

Empirical Formulas for Estimation of Fission Prompt Neutron Multiplicity for Actinide Nuclides

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Empirical formulas were obtained for estimation of the prompt neutron multiplicity for actinides as a function of the incident neutron energy. Measured data for eleven nuclides (^{232}Th , ^{233}U , ^{235}U , ^{238}U , ^{237}Np , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{241}Am , ^{243}Am , and ^{245}Cm) were used as the basis to extract systematic relations of slopes and intercepts of a linear function of the prompt neutron multiplicity. The obtained formulas were compared with available measured data and existing evaluations. It was found that these formulas could be used to estimate the quantity for nuclides for which no or scarce data is available.

1. Introduction

With the increase of interest to extended burn-up of nuclear fuel and incineration of long-lived actinide nuclides, the need for accurate nuclear data for actinide nuclides has been rising these years. However the quality of nuclear data for minor actinides remains to be improved.¹ The figure-of-merit of a nuclide in nuclear design of a reactor is evaluated by the product $\bar{\nu}(E_n)\sigma_f(E_n)$ of the average prompt neutron multiplicity $\bar{\nu}(E_n)$ and fission cross section $\sigma_f(E_n)$, which means that the prompt neutron multiplicity is one of the important quantities that determines the performance of the system. Efforts have been devoted to deduce formulas that can be used for evaluation of the quantity $\bar{\nu}(E_n)$ for any actinides, but existing formulas were obtained on the basis of the measurements available in the seventies. Thus it is useful to re-examine the preceding works and experimental data available nowadays so as to construct formulas that makes it possible to evaluate the required data in compiling evaluated nuclear data libraries. Actually, the present formulas are planned to be used for evaluation of prompt neutron multiplicity for some actinides in Japanese Evaluated Nuclear Data Library, version 4 (JENDL-4) presently under compilation.

2. Preceding Studies

It would be useful to review the preceding studies on the prediction of the average number of prompt neutrons for actinides. Here two models are discussed that have been used to evaluate the $\bar{\nu}(E_n)$ -values in several nuclear data files.

2.1. Howerton's Mode. Howerton^{2,3} assumed that $\bar{\nu}(E_n)$ can be expressed by a truncated pseudo-Taylor series expansion:

$$\bar{\nu}(E, A, Z) = C_0 + C_1(Z-92) + C_2(A-235) + C_3(E_n - E_{th}) + C_4(A-235) \cdot (E_n - E_{th}) + \dots \quad (1)$$

The constant C_0 was evaluated from systematics of the prompt neutron multiplicity at fission thresholds for ^{235}U and ^{238}U . Other coefficients C_i were also obtained from experimental data. This led to the resulting formula:

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$$\bar{\nu}(Z, A, E_n) = 2.33 + 0.06[2 - (-1)^{A+Z} - (-1)^Z] + 0.15(Z-92) + 0.02(A-235) + [0.130 + 0.006(A-235)](E_n - E_{th}). \quad (2)$$

This formula was extended so as to be applicable in the neutron energy region where multiple-chance fission can occur. (See eqs 9 – 11 of Reference 2.) This set of formulas has been used in many nuclear data files, including JENDL-3.3, to estimate the $\bar{\nu}(E_n)$ -values for nuclides for which no or scarce measured data were available.

However, there seems to be some problems in this method:

a) The experimental data used to derive the formula were limited to those available at 1977. Many measurements have been performed during the last thirty years, especially for heavier actinides. It is desirable to take these data into consideration.

b) It was assumed that $\bar{\nu}(Z, A, E_n)$ was adequately expressed in terms of truncated pseudo-Taylor series expansion around ^{235}U . However, it is rather questionable whether the functional forms and relevant parameters determined on the basis of the data in the narrow (U–Pu) region can be safely applied throughout the wider region of actinides up to Cf.

c) The value 3.73 used by Howerton^{2,3} for the $\bar{\nu}$ -value for spontaneous fission (abbreviated as sf hereafter) of ^{252}Cf is 0.724% lower than the recent value of $\bar{\nu}(^{252}\text{Cf}, \text{sf}) = 3.757$ adopted by Boldeman⁴ and widely used. In the present work, the experimental data were renormalized to this standard to remove biases due to diversity of the standard.

2.2. Bois-Fréhaut's Method. Bois and Fréhaut⁵ performed measurements on $\bar{\nu}$ for ^{235}U , ^{238}U , ^{239}Pu , ^{240}Pu , and ^{241}Pu as a function of the incident neutron energy over the energy range 1.5–15 MeV, and proposed another method on the basis of their measurements.

First, they assumed that the average number of prompt neutrons was expressed as a linear function,

$$\bar{\nu} = a(E_n - E_{th}) + \bar{\nu}_s, \quad (3)$$

where E_{th} is the fission threshold energy (tabulated in Tableaux I of Reference 5), $\bar{\nu}_s$ the average number of prompt neutrons at the threshold energy. They analyzed the measured data to obtain the slope a and intercept $\bar{\nu}_s$ for each of nuclides. Then they determined the quantities: $\bar{\nu} = f(N, Z, E_{th}, E_n)$, $a = f(N, Z)$, $\bar{\nu}_s = g(N, Z)$, as functions of N , Z , E_{th} , and E_n . After comparison with the data of sf for U, Pu, Cm, and Cf isotopes, two new terms $0.153(N-145)$ and $0.082(-1)^N$ were introduced to consider

fluctuations due to even-odd difference of N . The final form was

$$\bar{\nu} = (2.767 \pm 0.010) + (0.200 \pm 0.006)(Z - 94) + 0.153(N - 145) + 0.082(-1)^N + [(0.1496 \pm 0.0006) + (0.0031 \pm 0.0004)(N - 145)](E_n - E_{th}) \quad (4)$$

[for $94 \leq Z \leq 96$],

It should be noted that the coefficient of $(E_n - E_{th})$ in eq 4 is smaller than the coefficient $0.130 + 0.006(A - 235)$ of Howerton³ in the region heavier than ^{239}Pu .

There are some problems to be discussed on this method.

a) The term $0.082(-1)^N$ dependent on the even-odd nature of N was deduced from the data in the narrow region of U to Pu, so it remains to be verified if the term is applicable to wider range of actinides.

b) The N -dependent term $0.153(N - 145)$ was obtained by using the $\bar{\nu}$ (sf)-data. It is now known that the excitation energies of the fission fragments are lower in sf compared with neutron-induced fission (n,f), since the initial excitation energy of the fissioning nucleus is zero in sf, and it is equal to $B_n + E_n$ in (n,f), where B_n is the neutron binding energy of the fissioning nucleus. Thus the present author considers that the sf-data should be treated separately and not to be included in deriving systematic relations in $\bar{\nu}$.

c) The standard value $\bar{\nu}(^{252}\text{Cf}, \text{sf}) = 3.732$ of Axton⁶ was used by the authors as the reference. This value is 0.67% lower than the newer standard value of 3.757, determined by Boldeman⁴ and adopted in the present evaluation.

3. Analyses of Experimental Data

3.1. Experimental Data Base. Experimental data were taken from the experimental nuclear reaction data base EXFOR available at the web site of IAEA. Measured data relative to ^{252}Cf (sf) were renormalized by using $\bar{\nu}(^{252}\text{Cf}, \text{sf}) = 3.757$, and those relative to ^{235}U were renormalized by using the $\bar{\nu}(^{235}\text{U})$ -values of the current version of the Japanese Evaluated Nuclear Data Library (JENDL-3.3⁷).

As the first step, every renormalized data were plotted, and best-fit lines were obtained for 11 actinides ($^{232}\text{Th} - ^{245}\text{Cm}$) by least-squared method assuming a simple formula $\bar{\nu} = aE_n + \bar{\nu}_0$. Some notes should be given here: (a) Small structures observed near (n,f), (n,n'f) and (n,2nf)-thresholds were ignored, because our aim was to obtain global systematic relations; (b) Non-uniform frequency distribution of measured data points on the incident energy axis (in many cases more experimental data are available in the low-energy region) results in effectively placing more emphasis on low-energy region in the least-squared analysis. While being aware of this tendency, the author did not try to exclude some of the low-energy data, considering that low-energy data are more important in reactor physics. (c) For ^{241}Am , it was found that the data of Khokhlov cited in the recent paper of Kuzminov⁸ are different from those registered in EXFOR. Considering that this is due to some additional correction made after the measurement, we used the values reported in Reference 8.

The best-fit lines for 11 nuclides (^{232}Th , ^{233}U , ^{235}U , ^{238}U , ^{237}Np , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{241}Am , ^{243}Am , and ^{245}Cm) are compared in **Figure 1**. Two apparent tendencies can be observed: (1) In general, the $\bar{\nu}$ -values are greater for higher- Z nuclides; *i.e.* the intercept $\bar{\nu}_0$ is an increasing function of Z . This is due to higher total energy release E_R just after fission for heavier elements (*e.g.* $E_R = 177.1$, 198.9 , and 209.4 MeV for ^{232}Th , ^{239}Pu , and ^{245}Cm , respectively, at an incident energy 2 MeV, assuming Wang-Hu model⁹ for fission fragment mass distribution, and TUYU mass formula¹⁰); (2) From the lines for U and Pu isotopes, it is observed that the slope of the lines becomes steeper

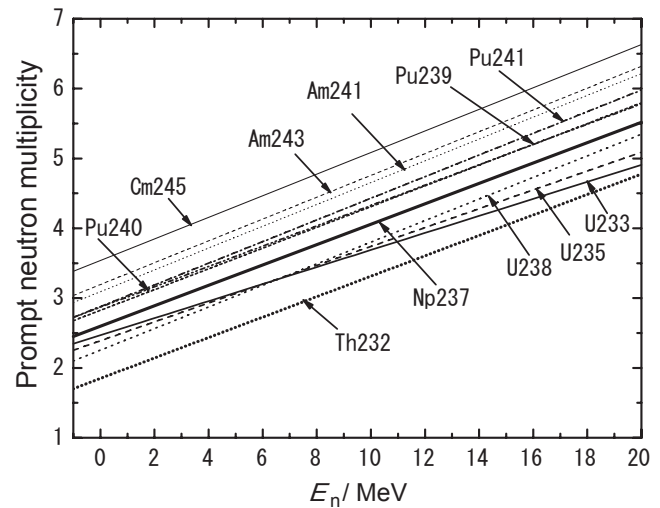


Figure 1. Comparison of least-squares fitting lines $\bar{\nu} = aE_n + \bar{\nu}_0$ for 11 nuclides

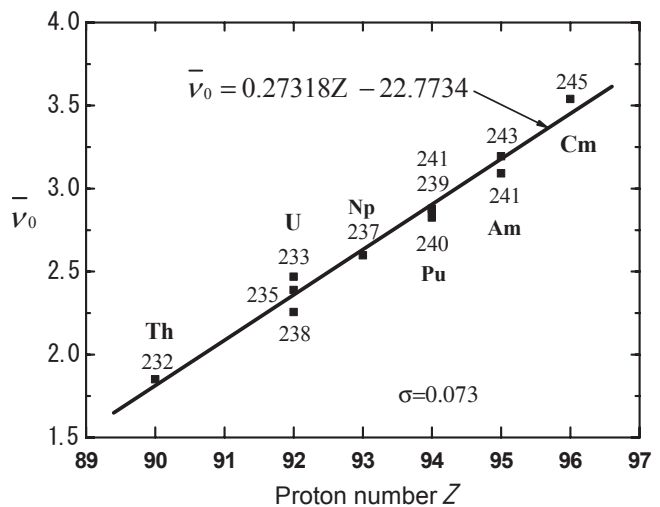


Figure 2. Correlation between the intercept $\bar{\nu}_0$ and the atomic number Z of target nuclides.

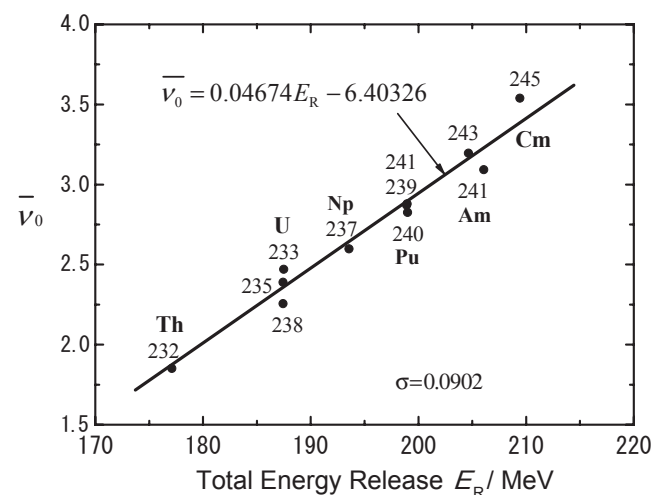


Figure 3. Correlation between the intercept $\bar{\nu}_0$ and the total energy release of fission E_R of target nuclides.

for heavier isotopes of the same element.

3.2. Intercept $\bar{\nu}_0$. We searched for correlations between the intercept $\bar{\nu}(E_n = 0 \text{ MeV})$ and other physical quantities, such as Z , N , E_R , fissility Z^2/A . After examination we found out two good correlations (**Figures 2 and 3**):

$$\bar{\nu}_0 \text{ vs. } Z: \quad \bar{\nu}_0 = 0.27318Z - 22.7734, \quad (\sigma = 0.073), \quad (5a)$$

$$\bar{\nu}_0 \text{ vs. } E_R: \quad \bar{\nu}_0 = 0.04674 E_R - 6.40326, \quad (\sigma = 0.090), \quad (5b)$$

where σ stands for the standard deviation. The two equations agree to within 1.6%, except Th, Pa, and ^{233}U for which they agree to within 3%. Good correlation between $\bar{\nu}_0$ and E_R , eq 5b, is self-evident, because $\bar{\nu}_0$ is approximately proportional to the total excitation energy of the fission fragments, which in turn is proportional to E_R . Good correlation between $\bar{\nu}_0$ and Z , eq 5a, is to be expected, because there is an explicit linear relation between E_R and Z .

The need for a term depending on even-odd character of N and/or Z of the target nucleus was not confirmed as long as the available experimental data are concerned. From physical point of view, the even-odd effect outstands in sf, because the excitation energy is not strong enough to break nucleon pairs, but this effect disappears at higher excitation energies.

3.3. Slope a . We searched for another correlation for the slope a , on the basis of empirical relations obtained in the previous subsection, and obtained two types of linear functions (**Figures 4 and 5**):

$$a \text{ vs. } A: a = -0.1636 + 0.0013A \text{ [Howerton (H) type]}, \quad (6a)$$

$$a \text{ vs. } N: a = -0.2256 + 0.00256N \text{ [Bois-Fr hault (BF) type]}. \quad (6b)$$

These equations should be compared with the corresponding terms in eqs 2 and 4. **Figure 4** shows that, when a is considered as a function of A , fit to the data for all nuclides resulted in a line that is considerably different from Howerton's line. This is due to the fact that Howerton's line was obtained from fitting to limited data for U and Pu isotopes that were available at that time. **Figure 5** shows that, regarded as a function of N , the present evaluations of a are rather close to Bois-Fr hault's line. This figure also shows that fit to specific nuclides, such as U or Pu isotopes, yields lines different from overall fitting. **Figure 6** compares the evaluations of a by Howerton, Bois-Fr hault, and the present author. It is worth noting that the two eqs 6a and 6b, obtained independently by different correlation analyses of H- and BF-type, provide results that are quite close to each other. This is one of the results of including the data of Am and Cm isotopes not considered in previous works.

3.4. New Proposal. In summary, we propose here two sets of empirical relations for the formula

$$\bar{\nu} = \bar{\nu}_0 + aE_n \quad (7)$$

with $\bar{\nu}_0$ as given by eqs 5a or 5b and the slope a as given by eqs 6a or 6b.

4. Verification

The empirical formulas obtained in the preceding section are verified by comparing with experimental data and existing evaluations such as Evaluated Nuclear Data File, format B, version VI, (ENDF/B-VI,¹¹ USA), Joint Evaluated Fission and Fusion File, version 3 (JEFF-3.0,¹² EU), Karlsruhe Evaluated Nuclear Data File, version 4 (KEDAK-4,¹³ Germany).

U-235: The present formulas agree well with JENDL-3.3 evaluation and experimental data within errors below 6 MeV, but a bit lower than that at higher energies. The evaluated values of $\bar{\nu}(E_n)$ for ^{235}U in JENDL-3.3 are based mainly on the measurement by Gwin^{14,15} and Fr hault.^{16,17} (**Figure 7**)

U-238: Three evaluations of $\bar{\nu}(E_n)$ in JENDL-3.3, ENDF/B-VI, JEFF-3.0 are all based on the data of Fr hault.¹⁸ The present formulas agree quite well above 6 MeV but higher than measurements below 6 MeV. (**Figure 8**)

Np-237: The evaluation of $\bar{\nu}(E_n)$ in ENDF/B-VI is a smooth

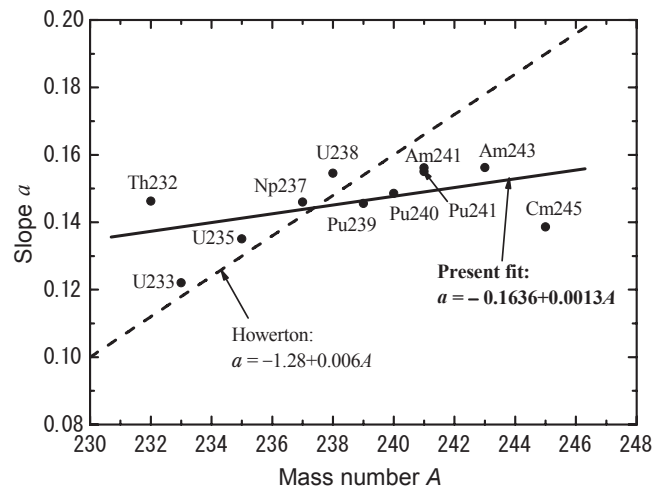


Figure 4. Correlation between the slope a and the mass number A of target nuclides. Howerton's relation was obtained only from U isotope data.

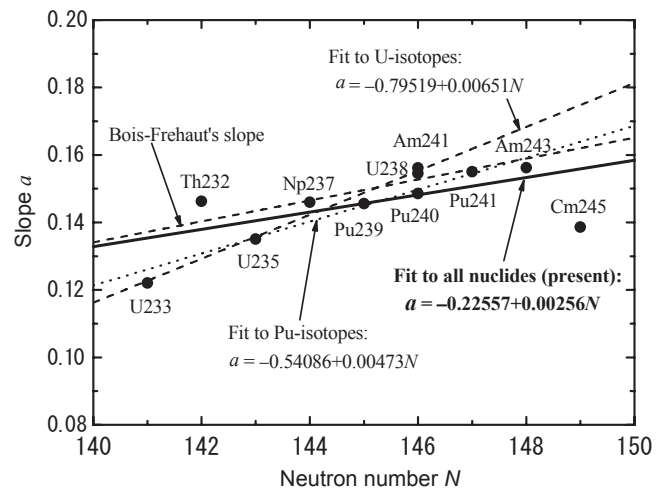


Figure 5. Correlation between the slope a and the neutron number N of target nuclides. Correlations observed among U data ($a = -0.79519 + 0.00651N$) and Pu data ($a = -0.54086 + 0.00473N$) are also shown. The present evaluation was obtained by least-squares fitting to all the data.

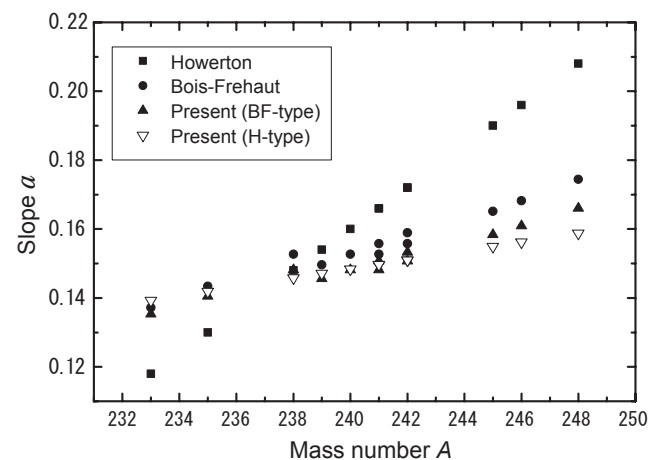


Figure 6. Comparison of evaluations of the slope a by Howerton, Bois-Fr hault, and the present author of H- and BF-type function. It is worth noting that the present two equations 6a and 6b, obtained independently by different correlation analyses of H- and BF-type, give results that are very close to each other.

approximation to the measurements by Malinovsky,^{19, 20} Veeseer,²¹ and Fréhaut.¹⁶ The evaluation of $\bar{\nu}(E_n)$ in JENDL-3.3 in addition considers Boikov²² and Mughabghab.²³ The present formulas agree quite well with JENDL-3.3 evaluation. (**Figure 9**)

Pu-239: The evaluation of $\bar{\nu}(E_n)$ in JENDL-3.3 is based on the experimental data of Gwin^{24,26} and Fréhaut.²⁵ The evaluation of $\bar{\nu}(E_n)$ in ENDF/B-VI is based on the evaluation of Fort,²⁷ after minor renormalization for consistency with the standards of the Cross Section Evaluation Working Group (CSEWG, Brookhaven National Laboratory). JEFF-3.0 adopted the evaluation of Fort,²⁷ supplemented by calculation by Vladuka²⁸ above 650 keV. No considerable difference was observed between the three evaluations. The present formulas give results that are essentially the same as the three evaluations. (**Figure 10**)

Am-241: No description is given on the evaluation method in the comment part of ENDF/B-VI file. Seemingly the line was adjusted to the value of 3.216 of Jaffy²⁹ at the thermal energy. The data of Khokhlov seem to be modified after publication at Gatlinburg Conference (1994).³⁰ In the present work, the data described in a recent report INDC(CCP)-435 (2002)³¹ was used. The evaluation of $\bar{\nu}(E_n)$ in JEFF-3.0 is higher than

measurements and other evaluations; the reason is not clear because no description on the evaluation method is given in the file. The present formula is consistent with Khokhlov, although a discrepancy of maximum 4% is observed in the 2 – 4 MeV region. (**Figure 11**)

Am-243: The evaluation of $\bar{\nu}(E_n)$ in JENDL-3.3 is based on Maslov's calculation.³² ENDF/B-VI adopted the Madland-Nix model calculation.³³ These two evaluations are more or less similar. However, JEFF-3.0 evaluation, based on Howerton's method, is steeper than the two. The present formulas give lines with intermediate slopes. (**Figure 12**)

Cm-243: Both JENDL-3.3 and ENDF/B-VI adopted Maslov's calculation.³² JEFF-3.0 is based on Howerton's method renormalized to the measured data of Jaffy²⁹ and Zhuravlev³⁴ at the thermal energy and gives a steeper line. Generally speaking, Maslov's calculations tend to be less steep than Howerton's formula, as can be confirmed for other nuclides. The present formulas provide lines with intermediate slopes. (**Figure 13**)

Cm-244: The evaluation of $\bar{\nu}(E_n)$ in JENDL-3.3 was obtained by Howerton's method. ENDF/B-VI also adopted Howerton's method but the value was renormalized to the thermal value computed from semi-empirical work of Gordeeva and Smirenkin³⁵ as revised by Manero and Konshin.³⁶ The evaluation of JEFF-3.0 was taken from KEDAK-4¹³ but the details of the method are not known. The present formula

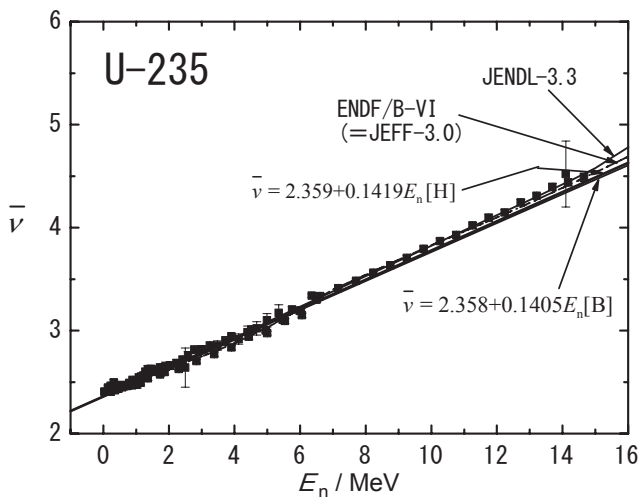


Figure 7. Prompt neutron multiplicity vs. incident neutron energy for ²³⁵U. The present evaluations are compared with existing evaluations (JENDL-3.3, ENDF/B-VI, JEFF-3.0) and experimental data. The equality sign, such as in “ENDF/B-VI = JEFF-3.0”, means that the two evaluations are *exactly identical*. The symbol [B] stands for an evaluation using Bois-Fréhaut-type equations 5a and 6a, while [H] stands for an evaluation using Howerton-type equations 5b and 6b.

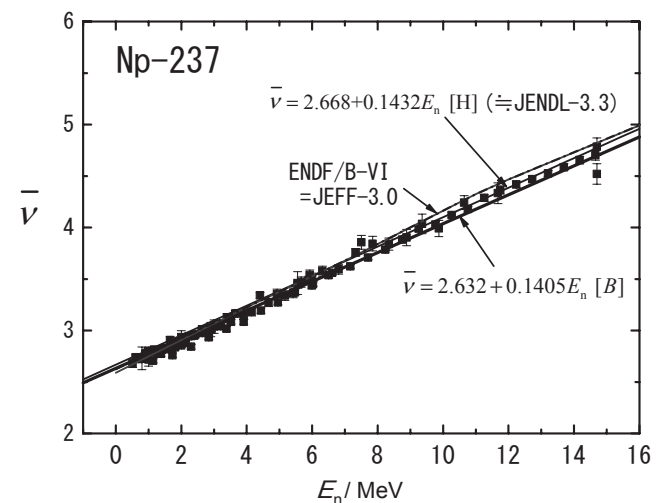


Figure 9. Prompt neutron multiplicity vs. incident neutron energy for ²³⁷Np. See the caption of Figure 7 for further explanation. The sign “≠” means that the two evaluations are *not identical but very close*.

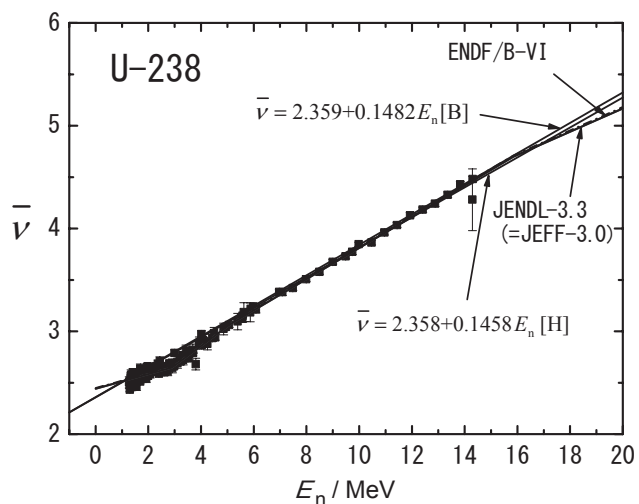


Figure 8. Prompt neutron multiplicity vs. incident neutron energy for ²³⁸U. See the caption of Figure 7 for further explanation.

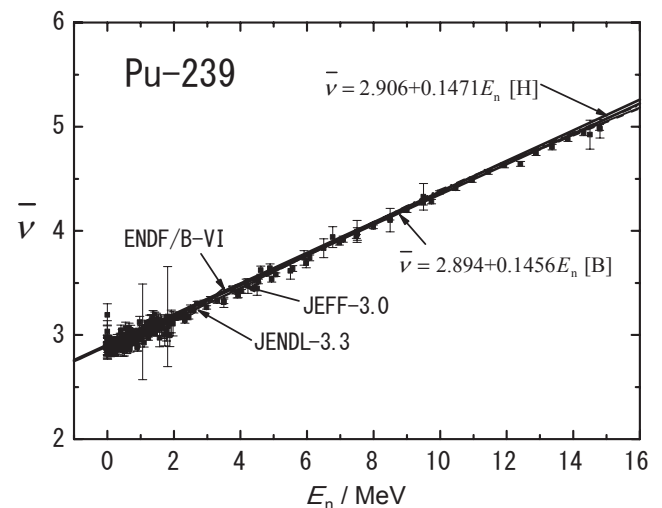


Figure 10. Prompt neutron multiplicity vs. incident neutron energy for ²³⁹Pu. See the caption of Figure 7 for further explanation.

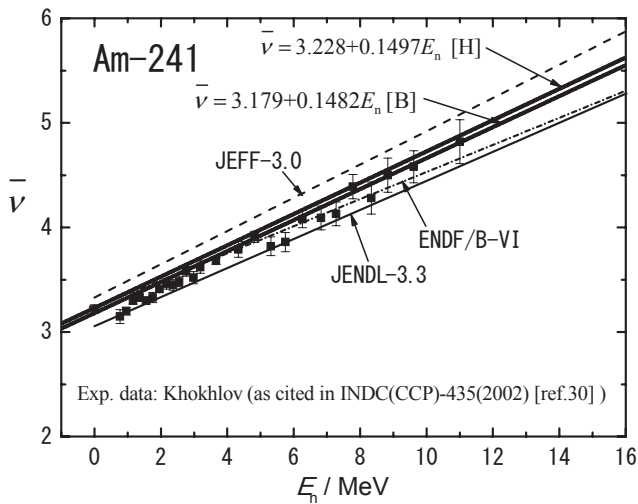


Figure 11. Prompt neutron multiplicity vs. incident neutron energy for ^{241}Am . See the caption of Figure 7 for further explanation.

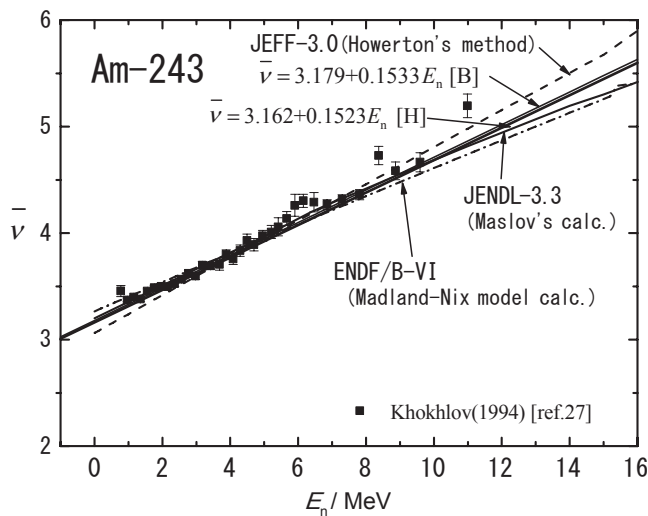


Figure 12. Prompt neutron multiplicity vs. incident neutron energy for ^{243}Am . See the caption of Figure 7 for further explanation.

gives lines located between ENDF/B-VI and JEFF-3.0, with rather smaller slopes. (Figure 14)

Cm-245: Both JENDL-3.3 and ENDF/B-VI adopted Maslov's calculation. JEFF-3.0 is based on the measurements of Khokhlov.³⁷ The present formulas are closer to Khokhlov's experiments than to Maslov's calculation,³⁸ the energy-dependence of which is rather weak. (Figure 15)

Cm-246: JENDL-3.3, ENDF/B-VI, and JEFF-3.0 all adopt Maslov's calculation. The slope of the present formulas is steeper than that of Maslov's calculation. (Figure 16)

Cm-247: JENDL-3.3 and JEFF-3.0 adopted Zhuravlev's measured value³⁴ at the thermal energy with the energy dependence calculated with Howerton's method. However, Zhuravlev's thermal data is exceptionally high among curium isotopes. ENDF/B-VI adopted a thermal value computed from semi-empirical work of Gordeeva and Smirenkin³⁵ as revised by Manero and Konshin,³⁶ together with the energy dependence of Howerton. The present evaluation gives values lower than any of the existing evaluations. (Figure 17)

Cm-248: JENDL-3.3 adopted the Howerton's method which was also adopted by JEFF-3.0. ENDF/B-VI evaluation was obtained by the same method as for Cm-247. The present evaluations are less steep. (Figure 18)

5. Conclusions

1) Assuming a linear function of the form $\bar{\nu} = \bar{\nu}_0 + aE_n$, the

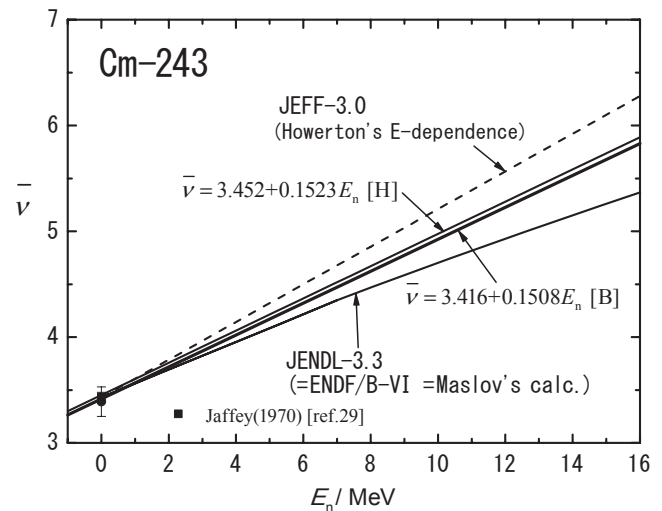


Figure 13. Prompt neutron multiplicity vs. incident neutron energy for ^{243}Cm . See the caption of Figure 7 for further explanation.

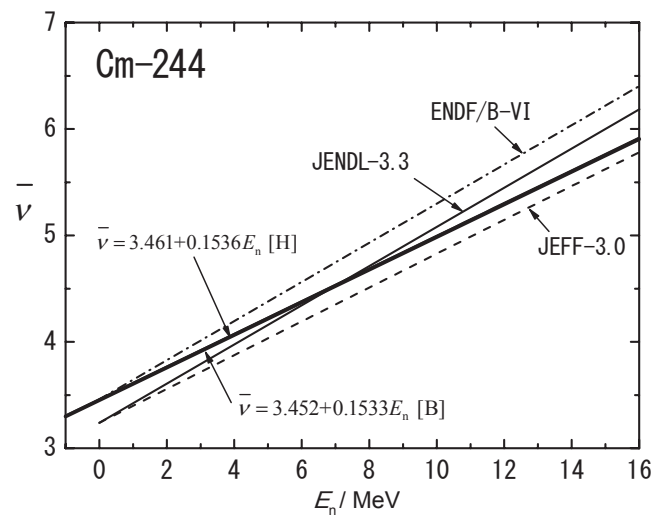


Figure 14. Prompt neutron multiplicity vs. incident neutron energy for ^{244}Cm . See the caption of Figure 7 for further explanation.

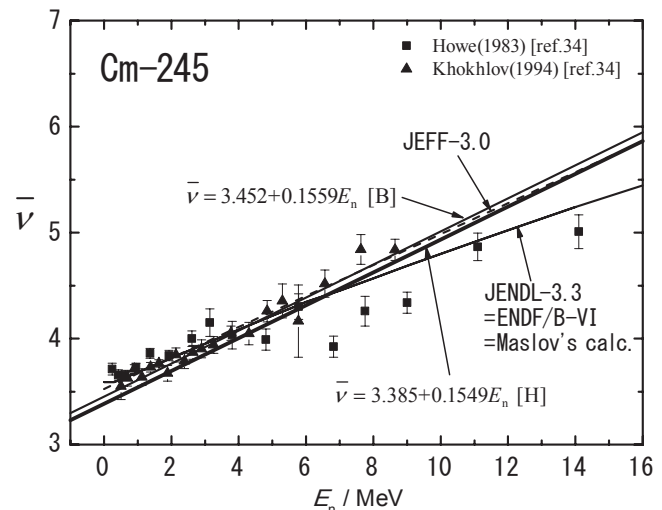


Figure 15. Prompt neutron multiplicity vs. incident neutron energy for ^{245}Cm . See the caption of Figure 7 for further explanation.

author determined the parameters $\bar{\nu}_0$ and a by least-squared fitting to experimental data for 11 actinides. Two empirical equations 5a and 5b for $\bar{\nu}_0$ and 6a and 6b for a , respectively, with nearly equal standard deviations were obtained. The present formulas give the slope parameter a that is considerably

smaller than Howerton's formula and rather close to (actually a bit smaller than) Bois-Fréhault's formula for actinides with $A > 237$.

2) The obtained equations were verified by comparing the predicted $\bar{\nu}$ -values with experimental as well as evaluated data

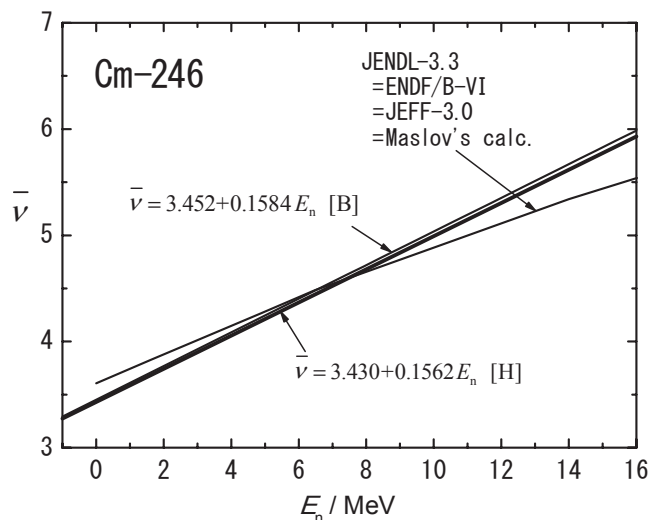


Figure 16. Prompt neutron multiplicity vs. incident neutron energy for ^{246}Cm . See the caption of Figure 7 for further explanation.

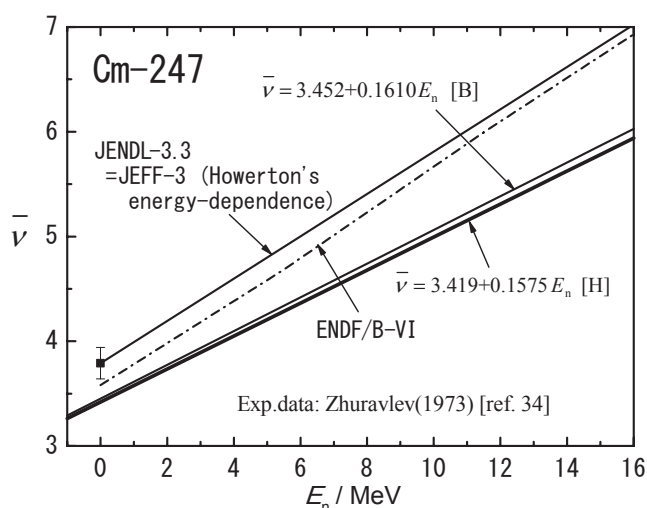


Figure 17. Prompt neutron multiplicity vs. incident neutron energy for ^{247}Cm . See the caption of Figure 7 for further explanation.

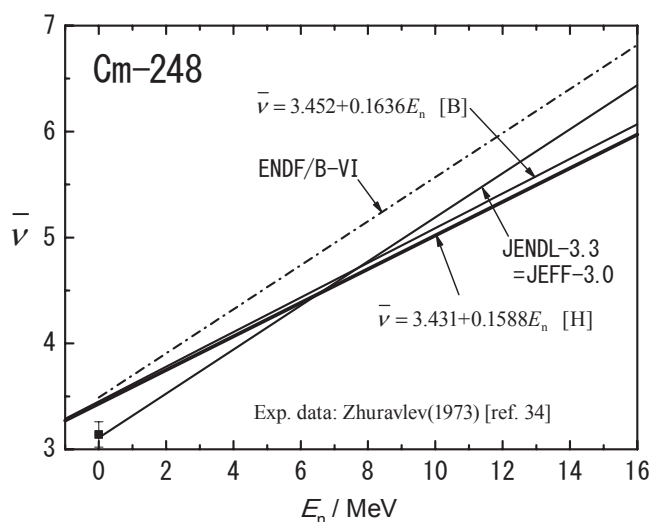


Figure 18. Prompt neutron multiplicity vs. incident neutron energy for ^{248}Cm . See the caption of Figure 7 for further explanation.

for 11 nuclides. It was found that the proposed formulas provide overall good representation without any *ad hoc* adjustment. These formulas can be used to predict $\bar{\nu}$ -values for nuclides for which no or scarce data is available.

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