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Prompt Neutron Emission Multiplicity Distributions and Average Values, $\bar{\nu}$, at 2200 Meter per Second for the Fissile Nuclides

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Abstract: The prompt neutron emission multiplicity distribution, P_ν, is of interest for methods of self-calibration and for auto-correlation to assay fissionable material for nuclear safeguards. $\bar{\nu}$, the average value of P_ν, is of interest at neutron thermal energies since it is related to the neutron multiplication factor and it is used as a normalizing point for energy dependent values of $\bar{\nu}$. Values of P_ν and $\bar{\nu}$ have been determined at the standard neutron energy of 0.0253 ev for the neutron induced fission of the four fissile nuclides, ^{233,235}U, and ^{239,241}Pu. Revised $\bar{\nu}$ values have been obtained by re-evaluating $\bar{\nu}$ experiments measured at 2200 meter/second relative to the $\bar{\nu}$ from the spontaneous fission of ^{252}Cf. These revised values of $\bar{\nu}$ have been used to renormalize the measured P_ν values. The revised values of $\bar{\nu}$ are all about 1/4% to 1/2% smaller than the corresponding values of ENDF/B-V.

($\bar{\nu}$, P_ν, E_n = 0.025 ev, ^{233}U, ^{235}U, ^{239}Pu, ^{241}Pu)

I. Introduction

The neutron emission multiplicity distribution, P_ν is the probability that a given fission results in the emission of ν neutrons. Self-calibrated instruments developed to assay fissionable material perform auto-correlation on the pulse stream of detected prompt neutrons from the sample. The amount of material involves the quantity:[1]

$$< \nu(\nu-1) > = \sum_\nu \nu(\nu-1)\, P_\nu. \qquad (1)$$

Higher moments, e.g., $<\nu(\nu-1)(\nu-2)>$, are useful in considering self-multiplication within an assay sample, since multiplication distorts the inherent P_ν distribution and its moments. The average value of neutrons emitted in a fission, $\bar{\nu}$, is not sufficient information. $\bar{\nu}$ is related to the neutron multiplication factor and measurement at thermal energies is necessary both for thermal reactors and as a normalizing point for energy dependent values of $\bar{\nu}$. The most careful experiments compared the $\bar{\nu}$ values from the thermal neutron fission of the fissile nuclides with the $\bar{\nu}$ value for the spontaneous fission of ^{252}Cf, taken as $\bar{\nu}$ = 3.757 ± 0.010 neutrons per fission from an earlier evaluation.[2] $\bar{\nu}$ ratio experiments on the fissile nuclides are reassessed by correcting for the effect of delayed gamma-rays, for the different mean energies, for the various fission neutron spectra involved, and for the loss of those events corresponding to fission fragments of low energy because of the thickness of the fission foil. Values for fissile nuclides' $\bar{\nu}$ renormalize the experimental P_ν values for these nuclides.

II. Generation of P_ν from Experiment

The efficiency, ε, of the neutron detector for the detection of a single neutron is less than unity. Allowance for neutrons emitted but not detected gives the probability Q_n of actually observing n neutrons, even if ν were emitted ($n < \nu$) being just:

$$Q_n = \Sigma\, P_\nu\, [\nu!/n!(\nu-n)!]\, \varepsilon^n\, (1-\varepsilon)^{\nu-n} \qquad (2)$$

and

$$P_\nu = \Sigma\, Q_n\, [n!/\nu!(n-\nu)!]\, \varepsilon^{-n}\, (\varepsilon-1)^{n-\nu} \qquad (3)$$

The detector efficiency is determined using a calibrating nuclide, with a known $\bar{\nu}$ value.

$$g = \varepsilon\, \bar{\nu}\, q, \qquad (4)$$

where q is the fission rate and g is the gross count rate for the calibrating nuclide. First, we determine the various prompt $\bar{\nu}$ values for the fissile nuclides and correct the efficiency of the P_ν experiments and then calculate a revised set of prompt P_ν values at thermal neutron energies.

III. Review of $\bar{\nu}$ Experiments of the Fissile Nuclides

Scintillator tanks are filled with a material with a large neutron capture cross section, e.g. gadolinium. Neutrons produced from fission enter the tank, are moderated and captured. The neutron capture gamma rays cause scintillations to be detected by photomultiplier tubes adjacent to the tank. The tube output is gated with the fission pulse to discriminate against background radiation. Scintillator tanks are sensitive to gamma rays emitted from the decay of isomeric levels in the various fission product nuclides. Some of these gamma-rays are delayed beyond the gating time of the photomultipliers, and the correction for this effect was taken from Boldeman's 1977 review[3] and Gwin's[4] measurement of all these nuclides. Boldeman measured only ^{235}U and ^{239}Pu and assumed that ^{233}U was identical to ^{235}U and ^{241}Pu was identical to ^{239}Pu. Gwin measured the effect in ^{235}U, ^{239}Pu and ^{241}Pu, and agreed with Boldeman. Gwin found the effect in ^{233}U to be twice as large as Boldeman's assumed correction. Boldeman's corrections were for all data except that of ^{233}U, where we doubled Boldeman's correction. We used Gwin's estimate of a 30 % uncertainty. Table I shows the percentage corrections for all effects to the various experiments, including other corrections to be discussed.

The efficiency of liquid scintillator tanks is dependent upon the energy of the neutron being moderated and captured in the tank. Each of the fissioning nuclides has a different energy spectrum of neutrons produced in a fission. The mean energies of the various fission spectra were determined in a recent evaluation[5].

The thickness of the fission foil can cause loss of events corresponding to fission fragments of low energy, which never escape from the foil. Low kinetic energy fission fragments correspond to the release of higher numbers of neutrons and therefore thick foils depress the $\bar{\nu}$ value as a result of the loss of these low energy and corresponding high neutron events. For the foil thickness correction, we used three different estimates: Gwin's[4] estimate for Gwin's measurement; Malinovskii's[6] estimate for the various Obninsk measurements and Boldeman's[7] estimate for all other measurements. We assumed a 50% uncertainty on this correction.

For some measurements, the $\bar{\nu}$ ratios were determined with a fissile nuclide fissioning by use of neutrons in the kev energy range and then extrapolated back to a thermal value by use of a constant slope curve, which had been fit between the value at thermal energies and at 2 Mev. It is not clear that application of this constant slope is appropriate for the neutron energy range of 30 to 60 kev. Data recorded by Gwin[8] and also by Prokhorova[9] would imply that the $\bar{\nu}$ ratio of $^{235}U/^{252}Cf$ is slightly smaller in the 50 to 100 kev energy region than at thermal neutron energies. Although the $\bar{\nu}$ ratio is definitely larger in the region 0.5–1.0 Mev, it appears to have a dip between the thermal energy region and the Mev range. As a result, we have chosen to assume that the $\bar{\nu}$ ratio between 30 to 60 kev is equivalent to the thermal ratio and have added the effect of the constant slope assumption as an uncertainty instead of a correction.

Table II compares our results with ENDF/B-V and with earlier evaluations of the prompt $\bar{\nu}$ ratios for these fissile elements. Using the recommended value for prompt $\bar{\nu}$ of ^{252}Cf and values for the delayed component of $\bar{\nu}$, Table III compares the present estimate for total $\bar{\nu}$ of these fissile nuclides with similar estimates from ENDF/B-V and earlier evaluations.

IV. Method of Comparing Different P_ν Sets

The Q_n and P_ν are vectors, whose components are probabilities, related by an operation and its inverse which converts one set of probabilities into the other, i.e. equations (2) and (3). Certain ratios of the various moments of the distributions are independent of the efficiency, e.g.:

$$< \nu\ (\nu-1)\ ....\ (\nu-k) > / \ \bar{\nu}^{k+1} =$$

$$< n\ (n-1)\ ...\ (n-k) > / \ <n>^{k+1}. \qquad (5)$$

Diven's parameter, for $k = 1$, $<\nu(\nu-1)> / \bar{\nu}^2$, can be considered as a measure of the shape of the P_ν distribution. Another indicator of the distribution's shape, which is not conserved, (independent of ε) is the ratio of the mean square deviation to the square of the mean:

$$< (\nu-\bar{\nu})^2 > / \ \bar{\nu}^2 = (< \nu^2 > - \ \bar{\nu}^2) \ / \ \bar{\nu}^2. \qquad (6)$$

Equations (2) and (3) were used with the quoted distribution P_ν and the reported efficiency ε to derive the original Q_n set for each experiment. The efficiency was varied until the calculated $\bar{\nu}$ (i.e. $\Sigma\ \nu\ P_\nu$) value was obtained corresponding to the recommended value. It should be noted that the efficiency, ε, is inversely proportional to the value of $\bar{\nu}$. The ratio of the new to the old efficiencies is the same as the ratio of the old to the new $\bar{\nu}$ values.

Although the experimenter requires the efficiency to derive the P_ν values from the measured Q_n values, we do not need the originally measured values of Q_n. The ratio of the efficiencies or of the $\bar{\nu}$ values are sufficient information for us to redetemine a corrected set of P_ν values.

After various sets of P_ν for the same nuclide are transformed to yield the renormalized value of $\bar{\nu}$, any remaining differences between the corresponding P_ν can be ascribed to systematic errors other than those in ε or $\bar{\nu}$, or to random errors. Evaluation of the standard deviation of corresponding values of P_ν is based on the differences between the P_ν sets, uncertainties of individual P_ν values, as well as on the uncertainty involved in the normalizing value of $\bar{\nu}$. Our earlier work[10] contains more details on this method.

The recommended prompt P_ν values for the four fissile nuclides are shown in Table IV for fission at the thermal neutron energy, E_n = 0.0253 eV. In addition to the P_ν values and $\bar{\nu}$, values are also given in these Tables for the average value of the quantities $\nu(\nu-1)$ and ν^2 and for the variance, $\sigma^2(\nu)$, which is the average of $(\nu - \bar{\nu})^2$.

V. Discussion

From Table III, it can be seen that there have been changes in the $\bar{\nu}$ values for the fissile nuclides although the value for the standard, ^{252}Cf, has been basically unchanged. These differences are primarily due to the various corrections that have been made to the prompt $\bar{\nu}$ ratio measurements as indicated in Table II. Recommended values are systematically lower than the values in ENDF/B-V. In most cases, the difference corresponds to between one and two standard deviations on the latest value. The uncertainty estimates associated with these $\bar{\nu}$ values are approximately twice as large as other recent evaluations, due primarily to the ^{252}Cf $\bar{\nu}$ uncertainty. The other evaluations are based on the least square fitting, (LSF), of all thermal neutron parameter data. Fitting a large amount of data produces very small standard deviations, which may or may not be justified. An uncertainty of one–tenth of one percent is recommended in the LSF evaluations for $\bar{\nu}$ of ^{252}Cf, although the three or four best measurements disagree by eight–tenths of one percent. $\bar{\nu}$ for ^{252}Cf has a much smaller uncertainty by a factor of between two to two and a half compared to this evaluation. This effect is then reflected in the uncertainty for each of the fissile nuclide's $\bar{\nu}$ values.

The recommended values for the prompt neutron emission probabilities shown in the last table can be used in nuclear safeguards for various correlation counting experiments, where the average value, $\bar{\nu}$, is insufficient. Although the values given are only for thermal neutrons, work is continuing to determine a best set of P_ν values at higher neutron energies. Detailed tables and full references are available from the authors.

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Table I. Percentage Corrections to Measured $\bar{\nu}$ Ratios for Various Effects

Author	Nuclide	Foil Thickness	Delayed γ	Fission Spectrum
Gwin[4]	^{233}U	0.04 % ± 0.02 % *	− 0.4 % ± 0.12 % *	− 0.068 % ± 0.034 % ***
Gwin[4]	^{235}U	0.2 % ± 0.1 % *	− 0.2 % ± 0.06 % *	− 0.091 % ± 0.046 % ***
Gwin[4]	^{239}Pu	0.2 % ± 0.1 % *	− 0.4 % ± 0.12 % *	− 0.045 % ± 0.028 % ***
Gwin[4]	^{241}Pu	0.04 % ± 0.02 % *	− 0.4 % ± 0.12 % *	− 0.051 % ± 0.033 % ***
Boldeman[11]	^{233}U	0.12 % ± 0.06 %	− 0.32 % ± 0.10 %	− 0.30 % ± 0.15 % ***
Boldeman[11]	^{235}U	0.235 % ± 0.118 %	− 0.16 % ± 0.05 %	− 0.39 % ± 0.19 % ***
Boldeman[11]	^{239}Pu	0.03 % ± 0.015 %	− 0.43 % ± 0.13 %	− 0.20 % ± 0.13 % ***
Boldeman[11]	^{241}Pu	0.14 % ± 0.07 %	− 0.43 % ± 0.13 %	− 0.23 % ± 0.15 % ***
Mather[12]	^{233}U	0.30 % ± 0.15 %	− 0.40 % ± 0.12 %	− 0.13 % ± 0.065 % ***
Mather[13]	^{235}U	0.425 % ± 0.213 %	− 0.20 % ± 0.06 %	− 0.18 % ± 0.09 % ***
Mather[12]	^{239}Pu	0.26 % ± 0.13 %	− 0.54 % ± 0.16 %	− 0.09 % ± 0.06 % ***
Conde[14]	^{235}U	0.3 % *	− 0.25 % ± 0.08 %	− 0.43 % ± 0.22 % ***
Colvin[15]	^{233}U	0.28 % ± 0.14 %		− 0.067 % ± 0.034 %
Colvin[15]	^{235}U	0.28 % ± 0.14 %		− 0.09 % ± 0.045 %
Colvin[15]	^{239}Pu	0.25 % ± 0.13 %		− 0.045 % ± 0.028 %
Colvin[15]	^{241}Pu	0.25 % ± 0.13 %		− 0.050 % ± 0.032 %
Hopkins[16]	^{233}U	0.03 % ± 0.015 %	− 0.20 % ± 0.06 %	− 0.158 % ± 0.078 %
Hopkins[16]	^{235}U	0.28 % ± 0.14 %	− 0.10 % ± 0.03 %	− 0.208 % ± 0.104 %
Hopkins[16]	^{239}Pu	0.25 % ± 0.13 %	− 0.30 % ± 0.09 %	− 0.104 % ± 0.065 %
Nurpeisov[17]	^{233}U	0.0 % ± 0.32 % **		− 0.51 % ± 0.26 % ***
Prokhorova[9]	^{235}U	0.175 % ± 0.09 %		− 0.69 % ± 0.35 % ***
Meadows[18]	^{235}U	0.076 % ± 0.04 %		− 1.2 % ± 0.80 % ***
Bolodin[19]	^{239}Pu	0.0 % ± 0.31 % **		− 0.34 % ± 0.22 % ***

* Author's own correction
** Sample assumed to be 10 gm/m^2 thick and correction applied as an uncertainty
*** Author's estimated spectral correction removed before applying cited correction

Table II. Comparison of Recommended Prompt $\bar{\nu}$ Ratios for the Fissile Nuclides

Author	^{233}U/^{252}Cf	^{235}U/^{252}Cf	^{239}Pu/^{252}Cf	^{241}Pu/^{252}Cf
this work	0.6594 ± 0.0011	0.6423 ± 0.0009	0.7652 ± 0.0011	0.7797 ± 0.0013
Divadeenam[20]	0.6615 ± 0.0010	0.6407 ± 0.0008	0.7836 ± 0.0014	0.7771 ± 0.0018
ENDF/B-V[21]	0.6620	0.6441	0.7679	0.7817

Table III. Comparison of Recommended Total $\bar{\nu}$ Values for the Fissile Nuclides

Author	^{233}U	^{235}U	^{239}Pu	^{241}Pu	^{252}Cf
this work	2.484 ± 0.008	2.430 ± 0.007	2.881 ± 0.009	2.945 ± 0.009	3.766 ± 0.010
Axton[22]	2.495 ± 0.004	2.4334 ± 0.0036	2.8822 ± 0.0051	2.9463 ± 0.0058	3.7676 ± 0.0047
Divadeenam[29]	2.493 ± 0.004	2.4251 ± 0.0034	2.8768 ± 0.0057	2.937 ± 0.007	3.7675 ± 0.0040
ENDF/B-V[21]	2.495	2.437	2.891	2.953	3.766
Lemmel[23]	2.479 ± 0.006	2.416 ± 0.005	2.862 ± 0.008	2.924 ± 0.010	3.746 ± 0.009
Hanna[24]	2.474 ± 0.060	2.4229 ± 0.0066	2.8799 ± 0.0090	2.934 ± 0.012	3.765 ± 0.012
Westcott[25]	2.494 ± 0.069	2.430 ± 0.008	2.871 ± 0.014	2.969 ± 0.023	3.772 ± 0.015

Table IV. Recommended Prompt P_ν Values

P_ν	^{233}U	^{235}U	^{239}Pu	^{241}Pu
P_0	0.0262 ± 0.0012	0.0317 ± 0.0015	0.0109 ± 0.0001	0.0108 ± 0.0005
P_1	0.1550 ± 0.0022	0.1720 ± 0.0014	0.0995 ± 0.0028	0.0895 ± 0.0014
P_2	0.3328 ± 0.0038	0.3363 ± 0.0031	0.2750 ± 0.0003	0.2860 ± 0.0017
P_3	0.3225 ± 0.0020	0.3038 ± 0.0004	0.3270 ± 0.0041	0.3313 ± 0.0041
P_4	0.1325 ± 0.0057	0.1268 ± 0.0036	0.2045 ± 0.0087	0.2140 ± 0.0039
P_5	0.0272 ± 0.0024	0.0266 ± 0.0026	0.0728 ± 0.0133	0.0749 ± 0.0050
P_6	0.0037 ± 0.0018	0.0026 ± 0.0009	0.0097 ± 0.0027	0.0112 ± 0.0024
P_7	0.0001 ± 0.0001	0.0002 ± 0.0001	0.0006 ± 0.0009	0.0023 ± 0.0013
$\bar{\nu}$	2.477 ± 0.008	2.413 ± 0.007	2.875 ± 0.009	2.929 ± 0.009
$<\nu(\nu-1)>$	4.850	4.635	6.738	7.017
$<\nu^2>$	7.460	7.049	9.613	9.946
$\sigma^2(\nu)$	1.324	1.226	1.347	1.367

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