Nuclear Instruments and Methods in Physics Research A 784 (2015) 455–459

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Neutron triples counting data for uranium

Stephen Croft [a,*], Adrienne M. LaFleur [b], Robert D. McElroy Jr. [a], Martyn T. Swinhoe [b]

[a] Oak Ridge National laboratory, 1 Bethel Valley Road, Oak Ridge, TN 37831, USA
[b] Los Alamos National Laboratory, Los Alamos, NM 87545, USA

ARTICLE INFO

Available online 22 October 2014

Keywords:
Nuclear safeguards
Nondestructive assay
Neutron multiplicity counting

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 $P(\nu)$. In the case of ^{238}U very few measurements of $P(\nu)$ 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 and 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 that 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.

Published by Elsevier B.V.

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 [2–4] 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 called 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, $P(\nu)$, 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 $P(\nu)$, 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

* Corresponding author. Tel.: +1 865 241 2834.
E-mail address: crofts@ornl.gov (S. Croft).

http://dx.doi.org/10.1016/j.nima.2014.09.086
0168-9002/Published by Elsevier B.V.

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 that 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 nu-bar, $\bar{\nu}$, 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 1 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 nu-bar value for ^{238}U is (0.943 ± 0.007) times that of ^{242}Pu, which taking nu-bar 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, namely, Hwang et al. [12] and Popeko et al. [11]. The measurements are challenging because the specific spontaneous fission rate of ^{238}U is low so that long count times and careful background corrections are needed even when multi-plate fission chambers are used to provide the fission trigger for the neutron detector. After readjusting the $P(\nu)$ distribution of Hwang et al. to the consensus value of nu-bar, in their evaluation Holden and Zucker simply took the average of the two distributions (and also quantities derived from it) as the 'best estimate' with the sample standard deviation (numerically equal to the absolute value of the difference divided by the square root of two) quoted as the associated uncertainty (note that in Table 2 of Santi and Miller the uncertainty in $P(2)$ is given as 0.039 when it should be 0.0839 which we take to be a typographical error). Subsequently the Chinese group of Huang et al. [13] (note the different spelling although it is the same group as that of Ref. [12]) published an English version of their 1974 work with the addition of an intersting discussion of Popeko et al.'s measurement. Hwang et al. used a somewhat conventional setup comprising a large volume Cd-loaded liquid scintillation tank with an efficiency to ^{240}Pu neutrons of (0.691 ± 0.011) detections per neutron and a $>95\%$ chance of detection within a 30 μs coincidence gate following fission. The counting rate from the fission chamber was about 0.3/min and during the course of the experiment they accumulated 3144 spontaneous fission events (although in their numerical results table, on which we base the calculations discussed below, the sum of events is only 3090). Popeko et al. in contrast present results for three fission gated runs: 4750 fissions with a detector of efficiency (0.216 ± 0.006), 8800 fissions with a detector of efficiency (0.264 ± 0.002), and Run 3 with a detector of (0.383 ± 0.002) in which 42,682 fissions were observed. It is well established that both high efficiency (>0.5 detections per neutron) and excellent statistical precision are generally needed if robust and physically meaningful multiplicity distributions are to unfolded from the observed data [14]. On this basis, and also because of its much larger sample size, Run 3 is of the 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 efficiency 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 2–5 to supplement the fission triggered data sets for multiplicity 0–5. 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, it 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 $\nu_m = \Gamma_m \nu_1^m$.

For each of these two distributions, Γ_2 and Γ_3 were 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.

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_3 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:

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].

Data source	n=0	1	2	3	4	5
Hwang et al.	504	1206	1043	292	41	4
Popeko et al.	18,209	17,522	5945	906	93	7

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

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