}$ and Multiplicity Counting*

$^{252}\text{Cf}$ , which may be produced by the transmutation from curium oxide targets, emits about  $2.31 \times 10^6 \text{ n.s}^{-1} \cdot \mu\text{g}^{-1}$  and with a half-life of  $(2.645 \pm 0.008)\text{y}$  [6] this results in both a compact and convenient source of fission neutrons of well defined characteristics. The yield from other

nuclides can often be neglected or accounted for quite accurately [7], and the yield from ( $\alpha$ , n) reactions in the carrier and encapsulation is orders of magnitude lower. It is no surprise therefore that once  $^{252}\text{Cf}$  became readily available commercially in 1971 it found widespread use in a diverse variety of important applications [8]. The characterization and calibration of instrumentation under standard conditions is what concerns us here.

The Cf reference sources used for the characterization and calibration of correlated neutron counting system are typically lightly encapsulated in two capsules of stainless steel. During the characterization process the Singles, Doubles and Triples rates may be determined for a selection of sources of differing strength in order to evaluate the dead time correction parameters by relying on the fact that the properties of the fissioning system remain constant. Although an important practical use of Cf reference sources we shall defer the discussion of this aspect to another time.

The relative abundance of Cf isotopes depends on the production batch [9]. For the present discussion we need only be aware of approximate bounding values for purposes of guidance and we take the following ranges to be representative of freshly made material.

Isotope	Atom. %
$^{249}\text{Cf}$	1.8 - 6.7
$^{250}\text{Cf}$	9.0 - 11
$^{251}\text{Cf}$	2.8 - 3.3
$^{252}\text{Cf}$	80 - 86
$^{253}\text{Cf}$	<0.04
$^{254}\text{Cf}$	<0.04

The neutron output of a freshly manufactured commercially distributed sources is therefore usually utterly dominated by  $^{252}\text{Cf}$  and may reasonably be treated as characteristic of that fissioning system. As the source ages however it is well known that  $^{250}\text{Cf}$  comes into play [7] and we caution against using old sources or sources manufactured from aged recycled materials. Although when the relative isotopic composition of the source material is known an allowance for the decay in neutron output can be made, we note here that the contribution from other species is also quite noticeable (we believe, based on observation) in the ratio of correlated neutron rates.

The calibration of a passive neutron multiplicity counter has three adjustable parameters in addition to the dead-time coefficients. We shall assume here that the counts are distributed between multiple  $^3\text{He}$  proportional counter cluster/preamplifier-discriminator boards and marshaled into a fast shift-register multiplicity analyzer via a derandomizer circuit so that at rates below a few 100kHz dead time corrections are not accuracy limiting when suitable dead time coefficients are used. The three calibration parameters of interest are therefore [3] the neutron detection efficiency, the Doubles gate utilization factor and the Triples gate utilization factor (see [10,11] for a discussion of GUFs). The point-model interpretational equations used in multiplicity counting are well established [3] but may be refined [4] on a case by case basis to allow for geometry, energy and other dependences.

When counting Pu items it is conventional to express the correlated neutron signal in units of equivalent or effective grams of  $^{240}\text{Pu}$ ,  $^{240}\text{Pu}_{\text{eff}}$  because  $^{240}\text{Pu}$  is often the most dominant isotope for weapons and civil grade material. For our present purposes the basic nuclear data for  $^{252}\text{Cf}$  and  $^{240}\text{Pu}$  are therefore of most importance. The  $^{240}\text{Pu}_{\text{eff}}$  worth of  $^{238}\text{Pu}$  and  $^{242}\text{Pu}$ , the other two commonly encountered isotopes of Pu with significant spontaneous fission probability, has been treated elsewhere [12, 13]. Drawing on the compilation of Holden and Hoffman [14] we are led to estimates of the spontaneous fission (SF) half lives and Relative Standard Deviations (RSD) of:

$$^{252}\text{Cf}: (85.61 \pm 0.29) \text{ y (RSD} \sim 0.34\%)$$

$$^{240}\text{Pu}: (1.148 \pm 0.010) \times 10^{11} \text{ y (RSD} \sim 0.87\%)$$

The individual experimental data included in the compilation is often in conflict and scope undoubtedly exists to improve upon these half-life estimates given concerted effort to do so and the careful application of best techniques.

The neutron multiplicity distributions of  $^{252}\text{Cf}$  and  $^{240}\text{Pu}$  have been measured by several authors but great care is required to evaluate the data in a self-consistent fashion. Drawing on the outstanding work in this area of Holden and Zucker (see reference [15] and references therein) for inspiration we are led to make estimates of the first three prompt emission factorial moments as follows:

$^{252}\text{Cf}$ :

$$v_1 = (3.757 \pm 0.010)$$

$$v_2 = (11.9478 \pm 0.0068)$$

$$v_3 = (31.636 \pm 0.065)$$

$$\rho_{2,3} = 0.9948$$

$^{240}\text{Pu}$ :

$$v_1 = (2.154 \pm 0.005)$$

$$v_2 = (3.789 \pm 0.013)$$

$$v_3 = (5.210 \pm 0.067)$$

$$\rho_{2,3} = 0.8878$$

It is of interest to note that in general there are more measurements of  $v_1$  than there are of the full multiplicity distributions  $P(v)$ . Furthermore, a given experiment often deals with more than one nuclide so that as a rule the  $v_1$ -values are evaluated first and used to normalize the  $P(v)$  distributions of different experiments. This makes it impossible to evaluate the covariances between  $v_1$  and  $v_2$ , and,  $v_1$  and  $v_3$ , from the data available in [15]. The correlation coefficient  $\rho_{2,3}$ , is the ratio of the covariance between  $v_2$  and  $v_3$ , divided by the product of the respective standard errors (which are shown as the  $\pm$  uncertainty). It is clear that  $v_2$  and  $v_3$  are strongly correlated. It should also be clear that evaluating covariances between  $^{252}\text{Cf}$  and  $^{240}\text{Pu}$  systems is impossible from evaluated data.

It is a number of years since a concerted effort was placed on generating basic multiplicity data. The existing database represents a significant investment and it is unlikely that an improvement to these estimates can be made without a skilled and experienced team supporting the effort. The existing database of measurements can, however, be usefully augmented where new measurements are available and reviewed afresh with the particular aim of evaluating covariance terms. This is especially important because most of the expressions linking  $^{252}\text{Cf}$  and  $^{240}\text{Pu}$  multiplicity equations involve combinations of these basic nuclear parameters.

The neutron emission spectrum of  $^{252}\text{Cf}(\text{SF}, n)$  is reasonably well known [16] and although those of  $^{238, 240, 242}\text{Pu}$  are less so [17-19] and could gainfully be re-evaluated, the relative differences in efficiency are not large (a fraction of a percent relative) for a well designed PNMC and can be estimated by detailed transport calculation or by from the measured systematic trend of efficiency as a function of mean energy from a variety of broad spectrum isotopic sources. This is fortunate because it would seem from a survey of the available literature that experimental and theoretical evidence for how the hardness of the emission spectrum trends with mass number for a given element is inconclusive (e.g. whether the mean energy increases or decreases marginally as one progresses across the  $^{250}\text{Cf}$ ,  $^{252}\text{Cf}$  and  $^{254}\text{Cf}$  spontaneous fissioning systems, appears to be an open question within uncertainties). (See [20-23] for additional stimulation.)

The absolute yield of delayed neutrons emitted with period greater than about 0.25s following the spontaneous fission of  $^{252}\text{Cf}$  has been determined experimentally [24] to be approximately  $(0.0086 \pm 0.0011) \text{ n.fis}^{-1}$  where the uncertainty is quoted at  $1-\sigma$  (standard deviation). We take this to be a reasonable estimate of the overall yield since based on detailed analysis of delayed neutron precursor yields and properties [25] supports a value about  $(0.0069 \pm 0.15) \text{ n.fis}^{-1}$ . The corresponding delayed neutron yield for  $^{240}\text{Pu}$  is  $(0.0089 \pm 0.0011) \text{ n.fis}^{-1}$  again based on the summation method. This value agrees within uncertainties with other evaluations [25].

The  $^{241}\text{Am}/\text{Li}(\alpha, n)$  isotopic neutron source spectrum is a sufficiently close analog to the delayed neutron spectrum [26] for our purposes such that an emission calibrated Am/Li source can be used to determine the delayed neutron counting efficiency directly to adequate accuracy. As we shall come to see below the presence of delayed neutrons requires only a small addition to the multiplicity equations and so it may be handled adequately with existing nuclear data. The reason for this is twofold, first the delayed neutron fraction is small and second the ratio of the delayed neutron and prompt fission neutron detection efficiencies is in the vicinity of unity for appropriately designed multiplicity counters. In passing we note that the distributions of counts between the various rings of  $^3\text{He}$  proportional counters in the walls of a multiplicity counter also carry information about the emergent energy spectrum [27]. For point-like sources this may be a useful additional benchmark and diagnostic between Pu and Cf.

### ***Mathematical Results***

First let us review the approach used in the measurement of Pu mass. For cases where the sample self multiplication,  $M$ , can be taken equal to unity, the expressions for the Singles (S), Doubles (D), and Triples (T) rates are given by

$$S = \varepsilon F \nu_t (1 + \alpha) \quad (1)$$

$$D = \varepsilon^2 F f_d (\nu_2/2) \quad (2)$$

$$T = \varepsilon^3 F f_t (\nu_3/6) \quad (3)$$

Here  $f_d$  and  $f_t$  are the Doubles and Triples gate fractions respectively, and the  $\nu_i$ 's ( $i = t, 2, 3$ ) are the spontaneous fission factorial moments with  $\nu_t$ , the mean number of prompt plus delayed neutrons emitted per SF, given by

$$\nu_t = \nu_1 + \nu_d, \quad (4)$$

where  $\nu_d$  is the contribution from delayed neutrons.  $F$  is the SF rate,  $\varepsilon$  the unique neutron detection efficiency, and  $\alpha$  the relative ( $\alpha, n$ ) to (SF,  $n$ ) production rate.

The approach to obtaining the  $^{240}\text{Pu}$  effective mass ( $m_{\text{eff}}$ ),  $\varepsilon$ , and  $\alpha$ , from the measured values for  $S$ ,  $D$ , and  $T$ , is then captured in equations [5-7] below.

$$(1 + \alpha) = \frac{1}{(f_t/f_d^2)} \frac{ST}{D^2} \frac{(\nu_2/2)^2}{\nu_t(\nu_3/6)} \quad (5)$$

$$F = \left(f_t/f_d^2\right)^2 \frac{f_d D^3}{T^2} \frac{(\nu_3/6)^2}{(\nu_2/2)^3} \quad (6)$$

$$\varepsilon = \frac{1}{(f_t/f_d^2)} \frac{T}{f_d D} \frac{(\nu_2/2)}{(\nu_3/6)} \quad (7)$$

The choice of expression for the Triples to Doubles gate-fraction ratio will soon be evident.

The  $^{240}\text{Pu}$  effective mass  $m_{\text{eff}}$ , is obtained from  $F$ , the fission rate using

$$m_{\text{eff}} = \frac{F}{g}, \quad (8)$$

where the value for 'g', the specific fission rate (fissions  $\text{s}^{-1} \text{ gram}^{-1}$ ) is found in the nuclear data literature, and may be calculated using the half-life information presented earlier.

Neglecting the contribution from delayed neutrons (so that  $\nu_t = \nu_1$ ), and assuming the following relation for the gate fractions

$$f_t = f_d^2, \quad (9)$$

equations [5-7] reduce to the form given in Appendix XI of the ANSI C1500 standard [5] (equations [XI.1-XI.3]) shown below. Equation (5) becomes

$$\alpha = \frac{3ST}{2D^2} \frac{\nu_{s2}^2}{\nu_{s1}\nu_{s3}} - 1. \quad (10)$$

Equation (6) with a substitution for  $T$  in terms of  $(1+\alpha)$  from Eq. (10) becomes

$$F = \frac{S^2 f_d \nu_{s2}}{2D \nu_{s1}^2 (1 + \alpha)^2}. \quad (11)$$

Finally, creating an expression for T/D using equations (10) and (11) and substituting for this ratio in Eq. (7) gives

$$\varepsilon = \frac{S}{F\nu_{s1}(1+\alpha)}. \quad (12)$$

Equations [5-7] are thus the general forms of the ANSI C1500 standard equations given above ([10-12]), when the Doubles and Triples gate fractions are determined independently, and when the contribution from delayed neutrons is taken into account. Equation (9) holds approximately when the die-away profile of the counter is close to being a single exponential, but this behavior is never ideal because the structure of the chamber adds harmonics and the electronic counting chain exerts a further influence. It is our experience that  $f_t$  is best treated as an independent variable (i.e. better fits to calibration data are obtained).

We now carry over this approach in the use of  $^{252}\text{Cf}$  sources for the calibration of a neutron counter. As previously described the  $^{252}\text{Cf}$  sources can be considered as pure sources of spontaneous fission neutrons with sample self multiplication  $M = 1$ , and the ratio of ( $\alpha$ , n) to (SF, n) neutrons  $\alpha = 0$ . Consequently one can imagine the multiplicity equations describing the behavior of real Cf sources more accurately than they do real Pu items. Using Eq. (1) and recognizing the possible difference in efficiency  $\varepsilon$  for fission neutrons versus delayed neutrons, we have for the Singles rate

$$S = F(\varepsilon_F\nu_1 + \varepsilon_d\nu_d) \quad (13)$$

where we have also made use of the fission moment relation in Eq. (4). For  $^{252}\text{Cf}$  calibration sources the certification is generally in yield, Y, rather than fission rate F, so that  $F = Y/\nu_t$ , or

$$F = \frac{Y}{\nu_1 + \nu_d} \quad (14)$$

and Eq. (13) can be rewritten in terms of Y as

$$S = \varepsilon_F Y \left[ \frac{1 + (\varepsilon_d/\varepsilon_F)(\nu_d/\nu_1)}{1 + (\nu_d/\nu_1)} \right] \quad (15)$$

The Doubles and Triples rates follow similarly from Eqs. (2) and (3):

$$D = (\varepsilon_F^2 f_d) Y \left[ \frac{(\nu_2/2)}{\nu_1 + \nu_d} \right] \quad (16)$$

$$T = (\varepsilon_F^3 f_t) Y \left[ \frac{(\nu_3/6)}{\nu_1 + \nu_d} \right] \quad (17)$$

Equation (15) immediately yields a value for the efficiency  $\varepsilon_F$ :

$$\varepsilon_F = \frac{S}{Y} \left[ \frac{1 + (\nu_d/\nu_1)}{1 + (\varepsilon_d/\varepsilon_F)(\nu_d/\nu_1)} \right] \quad (18)$$

By substituting for  $\varepsilon_F$  in Eqs. (16) and (17), the Doubles and Triples gate fractions ( $f_d$ ,  $f_t$ ) can be obtained explicitly from the Doubles and Triples rates respectively, but  $f_d$  and  $f_t$  by themselves are not of fundamental interest. We choose instead to use Eqs. (16) and (17) to write

$$(\varepsilon_F^2 f_d) = \frac{D}{Y} \left[ \frac{\nu_1 + \nu_d}{(\nu_2/2)} \right] \quad (19)$$

and

$$(\varepsilon_F^3 f_t) = \frac{T}{Y} \left[ \frac{\nu_1 + \nu_d}{(\nu_3/6)} \right] \quad (20)$$

In these expressions D and T are measured parameters, and by dividing the count into a series of shorter intervals the counting precision and covariances can be extracted empirically. Y is the certified yield as determined absolutely by Mn-bath measurements and the remaining parameters are obtained from nuclear data and are known to the levels discussed earlier. Equations (19) and (20) can now be used in the mass calibration of a neutron counter using either the measured Doubles or Triples rates. Coming full cycle to Eq. (2) and making use of Eq. (8) we define the specific Doubles calibration coefficient for non-multiplying (dilute) Pu.

$$a_{D,Pu} = \frac{D}{m_{eff}} \quad (21)$$

where the Doubles mass-calibration constant is given by

$$a_{D,Pu} = g_{Pu} (\varepsilon^2 f_d)_{Pu} (\nu_2/2)_{Pu} \cdot \quad (22)$$

Substituting for the term  $(\varepsilon^2 f_d)$  obtained from the  $^{252}\text{Cf}$  calibration in Eq. (19) we have

$$a_{D,Pu} = g_{Pu} \left( \frac{\varepsilon_{Pu}}{\varepsilon_F} \right)^2 \left[ \frac{D}{Y} \frac{(\nu_1 + \nu_d)}{(\nu_2/2)} \right]_{Cf} (\nu_2/2)_{Pu} \quad (23)$$

where the term  $(\varepsilon_{Pu}/\varepsilon_F)$  accounts for the relative difference in efficiencies between  $^{240}\text{Pu}$  and  $^{252}\text{Cf}$ , and for the Doubles gate fraction it is assumed that  $f_d = f_{Pu} = f_{Cf}$ .

Typically for waste applications we use linear calibrations. Determining the Doubles (or Reals) calibration is crucial and is the focus of this discussion. The Singles rate is often not treated quantitatively because  $\alpha$  is difficult to estimate *a priori* except for a pure Pu product. The Triples rate is used to interpret low-level cosmic ray background suppression or test for Cm/Cf contamination. Though not presented here, the Triples mass-calibration constant can be expressed in the same way as in Eq. (23), by substituting for  $(\varepsilon^3 f_t)$  using Eq. (20) in the expression for Triples given in Eq. (3).

The usefulness of the expression in Eq. (23) is that it shows the interrelationship between the measured data and the evaluated parameters explicitly, and in deriving meaningful uncertainties it is important to work from the base equations and to include correlations appropriately. An

understanding of the limitation in calculating  $a_{D,Pu}$  can be readily obtained by examining the extent to which the constituents are known.

The relative uncertainties for the parameters obtained from nuclear data discussed earlier are  $\sim 0.34\%$  for  $(v_2/2)_{Pu}$ ,  $\sim 0.06\%$  for  $(v_2/2)_{Cf}$ , and  $\sim 0.27\%$  for  $(v_1)_{Cf}$  based on the uncertainty in  $(v_1)_{Cf}$ . The specific fission rate  $g_{Pu}$  is less well known at  $\sim 0.87\%$  and is dominated by the uncertainty in the SF half-life. The measurement of the Doubles rate,  $D$ , depends only on statistics and deadtime and can often be reduced to  $< 0.1\%$ . The certified yield of the  $^{252}Cf$  can vary from  $\sim 0.3$ - $1.0\%$  depending on the origin of certification. The approximation  $f_d = f_{Pu} = f_{Cf}$  introduces negligible bias compared to other sources of uncertainty. The evaluation of  $\epsilon_{Pu}/\epsilon_F$  is somewhat problematic but can be estimated with the aid of a Monte Carlo transport code using knowledge of the nuclear spectrum, neutron interactions, and chamber design. Ring-ratio data may help in refining the modeling. In general a well designed PNCC has a flat efficiency profile and  $\epsilon_{Pu}/\epsilon_F$  is close to unity. The difference in mean energy between  $^{240}Pu$  and  $^{252}Cf$  is  $\sim 200$  keV with an uncertainty of  $\sim 30\%$ . The uncertainty in the *deviation* of  $\epsilon_{Pu}/\epsilon_F$  from unity is therefore only known to  $\sim 30\%$  if we interpolate on an efficiency vs mean energy trend line. So if  $(\epsilon_{Pu}/\epsilon_F) = 1.015$ , we expect the uncertainty to be  $\sim 0.005$  to first order (i.e.  $\sim 0.5\%$  relative) less for a counter with a flatter efficiency. As discussed previously, this is an area where greater attention is needed in the investigation of spectral differences.

Combining these contributions in quadrature and, for the reasons discussed above, ignoring correlations, the resultant overall uncertainty in  $a_{D,Pu}$  is found to be in the approximate range  $1.1\% - 1.5\%$ . As indicated the uncertainties on the specific spontaneous fission rate of  $^{240}Pu$ , the determination of the emission rate of the Cf reference sources and the relative spectral difference between the principal isotopes are major contributors. We believe that these uncertainties can be reduced significantly by future re-evaluations and experimentation.

### ***Conclusions and Discussion***

This paper addresses the use of  $^{252}Cf$  in estimating the Pu parameters required for neutron multiplicity counters. We formulate the problem in a convenient way amenable to practical implementation and include a discussion of the uncertainties involved. The basic nuclear data required has either been provided or referenced. The availability of Pu for calibrating neutron counters is limited and restrictions on use and transport tightening. Reliance on  $^{252}Cf$  for initial calibrations, performance testing and benchmarking of calculations is consequently of growing importance. In this work we have extended the discussion given in the optional part of the standard ASTM C 1500 to show how best to extract the sought after parameters for multiplicity counting.

$^{252}Cf$  provides a near ideal point like source of fission neutrons for the characterization and calibration of passive neutron multiplicity counters. For benchmarking of sophisticated calculational radiation transport tools the uncertainties in the emission spectrum and the uncertainty to which the absolute neutron output can be determined at National Laboratory facilities is an important part of the overall uncertainty budget. It is important to stress that for demanding applications such as those described here there is a need for effort to reduce these

basic uncertainties to be ongoing. The other important source of uncertainty is in how well the materials and dimensions of the physical instrument can be assessed and entered into the model.

Modern multiplicity counters offer low background rate, high detection efficiency, low dead-time and excellent long-term stability (<0.1%) over years of operation. For neutron sources up to a few  $1 \times 10^5$  n.s<sup>-1</sup> (say) they offer the potential to measure Doubles-to-Singles and Triples-to-Doubles emission rates etc. with exquisite precision sufficient to distinguish between aged Cf-sources that differ in isotopic composition. It is therefore important to use fresh materials and controlled substrates and consistent encapsulation when specifying Cf-sources for high accuracy work.

We see no compelling evidence why gate utilization factors determined using <sup>252</sup>Cf cannot be adopted for all practical purposes for the measurement of compact spontaneously fissile Pu items. However efficiencies need correcting for spectral differences and here data for <sup>238</sup>, <sup>240</sup>, <sup>242</sup>Pu is found to be wanting and we appeal to the international community to address this issue.

If we are to use <sup>252</sup>Cf as a surrogate for Pu reference materials and exploit the full potential of the multiplicity equations presented here then at present the ratios of half-lives and prompt neutron multiplicity-moments data is limiting. In principle, however, it is possible to make a far more precise determination of the <sup>240</sup>Pu<sub>eff</sub> worth of <sup>252</sup>Cf by direct means similar to those applied to <sup>238</sup>Pu and <sup>242</sup>Pu [12, 13].

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