LA-UR- 04-8040

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Title: **Revised Prompt Neutron Emission Multiplicity Distributions for ^{236,238}Pu**

Author(s): **P. A. Santi, D. H. Beddingfield, and D. R. Mayo**

Submitted to: **For Informal Distribution**

(TECHNICAL REPORT)

Los Alamos
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Revised Prompt Neutron Emission Multiplicity Distributions for $^{236,238}Pu$.

P. Santi, D. Beddingfield, and D. Mayo
Safeguards Science and Technology Group (N-1)
Los Alamos National Laboratory, MS E540
Los Alamos, NM 87544.

The prompt neutron emission multiplicity distributions and average values (P_ν and $\langle\nu\rangle$, respectively) for nuclei which decay via spontaneous fission have been re-evaluated. In the cases of ^{236}Pu and ^{238}Pu, inconsistencies were found in the recommended values for P_ν and $\langle\nu\rangle$ that were given in the most recent compilation of neutron emission multiplicity distributions from the fission of Uranium and TransUranium nuclides [1]. In correcting for these inconsistencies, the values of $\langle\nu\rangle$ for ^{236}Pu and ^{238}Pu have now been revised to 2.07±0.13 and 2.19±0.07 respectively. The corresponding neutron emission probabilities for these two nuclei have also been revised in order to take into account the new recommended $\langle\nu\rangle$ values.

I. Introduction

Neutron multiplicity counting is an important nondestructive assay technique for performing safeguard measurements of the mass of plutonium in impure materials [2]. This technique relies on the fact that the number of neutrons that are emitted in the spontaneous fission of the even mass Pu isotopes is statistical in nature and well known. In order to continue the development of methods of standardizing the instrumentation used to perform neutron multiplicity measurements, as well as to continue to address issues of measurement bias and precision that occur when assaying Pu metal or impure items, Monte Carlo calculations are needed to study the various effects of sample multiplication, geometry, and (α,n) reactions from impurities on the measured moments of the neutron multiplicity distributions. The code that is used most extensively at Los Alamos National Laboratory for these calculations is the particle transport code MCNPX [3] which models the transport of neutrons through a given physical setup based on the known microscopic physical processes that emit neutrons as well as the point wise cross section data which describes the interactions between the neutron and the medium. The accuracy of these calculations is dependent in part on the accuracy of the neutron emission multiplicity distributions, P_ν, which describe how probable a given number of neutrons will be emitted from a given fission event, as well as the average number of prompt neutrons emitted, $\langle\nu\rangle$, for the various relevant nuclei. To ensure the accuracy of these neutron emission multiplicity distributions for nuclei which decay via spontaneous fission, a review of current status of these distributions was undertaken prior to their inclusion into MCNPX.

II. Method of Revising P_ν Data Sets

A previous set of compilations of P_ν and $\langle\nu\rangle$ for a wide range of nuclei had been done by Holden and Zucker in the mid 1980's [1, 4-5]. In these compilations, corrections were made to the various sets of P_ν that were available in the literature to compensate for the fact that the consensus value of $\langle\nu\rangle$ for various nuclei has improved over time. The neutron emission multiplicity distributions for a given nucleus are related to the average number of neutrons emitted by:

$$\sum \nu P_\nu = \langle \nu \rangle \tag{1}$$

where ν is the number of neutrons emitted per fission. It is evident from equation (1) that a change in the value of $\langle\nu\rangle$ for a given nucleus requires a subsequent change in the set of P_ν values. Since $\langle\nu\rangle$ can be determined independently and with greater accuracy than P_ν, the detection efficiency, ε, of the various neutron detector systems that were used to measure neutron emission probabilities were often determined based on a calibrating nuclide with a well known <ν> using the relationship:

$$g = \varepsilon \langle \nu \rangle q \qquad \textbf{(2)}$$

where q is the fission rate of the sample of the calibrating nuclide and g is the gross measured count rate from the calibration sample. Changes in the values of <ν> for the nuclides that were used to originally calibrate the neutron detector will subsequently affect both the values of <ν> that was measured in a given experiment as well as the neutron emission probabilities. The values of <ν> for the calibrating nuclei used in the original measurements were often quoted along with the measured values for <ν>.

In correcting the neutron emission probabilities for these changes in the average neutron multiplicity, Holden and Zucker developed a method of first reconstructing the measured probabilities of actually observing n neutrons from the fission of a given nuclide (Q_n) based the published values of the neutron detection efficiency ε, and the neutron emission probabilities (P_ν) through the relationship:

$$Q_n = \sum_{\nu} P_\nu \cdot \left[\frac{\nu!}{n!(\nu - n)!} \right] \cdot \varepsilon^{n} \cdot (1 - \varepsilon)^{\nu - n} \qquad \textbf{(3)}$$

In order to reconstruct a set of P_ν that was consistent with the updated value of <ν> for that nucleus as well as consistent with the originally measured set of Q_n values, the quoted neutron detection efficiency for a given experiment was varied until the values for P_ν satisfied equation (1) for the updated value of <ν>. The relationship between P_ν and the measured Q_n values and neutron detection efficiency is given by inverting equation (3):

$$P_\nu = \sum_{n} Q_n \cdot \left[\frac{n!}{\nu!(n - \nu)!} \right] \cdot \varepsilon^{-n} \cdot (\varepsilon - 1)^{n - \nu} \qquad \textbf{(4)}$$

III. Revised <ν> for ^{236}Pu and ^{238}Pu

In the cases of ^{236}Pu and ^{238}Pu, the only simultaneous measurement of both <ν> and P_ν was performed by Hicks, et al. [6]. This measurement used a ^{240}Pu sample as the reference for determining the efficiency of the neutron detector as well as the absolute values of <ν> for the nuclei of interest. An original set of corrections were performed on the data sets for ^{236}Pu and ^{238}Pu by Holden and Zucker using a consensus value <ν> of 2.140±0.005 for ^{240}Pu [4]. In a later compilation [1], the recommended value for ^{240}Pu was revised to 2.154±0.005 taking into account a recent set of measurements by Boldeman, et al. [8]. While the recommended value of <ν> had changed for ^{240}Pu in the latest compilation, the values of <ν> for ^{236}Pu and ^{238}Pu had not been subsequently revised. Thus the revised values for ^{236}Pu and ^{238}Pu from Ref. [6] were inconsistent with the quoted recommended value for ^{240}Pu. In Table 1, we present the corrected values for <ν> for these two nuclei from the measurements presented in Ref. [6]. The corrections amounted to a 1.5% and 0.5% increase in the value of <ν> for ^{236}Pu and ^{238}Pu, respectively, relative to the revised values that were presented in latest compilation [1]. As had been done previously by

Table 1: Revised <ν> for ^{236}Pu and ^{238}Pu

Nuclide	Reference	Cited Value	Revised Value in Ref. [1]	Corrected Value
^{236}Pu	[6]	2.305±0.19	2.17±0.19	2.20±0.19
	[7]	1.89±0.2	-	1.93±0.2
	Consensus[(a)]	-	2.17±0.19	**2.07±0.14**
^{238}Pu	[6]	2.33±0.08	2.21±0.08	2.22±0.08
	[7]	2.04±0.13	-	2.10±0.13
	Consensus[(a)]	-	2.21±0.08	**2.19±0.07**

(a) Consensus values and error bars were calculated by taking a weighted average of the revised values from Refs. [6] and [7].

Holden and Zucker, the quoted errors on the revised values are simply the originally quoted errors from Ref. [6].

An additional inconsistency was noticed in Ref. [1] in the determination of the consensus values for these two plutonium nuclei relative to the determination of consensus values for other nuclei that were presented in the compilation. A second measurement of <ν> had been made for ^{236}Pu and ^{238}Pu, as well as for ^{240}Pu, ^{242}Pu and ^{242}Cm, by Crane, et al. [7] using a very similar experimental setup to the one used in the Hicks experiment. While the two experiments used very similar experimental techniques, the results from Ref. [7] for the Pu nuclei had not been included in the compilations to determine the consensus values of <ν> for these nuclei even though the results for ^{242}Cm from the Crane experiment was used in the compilations. Because the same experimental technique for all of the nuclei that were measured by Crane, it is unclear why only the results for ^{242}Cm were used in the compilation. In order to remain consistent in terms of using data from that experiment to determine recommended values of <ν>, the data for ^{236}Pu, ^{238}Pu and ^{242}Pu have been revised and included in the calculations for determining a new consensus value for these nuclei.

The experimental data from Ref. [7] utilized a two-point calibration based on ^{252}Cf and ^{244}Cm to convert relatively measured <ν> values into absolute <ν>. One potential issue with using this method can be seen in Figure 1 where the original absolute <ν> is plotted as function of the measured relative <ν> for all of the nuclei studied in Ref. [7]. Due to the fact that the nuclei used to calibrate the relationship between the absolute and relative <ν> values lie at large <ν> values relative to the other nuclei studied, it was necessary to extrapolate this relationship to a region where no calibrating nuclei existed inducing a systematic error. In revising the data, the impact of extrapolating the relationship between the measured relative <ν> and the absolute <ν> was minimized by using ^{240}Pu as a third calibration point. The fact that the absolute <ν> for ^{240}Pu is well known [1] and was much closer to the nuclei of interest, the amount of extrapolation needed to determine the corrected absolute <ν> for ^{236}Pu and ^{238}Pu was considerably reduced. Based on a linear fit of the revised data points for ^{252}Cf, ^{244}Cm and ^{240}Pu, new absolute values for <ν> were determined for ^{236}Pu, ^{238}Pu, ^{242}Pu, and ^{242}Cm and are presented as the solid blue squares in Fig. 1. In the case of ^{242}Cm, the change in the revised value from 2.48±0.11 [5] to 2.41±0.11 did not affect the consensus value of <ν> for this nucleus as determined by calculating the weighted average of all of the previous measurements. Similarly, for ^{242}Pu, the inclusion of the revised <ν> value of 2.33±0.16 into the weighted average calculation had no impact on the on the resulting consensus value for ^{242}Pu due the relatively large error bars that were originally assigned to this measured value. However, with only one other measurement of <ν> for ^{236}Pu and ^{238}Pu present

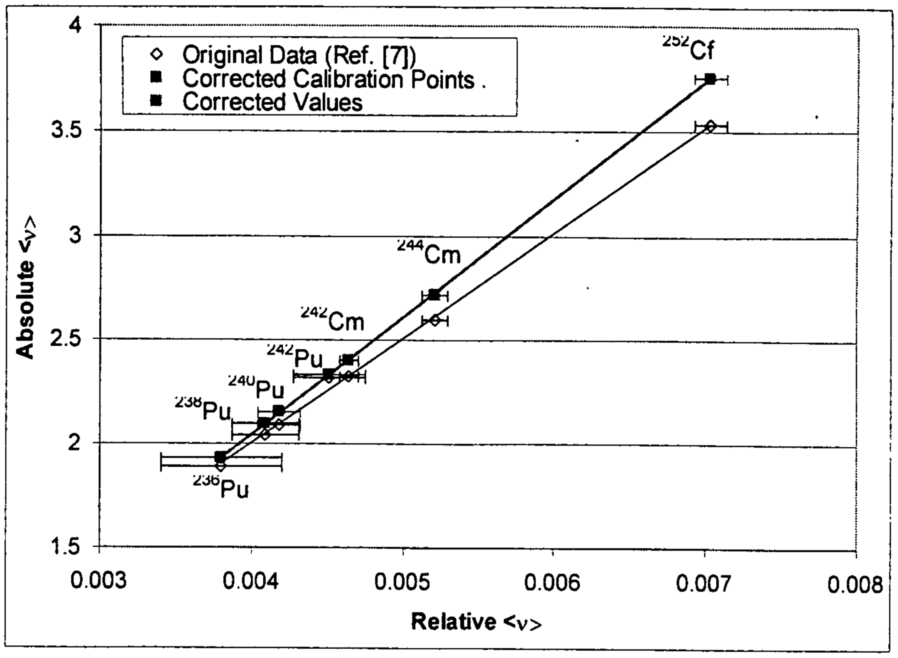

Figure 1: Absolute <ν> vs. Relative <ν> for the original data points in Ref. [7] (open circles) and the revised data points (solid squares). The black solid line is the linear fit to the two calibration points used in the original data set (^{252}Cf and ^{244}Cm) while the pink solid line is the linear fit to the three calibration points used to revise the data. The new calibration curve was based on the revised values of <ν> for ^{252}Cf (3.757±0.010) [5], ^{244}Cm (2.72±0.02) [5], and ^{240}Pu (2.154±0.005) [1].

in the literature, the inclusion of the revised <ν> values from Ref. [7] into the determination of the consensus values had a huge impact on the recommended values for these two nuclei.

The original and revised values of <ν> from Ref. [7] for ^{236}Pu and ^{238}Pu are presented in Table 1. The revised values from Ref. [7] are in good agreement with the revised values from Ref. [6]. This good agreement between the two sets of data allows us to take a weighted average of the two data points in order to determine consensus values and error bars for the two nuclei. The consensus values for ^{236}Pu and ^{238}Pu are presented in Table 1 along with the originally recommended values for these nuclei from Ref. [1]. The new recommended values for ^{236}Pu and ^{238}Pu are 5% lower and 1% lower, respectively, than the previously recommended values. The relatively large change in the recommended values of <ν> necessitates revising the sets of P_ν values for these two nuclei.

IV. Revised P_ν sets for ^{236}Pu and ^{238}Pu

In revising the P_ν sets from Ref. [6] for ^{236}Pu and ^{238}Pu, a slightly different method was used than the one used by Holden and Zucker in their compilations. Because the observed neutron multiplicity distributions for the various nuclei had been published in Ref. [6] without corrections for resolving time and background issues, it was possible to directly determine the set of Q_n values that were used to derive the published values of P_ν sets rather than reconstruct the Q_n values from the published P_ν values. The benefit of deriving the Q_n values from the raw data rather than reconstructing them is that one can exactly determine the neutron detection efficiency that was used to determine the P_ν values for that particular nucleus rather than rely on the published average neutron detection efficiency. This removes some of the ambiguity which is associated with the reconstructed Q_n values due to the fact that in Ref. [6], the author mentions that for the later runs the measured neutron detection efficiency was a few percent lower than for the earlier runs and only quotes what the measured absolute neutron detection efficiency was at the time of the ^{240}Pu run. Hence, by only using the quoted neutron detection efficiency to reconstruct the Q_n values, a systematic uncertainty can be introduced in revising the P_ν values due to the uncertainty in the actual neutron detection efficiency that was present at the time the nucleus was measured in the experiment.

While the resolving time correction (correcting for the fact that a single pulse may in fact contain two pulses) was explicitly stated in Ref [6], the stated correction calculation for the background in the publication was found to be erroneous due to the fact that, when rearranged, the uncorrected measured neutron multiplicity distribution would be equal to the 'corrected' measured neutron multiplicity distribution indicating that in fact no correction had been applied to the data. To properly correct the observed neutron multiplicity distributions for the published background rates, the correction method developed by Diven, et al. [9] was used which breaks down the observed neutron multiplicity distribution (Q'_n) of measuring n neutrons into the combination of the probability of observing x neutrons emitted from the fissioning nucleus (Q_x) and the probability of observing (n-x) background neutrons (B_{n-x}):

$$Q'_n = Q_0 \cdot B_n + Q_1 \cdot B_{n-1} + \ldots + Q_n \cdot B_0 \qquad \textbf{(5)}$$

The probability of observing n background neutrons in a correlation time, t_c, with a background rate b is given by:

$$B_n = \frac{(bt_c)^n \cdot e^{-btc}}{n!} \qquad \textbf{(6)}$$

Once the observed neutron probabilities had been corrected for background, the P_ν values were calculated based on the published neutron detection efficiency for the detector used in Ref. [5]. By varying the neutron detection efficiency, a set of P_ν values were produced which differed from the original published P_ν values by no more than 1%.

Having determined the Q_n values and neutron detection efficiencies that were originally used to derive the original sets of P_ν values for ^{236}Pu and ^{238}Pu, a revised set of P_ν values were determined that satisfied equation (1) for the new recommended $\langle\nu\rangle$ using the original Q_n values. Table 2 presents the revised sets of P_ν for ^{236}Pu and ^{238}Pu, along with original sets that were presented in Ref. [6] as well as the originally revised sets from the Holden and Zucker compilation [1]. While the neutron emission probability distributions are themselves interesting, the important values in terms of safeguards are the 1st, 2nd, and 3rd moments of the distributions ($\langle\nu\rangle$, $\langle\nu(\nu-1)\rangle$, and $\langle\nu(\nu-1)(\nu-2)\rangle$, respectively) which are related to the numbers of single neutron events, double neutron events, and triple neutron events that one measures in neutron multiplicity counting [2]. The moments of the original and revised probability distributions are presented in Table 2. The new 2nd and 3rd moments for ^{236}Pu are 9% to 12% lower than the moments presented in Ref. [1], while the 2nd and 3rd moments for ^{238}Pu are only 2% to 3% different than the originally revised values. It should be noted that while the changes in the neutron emission probabilities are quite large for ^{236}Pu, the current values are still within 1 σ of the originally measured values due to the large statistical error bars associated with the original measurement.

Table 2: Original and Revised P_ν values and moments for ^{236}Pu and ^{238}Pu

	^{236}Pu			^{238}Pu		
	Original	**Ref. [2]**	**Corrected**	**Original**	**Ref. [2]**	**Corrected**
P_0	0.062 ± 0.035	0.0706805	0.0802878	0.044±0.009	0.0540647	0.0562929
P_1	0.156 ± 0.09	0.1862416	0.2126177	0.175±0.026	0.205358	0.2106764
P_2	0.38 ± 0.13	0.3795474	0.3773740	0.384±0.026	0.3802279	0.3797428
P_3	0.28 ± 0.12	0.2545524	0.2345049	0.237±0.027	0.2248483	0.2224395
P_4	0.096 ± 0.086	0.0838837	0.0750387	0.124±0.021	0.1078646	0.1046818
P_5	0.033 ± 0.036	0.0250943	0.0201770	0.036±0.009	0.0276366	0.0261665
$\langle\nu\rangle$	2.305	2.17 [(a)]	2.07 [(a)]	2.33	2.21 [(a)]	2.19 [(a)]
$\langle\nu(\nu-1)\rangle$	4.252	3.7949	3.4658	4.398	3.9567	3.8736
$\langle\nu(\nu-1)(\nu-2)\rangle$	5.964	5.0462	4.4186	6.558	5.5960	5.4170
$\langle\nu^2\rangle$	6.557	5.9649	5.5377	6.728	6.1667	6.0607
$\langle\nu^2\rangle-\langle\nu\rangle^2$	1.2440	1.256	1.2448	1.2991	1.2826	1.2775
$\langle\nu(\nu-1)\rangle/\langle\nu\rangle^2$	0.8017	0.8059	0.8073	0.8099	0.8101	0.8099

(a) The P_ν data sets were made to conform to this value.

V. Summary

A review of the current status of the neutron emission probabilities for nuclei which decay by spontaneous fission has been completed. While most of the recommended values for $\langle\nu\rangle$ and P_ν in the most recent compilations by Holden and Zucker [1,5] have been independently verified, corrections to the recommended values for ^{236}Pu and ^{238}Pu have been performed due to inconsistencies that have been found with these values. The corrections have resulted in a 5% and 1% decrease in the recommended values of $\langle\nu\rangle$ for ^{236}Pu and ^{238}Pu, respectively.

VI. References

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