NEUTRON TRIPLES COUNTING DATA FOR URANIUM

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ABSTRACT

Correlated neutron counting using multiplicity shift register logic extracts the first three factorial moments from the detected neutron pulse train. The descriptive properties of the measurement item (mass, the ratio of (α, n) to spontaneous fission neutron production, and leakage self-multiplication) are related to the observed singles (S), doubles (D) and triples (T) rates, and this is the basis of the widely used multiplicity counting assay method. The factorial moments required to interpret and invert the measurement data in the framework of the point kinetics model may be calculated from the spontaneous fission prompt neutron multiplicity distribution $P(v)$. In the case of ^{238}U very few measurements of $P(v)$ are available and the derived values, especially for the higher factorial moments are not known with high accuracy.

In this work, we report the measurement of the triples rate per gram of ^{238}U based on the analysis of a set of measurements in which a collection of 10 cylinders of UO_2F_2 , each containing about 230 g of compound, were measured individually and in groups. Special care was taken to understand and compensate the recorded multiplicity histograms for the effect of random cosmic-ray induced background neutrons, which, because they also come in bursts, mimic fissions but with a different and harder multiplicity distribution. We compare our fully corrected (deadtime, background, efficiency, multiplication) experimental results with first principles expectations based on evaluated nuclear data. Based on our results we suspect the current evaluated nuclear data is biased, which points to a need to undertake new basic measurements of the ^{238}U prompt neutron multiplicity distribution.

KEYWORDS

nuclear safeguards; nondestructive assay; neutron multiplicity counting

1. INTRODUCTION

Neutron multiplicity counting [1] is widely used in nuclear safeguards for the assay of plutonium. Because the specific spontaneous fission rates are far lower the technique is not commonly used for uranium. However, it has been established [2,3,4] that with a suitable high efficiency detector, in low background conditions, quantities of typical safeguards concern can be counted with adequate precision in an acceptable time. In this case ^{238}U is the dominant source of spontaneous fission (SF) events. According to the most commonly applied method of autocorrelation time correlation counting, the observed singles (S), doubles (D) and triples (T) counting rates are related to the item parameters through the so call point-model multiplicity equations [1,5]. The factorial moments of the SF prompt neutron multiplicity distribution also enter into the point-model multiplicity equations. Thus, in order to be able to apply the equations quantitatively and with confidence the factorial moments of ^{238}U must be well known. Unfortunately very few measurements of the of the prompt neutron multiplicity distribution, $P(v)$, are available and the measurements that are available do not agree as well as one would like [6,7]. In this work we have approached the problem differently. Instead of trying to measure $P(v)$, as would be the goal of a basic science study, we have measured the quantities of applied interest, namely the S, D and T rates, from well characterized working reference materials using high performance Mini-Epithermal Neutron Multiplicity Counters (mini-ENMCs) [8].

2. REFERENCE MATERIALS

Ten tubes of well characterized UO_2F_2 were studied [9]. Each aluminum tube was 2.54 cm in outer diameter, 0.124 cm in wall thickness, and filled to about 21 cm with 230 g of anhydrous powder. The uranium enrichment was about 5 wt% and the purity, weights and so forth were

known by a battery of destructive analysis methods traceable to national standards and to an accuracy far superior to that of the neutron counting methods. These items were manufactured in order to calibrate nondestructive assay measurement systems at the Portsmouth, Ohio Gaseous Diffusion Plant (PORTS). Uranium holdup measurements in process equipment for nuclear criticality safety have been made at the PORTS by counting the gross neutrons produced from fluorinated compounds for many years [10]. During the certification process of neutron emission rate from this sub-set of reference items recently made we took advantage of the extensive destructive characterization to measure the correlated signatures as well.

3. REVIEW OF THE ^{238}U MULTIPLICITY DISTRIBUTION

Recently Santi and Miller [7] carried out a broad review of the prompt neutron emission probability distributions and average values for nuclides which undergo spontaneous fission. For ^{238}U they found the situation warranted no change from the previous comprehensive evaluation by Holden and Zucker [6]. Some eight determinations of the mean number of prompt neutrons emitted following fission (referred to as $\bar{\nu}$, and also commonly denoted by ν_1) are available for consideration. These guided Holden and Zucker to the choice of a recommended consensus value of (1.99 ± 0.03) (note however that in Table I of Santi and Miller the value given is 1.98 which we take to be a typographical error). For comparison based on Popeko et al's [11] measurement, the $\bar{\nu}$ value for ^{238}U is (0.943 ± 0.007) times that of ^{242}Pu , which taking $\bar{\nu}$ for ^{242}Pu to be (2.149 ± 0.008) [6, 7] returns a value of (2.026 ± 0.017) prompt neutrons per fission, numerically in agreement within the combined standard deviations. The value reported by Hwang et al [12] is relative to ^{240}Pu and is (1.96 ± 0.05) . There are only two determinations of the full prompt neutron emission probability distribution, $P(\nu)$. These are by the groups cited,

117 namely, Hwang et al [12] and Popeko et al [11]. The measurements are challenging because the
 118 specific spontaneous fission rate of ^{238}U is low so that long count times and careful background
 119 corrections are needed even when multi-plate fission chambers are used to provide the fission
 120 trigger for the neutron detector. After readjusting the $P(\nu)$ distribution of Hwang et al to the
 121 consensus value of $\bar{\nu}$, in their evaluation Holden and Zucker simply took the average of the
 122 two distributions (and also quantities derived from it) as the “best estimate” with the sample
 123 standard deviation (numerically equal to the absolute value of the difference divided by the
 124 square root of two) quoted as the associated uncertainty (note in Table II of Santi and Miller the
 125 uncertainty in $P(2)$ is given as 0.039 when it should be 0.0839 which we take to be a
 126 typographical error). Subsequently the Chinese group of Huang et al [13] (note the different
 127 spelling although it is the same group as [12]) published an English version of their 1974 work
 128 with the addition of an interesting discussion of Popeko et al’s measurement. Hwang et al used a
 129 somewhat conventional set-up comprising a large volume Cd-loaded liquid scintillation tank
 130 with an efficiency to ^{240}Pu neutrons of (0.691 ± 0.011) detections per neutron and a >95% chance
 131 of detection within a 30 μs coincidence gate following fission. The counting rate from the
 132 fission chamber was about 0.3/min and during the course of the experiment they accumulated
 133 3144 spontaneous fission events (although in their numerical results table, which we base the
 134 calculations discussed below on, the sum of events is only 3090). Popeko et al in contrast
 135 present results for three fission gated runs: 4750 fissions with a detector of efficiency
 136 (0.216 ± 0.006) , 8800 fissions with a detector of efficiency (0.264 ± 0.002) , and Run 3 with a
 137 detector of (0.383 ± 0.002) in which 42682 fissions were observed. It is well established that both
 138 high efficiency (>0.5 detections per neutron) and excellent statistical precision are generally
 139 needed if robust and physically meaningful multiplicity distributions are to be unfolded from the

observed data [14]. On this basis, and also because of its much larger sample size, Run 3 is of greatest statistical importance and is the one we shall adopt. It is also evident that whereas Popeko et al have a strong advantage in number of fissions observed, Hwang et al have a strong advantage in efficiency when it comes to probing the tail of the multiplicity distribution. Popeko et al also report a fourth run taken on a uranium block at a depth of 1100 mwe (meter water equivalent overburden) to reduce the cosmic ray induced neutron background with the 0.264 efficient detector. For this dataset there is no fission trigger and it is not clear how the ‘internal start’ mode can be used to create a observed neutron number distribution for multiplicity 2 to 5 to supplement the fission triggered data sets for multiplicity 0 to 5. Because of this we are discouraged from using Popeko’s Run 4 data although at face value it looks to be statistically the best dataset of the four. It is unfortunate that Popeko et al did not describe the method of data reduction so that this data set could be used to augment the other runs.

For our nuclear safeguards applications we are, however, less concerned with the ^{238}U multiplicity distribution than we are with the first two factorial moments ν_2 and ν_3 [7]. With only two measured distributions it is difficult to assign uncertainties based on a simple statistical analysis. In this work we therefore take a different slant. To avoid having to readjust and unfold the raw distributions (for which the publications do not give adequate information, for instance the fraction of events falling in the fission triggered gate or the ^{242}Pu or ^{240}Pu reference nuclide experimental distributions used to establish the metrology scale of the measurements) we make use of what we shall call the generalized Diven shape parameter defined as $\Gamma_m = \nu_m/\nu_1^m = n_m/n_1^m$. This has the property of being independent of efficiency and so, as the expression indicates, may be calculated from either the unfolded $P(\nu)$ distribution or $Q(n)$ the recorded

distribution (corrected for interlacing, background and so forth). On this basis we have two experimental $Q(n)$ distributions to work with in order to extract the Diven parameters from the observed data. These are given in Table 1. Since ν_1 is available from separate evaluation we can then obtain the factorial moments from the relation $\nu_m = \Gamma_m \nu_1^m$.

Data source	n=0	1	2	3	4	5
Hwang et al	504	1206	1043	292	41	4
Popeko et al	18209	17522	5945	906	93	7

Table 1. $Q(n)$ distributions taken from Hwang et al [12,13] and Popeko et al [11]. Note $Q(4)$ for Popeko et al is wrongly given as 95 in reference [13].

For each of these two distributions, Γ_2 and Γ_3 was evaluated along with the ratio Γ_3/Γ_2 . To estimate the uncertainty on these derived parameters, each $Q(n)$ value was changed up and down by one standard deviation (assumed to be equal to the square root of the number of counts) and half the difference was propagated in quadrature as if they were independent variables. The results are summarized in Table 2.

Data source	Γ_2	Γ_3	Γ_3/Γ_2
Hwang et al	0.7195±0.0104	0.3447±0.0205	0.479±0.023
Popeko et al	0.7492±0.0067	0.428±0.016	0.571±0.018
Adopted	0.7404±0.014	0.3960±0.040	0.5360±0.045

Table 2. Derived ^{238}U multiplicity distribution quantities.

We see that the two measurements are of similar apparent quality (as judged by the estimated uncertainties) and that agreement between them is reasonably good. The adopted values listed are the weighted means and the associated uncertainties are the external standard errors (which are 2.4, 3.2 and 3.2 times larger than the internal standard errors for Γ_2 , Γ_3 , and Γ_3/Γ_2 respectively). Taking ν_1 to be (1.99±0.03) [6] we derive from the adopted values of Γ_2 , Γ_3 , and Γ_3/Γ_2 the following estimates for the prompt neutron multiplicity distribution of ^{238}U :

- $\nu_2 = 2.93 \pm 0.10$

- $\nu_3 = 3.12 \pm 0.35$

- $\nu_3/\nu_2 = 1.067 \pm 0.091$

For comparison, the values for ν_2 and ν_3 listed by Santi and Miller [7] and by Zucker and Holden [6] based on their choice of $P(\nu)$ evaluation as described earlier are (2.87 ± 0.14) and (2.82 ± 0.48) respectively. The present values, arrived at independently and using a distinct analysis of the best available data are in good agreement with these previous reviews within the rather large fractional uncertainties. From the width parameter reported by Belen-kii et al [15], a value for ν_2 of (2.94 ± 0.30) can be deduced. From a technical perspective, there is no experimental reason why these uncertainties cannot be reduced by a factor of three or more when one considers for instance the quality of ^{240}Pu or ^{242}Pu data as a benchmark [6,7]. There are hints from integral measurements [4] that the third factorial moment in particular requires re-measuring.

4. MULTIPLICITY COUNTING

The singles count rate is dominated by the yield of $F(\alpha, n)$ reactions and has been discussed in detail elsewhere [9]. Consequently we cannot use the singles rate to directly quantify ^{238}U for these UO_2F_2 samples. According to the commonly used point-model equations for multiplicity shift register (MSR) counting the deadtime and background corrected doubles and triples rates may be written as follows [1]:

$$D = F\varepsilon^2 f_d \frac{v_{S2}}{2} M^2 \left[1 + \left(\frac{M-1}{v_{I1}-1} \right) (1+\alpha) v_{S1} \frac{v_{I2}}{v_{S2}} \right]$$

$$T = F\varepsilon^3 f_t \frac{v_{S3}}{6} M^3 \left[1 + \left(\frac{M-1}{v_{I1}-1} \right) \left[3 \frac{v_{S2}v_{I2}}{v_{S3}} + (1+\alpha) v_{S1} \frac{v_{I3}}{v_{S3}} \right] + 3 \left(\frac{M-1}{v_{I1}-1} \right)^2 (1+\alpha) \frac{v_{S1}v_{I2}^2}{v_{S3}} \right]$$

202 where,
 203 D and T are the doubles and triples rates in units of dps and tps respectively;
 204 F is the spontaneous fission (SF) rate, fis.s^{-1} and for ^{238}U and is given by the product of the ^{238}U
 205 mass, m_{238} in grams, and the specific SF rate, g_S , in fis.g^{-1} , that is $F = m_{238}g_S$;
 206 $g_S = (\ln(2)N_A/A)/T$ where A is the molar mass in grams, N_A is Avogadro's constant, mol^{-1} ,
 207 and T is the SF half-life in sec. Holden and Hoffman [16] collate fourteen experimental results
 208 and based on our analysis of that data we select an value of T of $(8.421 \pm 0.068) \times 10^{15}$ yr (1 yr =
 209 365.25 days) resulting in a value for g_S of $(0.0065984 \pm 0.000053) \text{ fis.g}^{-1}.\text{s}^{-1}$.
 210 ε is the neutron detection efficiency for neutrons emerging from the item, cnt.n^{-1} ;
 211 M is the leakage self-multiplication of the item;
 212 α is the ratio of (α, n) -to-SF-neutron creation rate inside the item and is given by $\alpha = S_\alpha / (Fv_{S1})$,
 213 S_α being the (α, n) production rate;
 214 f_d is the doubles gate utilization factor (GUF), the fraction of doubles that fall within the finite
 215 gating structure;
 216 f_t is the triples GUF;
 217 v_{Sj} is the j^{th} factorial moment of the SF prompt-neutron multiplicity distribution;
 218 and,
 219 v_{Ij} is the j^{th} factorial moment of the induced fission (IF) prompt-neutron multiplicity distribution.
 220
 221 Delayed neutrons are a small contribution and are neglected here. The right-hand products of M
 222 raised to a power and the square brackets represent the enhancement in the detected correlated
 223 neutron rates due to self-multiplication. Based on previous work [9] we estimate for our UO_2F_2
 224 samples the average α value to be to be about 7.2 with a range of ± 0.2 , and M to be about 1.0027

with a range of <0.0002 and independent of neutron source reaction with roughly 20% of the induced fission taking place in ^{235}U and the rest in ^{238}U . For fast (1 MeV) fission we estimate, using the tables of Zucker and Holden [17], the values of ν_{I1} , ν_{I2} , and ν_{I3} to be about 2.449, 4.937, and 8.112 respectively. The exact choice of energy is not crucial since the induced fission factorial moments appear only in ratios. The characteristics of the multiplicity counter are well established. The detection efficiency for fission neutrons varies with mean energy and over the range of interest here (1.9 MeV for ^{238}U SF to 2 MeV for induced fission) the range is 0.636 to 0.628 with an absolute relative standard deviation of about 1.3%. Detailed sensitivity calculations have been undertaken using Monet Carlo methods [9]. The mid-range value is adopted with half the spread being added to the uncertainty, namely (0.632 ± 0.012) counts per neutron. f_d and f_t have measured values of 0.8349 and 0.7032 using sealed ^{252}Cf sources and using the fact that ^{252}Cf is a de facto reference multiplicity distribution. Based on these chamber parameters and our ^{238}U nuclear data review described earlier we have established an algebraic predictive capability for the correlation counting experiments to be described. An important point is that even though the self-multiplication is close to unity, the strong internal $F(\alpha, n)$ source term results in a significant enhancement of the doubles and triples rates by typical factors of about Φ_2 (1.0569 ± 0.0049) and Φ_3 (1.115 ± 0.012) times respectively with a ratio Φ_3/Φ_2 of (1.0552 ± 0.0070) (where the uncertainty takes into account correlations).

In this brief paper we will use the measured and evaluated value of the ratio ν_3/ν_2 as a measure of the shape multiplicity distribution which reduces the influence of a number of possible systematic effects.

$$\left(\frac{v_3}{v_2}\right) = \frac{3 T / (f_t \Phi_3)}{\varepsilon D / (f_d \Phi_2)}$$

We have shown how the multiplication corrections can be made to better than 0.5%, that the efficiency is known to better than 2%, and we believe the gate utilization factor ratio is also known to better than 1% by extrapolation of the MSR predelay and gate width settings.

5. DATA COLLECTION AND ANALYSIS

Each tube of uranyl fluoride was counted multiple times and in combination in one of two identical mini-ENMCs of the type illustrated in Figure 1. The second counter was used to track the ambient background which moves with atmospheric changes. Both MSR electronics (Canberra Industries model JSR-15) and a List Mode Multiplicity Module (LANL LMMM) were used to acquire the correlated pulse trains. A detailed offline analysis was used to extract the rates and to separate the signal from the nuclear decay from cosmic-ray induced spallation events. A combination of bespoke analytical models and Monte Carlo simulations using the general purpose radiation transport code MCNPX [18] were used to justify and place uncertainties on the approach. The data were collected in a basement laboratory of a LANL facility but Los Alamos is at an elevation of about 2.2 km above sea level and so, with these relatively low mass items, correction for the cosmic-ray contribution to the weak triples rate is significant and is a keen focus of our present work. It is a significant extension to our earlier work that reported only on the singles data for the calibration of holdup measuring equipment. It is also of general interest for counting close to detection limits.

Figure 1. Schematic of the mini-ENMC

6. RESULTS

Table 3. lists the results and from this the sought after specific (per unit mass) rates to be extracted.

WRM ID	Date	Count Time	Background Count Rates [cps]			Net Count Rates [cps]		
			Singles $\pm \sigma_s$	Doubles $\pm \sigma_D$	Triples $\pm \sigma_T$	Singles $\pm \sigma_s$	Doubles $\pm \sigma_D$	Triples $\pm \sigma_T$
02	12/12/2012	60s x 360 cycles	$5.052 \pm 0.32\%$	$0.160 \pm 3.98\%$	$0.099 \pm 10.6\%$	$12.81 \pm 0.27\%$	$0.639 \pm 2.00\%$	$0.112 \pm 13.1\%$
08	11/29/2012	60s x 360 cycles	$4.952 \pm 0.32\%$	$0.132 \pm 3.64\%$	$0.064 \pm 10.4\%$	$13.40 \pm 0.27\%$	$0.653 \pm 1.84\%$	$0.116 \pm 8.04\%$
09	11/28/2012	60s x 250 cycles	$4.949 \pm 0.40\%$	$0.144 \pm 5.01\%$	$0.093 \pm 14.7\%$	$13.70 \pm 0.29\%$	$0.663 \pm 2.23\%$	$0.105 \pm 16.6\%$
14	11/30/2012	60s x 300 cycles	$4.985 \pm 0.34\%$	$0.151 \pm 4.36\%$	$0.096 \pm 12.5\%$	$13.69 \pm 0.29\%$	$0.675 \pm 2.17\%$	$0.115 \pm 13.8\%$
17	12/18/2012	60s x 360 cycles	$5.091 \pm 0.30\%$	$0.153 \pm 3.73\%$	$0.085 \pm 10.6\%$	$13.49 \pm 0.26\%$	$0.653 \pm 1.93\%$	$0.114 \pm 10.5\%$
18	12/10/2012	60s x 360 cycles	$5.075 \pm 0.33\%$	$0.146 \pm 3.74\%$	$0.080 \pm 12.4\%$	$13.53 \pm 0.26\%$	$0.640 \pm 1.79\%$	$0.092 \pm 12.8\%$
20	12/4/2012	60s x 376 cycles	$4.907 \pm 0.45\%$	$0.124 \pm 5.46\%$	$0.071 \pm 15.3\%$	$14.25 \pm 0.27\%$	$0.677 \pm 1.66\%$	$0.093 \pm 12.4\%$
26	12/6/2012	60s x 390 cycles	$5.064 \pm 0.31\%$	$0.140 \pm 3.35\%$	$0.066 \pm 8.93\%$	$14.23 \pm 0.24\%$	$0.687 \pm 1.66\%$	$0.141 \pm 6.80\%$
35	12/3/2012	60s x 360 cycles	$4.983 \pm 0.30\%$	$0.135 \pm 3.54\%$	$0.066 \pm 10.2\%$	$14.24 \pm 0.24\%$	$0.658 \pm 1.70\%$	$0.117 \pm 7.85\%$
38	12/20/2012	60s x 360 cycles	$4.935 \pm 0.32\%$	$0.139 \pm 3.89\%$	$0.076 \pm 10.5\%$	$14.15 \pm 0.26\%$	$0.652 \pm 1.79\%$	$0.116 \pm 9.13\%$

Table 3. Summary of results

For each of the ten items the experimental value of ν_3/ν_2 was extracted as described earlier by applying item specific corrections. The arithmetic mean value obtained was 0.907 with a sample standard deviation of 0.10 and a standard error (estimate of the population standard deviation) of

0.033. The weighted mean was slightly higher at 0.9394. The internal and external standard errors were numerically similar at 0.033 and 0.034 respectively. The selected value of (0.9394±0.034) is to be compared with the evaluated nuclear data value performed above of (1.067±0.091). The ratio of this to our measured value, (1.136±0.11), is consistent with unity within the joint uncertainty (which includes the systematic uncertainty in the measurement chamber efficiency and ratio of the multiplication enhancement factors) – but the fractional uncertainty is rather large. The weighted mean ratio of (0.955±0.042) (external standard error) favors our experimental value.

7. CONCLUSION

New integral measurements of the factorial moments for ^{238}U spontaneous fission are reported. These are placed on an absolute scale with reference to the de facto standard ^{252}Cf . The specimens used are far from ideal being of relatively low mass and with a strong (α, n) contribution compared to the typical stored bulk oxide materials of common interest in nuclear safeguards. However the use of a high performance multiplicity counter in a shielded location together with a second matched counter to track the for cosmic-ray spallation background yielded data of with a accuracy that significantly improves on the state of current knowledge. Our evaluation of basic nuclear data and new experimental results indicate that the triples factorial moment derived from $P(v)$ tables is not known to adequate accuracy for nuclear safeguards accountancy. We advocate for new $P(v)$ measurements and suggest uncertainties in the third factorial moment can be readily reduced by a factor of 5 using modern experimental practice.

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