Effects of Fission Yield Data in the Calculation of Antineutrino Spectra for ^{235}U(n,fission) at Thermal and Fast Neutron Energies

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(Received 16 October 2015; revised manuscript received 9 March 2016; published 1 April 2016)

Fission yields form an integral part of the prediction of antineutrino spectra generated by nuclear reactors, but little attention has been paid to the quality and reliability of the data used in current calculations. Following a critical review of the thermal and fast ENDF/B-VII.1 ^{235}U fission yields, deficiencies are identified and improved yields are obtained, based on corrections of erroneous yields, consistency between decay and fission yield data, and updated isomeric ratios. These corrected yields are used to calculate antineutrino spectra using the summation method. An anomalous value for the thermal fission yield of ^{86}Ge generates an excess of antineutrinos at 5–7 MeV, a feature which is no longer present when the corrected yields are used. Thermal spectra calculated with two distinct fission yield libraries (corrected ENDF/B and JEFF) differ by up to 6% in the 0–7 MeV energy window, allowing for a basic estimate of the uncertainty involved in the fission yield component of summation calculations. Finally, the fast neutron antineutrino spectrum is calculated, which at the moment can only be obtained with the summation method and may be relevant for short baseline reactor experiments using highly enriched uranium fuel.

DOI: 10.1103/PhysRevLett.116.132502

The current reactor antineutrino experiments, Double Chooz [1], Daya Bay [2], and RENO [3], have solved the long-standing question of the oscillation parameter θ_{13} value. Yet, a new puzzle has emerged on the shape of the measured antineutrino spectrum, as it differs from the best model predictions [4,5] with a prominent antineutrino excess at energies between 5 and 7 MeV [6,7]. These model predictions use the "conversion method," which relies on precisely measured electron spectra [8–11] to deduce the corresponding antineutrino spectra. Antineutrino spectra can also be calculated using a comprehensive set of nuclear data [12], an approach known as the "summation method." A recent study [13] found that the summation method gives an excess of antineutrinos in the 5–7 MeV region relative to the conversion method. This intriguing result calls for a thorough investigation into the method and data utilized in these calculations in order to solve the antineutrino excess puzzle.

There are two components in a summation calculation, one relating to the antineutrino spectra from each of the isotopes undergoing β^- decay and the other its fission yield, which provides the weighting factor for the individual spectra. Most of the recent work has centered on the former, whereas the fission yield component has garnered less attention. Fallot *et al.* [14] explored the effects of total absorption gamma-ray spectroscopy (TAGS) data. Hayes *et al.* [15] investigated the impact of first forbidden transitions. The antineutrino spectra were decomposed into the contribution from individual nuclei, and prominent ones were identified in the region of the observed excess in Refs. [13,16,17]. Recently, Hayes *et al.* [18] studied a number of possible sources of this antineutrino excess. Our goal is to thoroughly address the heretofore neglected fission yield component in the calculation of antineutrino spectra, investigating corrections of erroneous yields, consistency between decay and fission yield data, and realistic estimates of isomer population following fission. We note that fission yields impact a wide range of applications, and thus results of this work are relevant to a number of fields, such as antineutrino detection for reactor monitoring [19], decay heat [20], and nuclear forensics [21].

Another puzzle to be investigated in future experiments is the so-called reactor antineutrino anomaly, a short-distance deficit of measured antineutrinos [22], which has led to the suggestion of one or more sterile neutrinos and prompted the development of new short baseline experiments such as PROSPECT [23], where the detectors will be near a highly enriched uranium (HEU) reactor. As ^{235}U will contribute most of the fission events, the analysis will be simpler; however, since the total electron spectrum has only been measured at thermal energies, summation calculations may be needed to understand the effect of nonthermal neutrons. Thus, we also explore the fast neutron fission yields for ^{235}U and calculations of the corresponding antineutrino spectrum.

Currently, there are two distinct sets of recommended fission yield data, the ENDF/B-VII.1 [24] and JEFF-3.1 [25] libraries, which provide independent and cumulative fission yields for ground state (g.s.) and isomeric levels. The JENDL library [26] also has fission yield data, which are basically identical to those in ENDF/B. The independent fission yield (IFY) is the probability that a level is populated after a single fission event. The cumulative fission yield (CFY) is recursively defined as $\mathrm{CFY}_i = \mathrm{IFY}_i + \sum b_{ki}\mathrm{CFY}_k$, where b_{ki} is the probability that level k decays to level i.

Prior studies [27–29] have indicated issues with the current ENDF/B ^{235}U thermal fission yields for ^{86}Ge, which has an IFY value much larger than the IFYs for the other Ge isotopes, despite being quite neutron rich; see Fig. 1. The IFYs for ^{87,88}Ge also appear anomalous. We interpret these $^{86-88}$Ge IFYs as spurious errors since, for Ge isotopes, the yield distribution as a function of mass is expected to be well represented by a Gaussian distribution, as observed in the JEFF and fast ($E_{\mathrm{neutron}} = 500$ keV) ENDF/B yields and predicted by the GEF model [30]. Additionally, the sum of IFYs for $Z = 32$ is ~2.4 times larger than that for the $Z = 60$ complementary partners. From simple charge conservation and neglecting ternary fission, the sum of IFYs for Z and 92-Z should be equal, within uncertainties. From the historical releases of the ENDF/B library, we can track the evolution of the ^{86}Ge IFY, which was below 10^{-5} for ENDF/B-V and ENDF/B-VI.1, but changed to its current anomalous value for ENDF/B-VI.2 in 1993. We note that the data source [31] for the Ge yields evaluation [32] only contains data for $^{79-84}$Ge and the first observations of $^{86-89}$Ge were reported later [33,34]. We believe that the erroneous value originates from a misassignment of the ^{86}Se CFY in Ref. [31] to that of ^{86}Ge.

Since we have no access to the data or codes used to derive the ENDF/B yields, we have applied an *ad hoc* correction using a weighted Gaussian fit to the data, excluding outliers. For example, we show in Fig. 1 the original Ge IFYs, the Gaussian fit, the corrected IFYs, the fast IFYs, and GEF results for 2×10^{10} events. For thermal ENDF/B IFYs, the Gaussian fit correction was applied to ^{84,86,87,88}Ga, ^{86,87,88}Ge, ^{88}As, ^{96,98,100}Kr, ^{85,100}Rb, ^{120,130,131}Cd, ^{137}Sb, and ^{140}Te. For $Z = 32$, the sum of corrected IFYs is within 0.5% of the sum of $Z = 60$ IFYs. We have also identified similar anomalous deviations in the fast ^{235}U ENDF/B yields and the ^{93}Br, ^{105,106,107,108}Y, and ^{105,106,107,108}Zr IFYs were corrected.

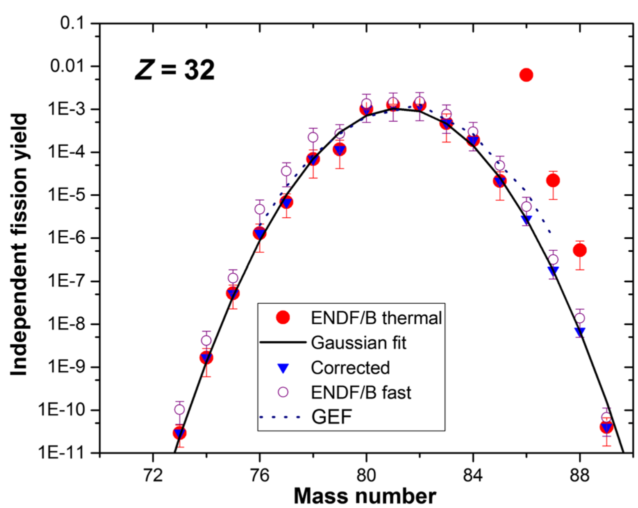

FIG. 1. Independent fission yields for germanium isotopes. Full (open) circles are the thermal (fast) ENDF/B-VII.1 values, the full line is a weighted Gaussian fit to the thermal values (excluding ^{86,87,88}Ge), triangles are the corrected thermal yields, and the dotted line is a GEF calculation.

The next set of corrections was applied to the g.s. and isomer IFYs of a given nucleus, which is of importance since, typically, one of them, due to angular momentum considerations, produces more energetic antineutrinos than the other. In cases where no experimental isomeric ratio, IFY(isomer)/[IFY(g.s.) + IFY(isomer)], was available and the spins were unknown, ENDF/B equally split the yields between the two states. If the spins were known, the Madland-England model [35] was used to estimate the yields. In this work we have used the measured $^{239}\mathrm{Pu}(n,\mathrm{F})$ isomeric ratios [36] for ^{98}Y, ^{99}Nb, and ^{136}I, as we expect they would be similar for ^{235}U. In an attempt to provide a more realistic isomeric ratio for cases with no experimental value, a survey of the yrast band population in even-even nuclides following the spontaneous fission of ^{252}Cf was performed. About 30 such cases are available in the ENSDF database [37], yielding an average population of 100%, 66%, 41%, 18%, and 8% for the yrast 2^+, 4^+, 6^+, 8^+, and 10^+ levels, respectively. This distribution was used to obtain g.s. and isomeric IFYs of ^{96,97,100}Y, ^{100,102,104}Nb, ^{128,130,131}Sn, ^{134}Sb, ^{146}La, ^{148}Pr, and ^{152,154}Pm. Finally, the isomer IFYs for ^{84}As, ^{85}Se, ^{86}Br, ^{109}Ru, and ^{143}Xe were added to the g.s. IFYs since there is currently no evidence for these isomers [37].

Next, the corrected IFYs were renormalized to 2 and CFYs were obtained. We note that changing the ^{86}Ge IFY changes its CFY and those of the nuclides further down its β-decay path. The antineutrino spectra are calculated as [12]

$$I(E_{\bar{\nu}}) = \sum \mathrm{CFY}_i \times I_i(E_{\bar{\nu}}),$$

where $I_i(E_{\bar{\nu}})$ is the antineutrino spectrum from the ith β-minus decaying level in the network.

In this work we use an updated version of the ENDF/B-VII.1 decay data sublibrary [16], with the b_{ki} data to convert from IFY to CFY, as well as the parameters needed to calculate the level to level spectra, such as β-minus intensities (I_β), energies, and spin or parity change. The main sublibrary update is the use of I_β's from TAGS data [20,38] and from fits to measured β-minus spectra [39]. For nuclides with measured I_β's, electron and antineutrino spectra are calculated including finite size, radiative, and screening corrections as given by Huber [4]. For nuclides with poorly known I_β's, theoretical spectra [40] are used; specifically, ^{86}Ge falls in this group.

One way of validating the IFY corrections is to calculate the delayed neutron multiplicity per fission, $\bar{\nu_d}$, obtained as $\bar{\nu_d} = \sum \mathrm{CFY}_i(P_{1ni} + 2 \times P_{2ni})$, where $P_{1(2)ni}$ is the β-delayed 1(2)-neutron emission probability. Higher order neutron emission is negligible. With the corrected yields, we obtain a $\bar{\nu_d}$ of 1.559×10^{-2} and 1.747×10^{-2} for ^{235}U thermal and fast neutrons, respectively, while the recommended ENDF/B-VII.1 [24] values are 1.585×10^{-2} and 1.67×10^{-2}. The values obtained using the original yields are 2.021×10^{-2} and 1.786×10^{-2}; thus, for thermal neutrons, the corrected yields produce a $\bar{\nu_d}$ very close to the recommended value, compared to a result which is 27% higher with the original yields. We have also verified that the corrected CFYs are in good agreement with recent measurements [41] of long-lived fission products. Additionally, we observed that the corrected yields induce changes in the decay heat of less than 15% at times shorter than 100 s after fission, as the corrections were mainly applied to short-lived nuclides.

We first explore the effect of these yield corrections by studying the ratio of the calculated antineutrino spectrum using the corrected fission yields over the same calculation with the original ENDF/B CFYs. Results are given in Fig. 2(a) for thermal neutrons and Fig. 2(b) for fast neutrons. In addition, a similar ratio is plotted, but one where the numerator involves correcting only a single nucleus IFY and then calculating new CFYs, which highlights some of the most relevant cases but also reflects the use of new b_{ki} data. For thermal neutrons, the corrected yields produce a spectrum that, in the 0–5 MeV region, is within 3% of the one calculated using the original CFYs. In the 5–7 MeV region, the corrected spectrum is 3%–10% lower than the uncorrected one, which can be traced to the changes in the ^{86}Ge IFY. This result is very relevant to the excess of antineutrinos in the 5–7 MeV region, as using original ENDF/B CFYs will produce more antineutrinos in the same region, simply due to an artifact in the yields. As a consequence, the analyses and conclusions in Refs. [13,18] which used the original ENDF/B-VII.1 CFYs may need to be revised. For fast neutrons, the corrected yields produce a spectrum that, from 0–8 MeV, is within 4% of the one calculated using original yields. Note that the original ENDF/B-VII.1 CFYs used b_{ki} data different from those in more recent decay data evaluations; therefore, care should be taken when combining them in summation calculations of antineutrino spectra. We have done so here simply to illustrate the effect of correcting the yields.

As mentioned previously, there are two major fission yield libraries, ENDF/B and JEFF. We examine their differences by plotting the ratio of the antineutrino spectrum calculated with ENDF/B yields (both original and corrected) over that calculated with the corresponding JEFF-3.1 yields, as shown in Fig. 3(a). No corrections were applied to the JEFF yields; additionally, fast neutrons correspond to an energy of 400 keV. Clearly, the use of the corrected ENDF/B yields results in better agreement with the JEFF yields. For thermal neutrons there is only a few percent difference between calculations using these two distinct fission yield libraries for antineutrinos up to 5 MeV, reaching ~6% in the 6 to 7 MeV region. This allows us to place estimates on the uncertainties introduced into summation calculations from the fission yield component. In the 7 to 8 MeV region, ^{92}Rb is the dominant contributor

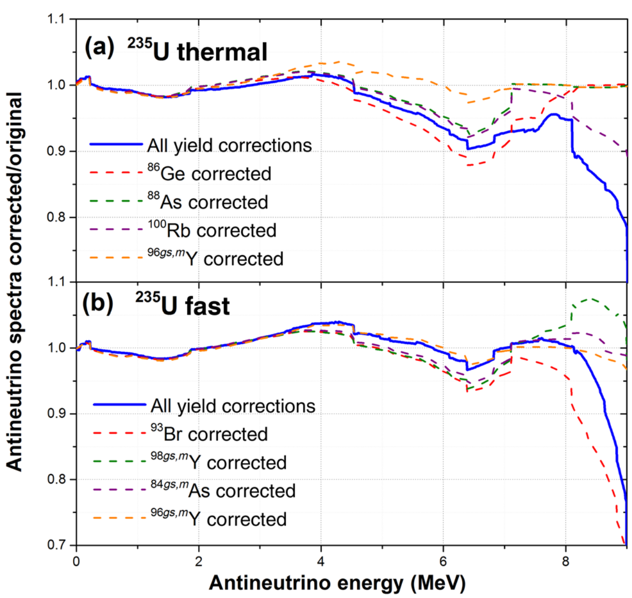

FIG. 2. (a) Thermal ^{235}U antineutrino spectra calculated using the corrected ENDF/B yields divided by the one using the original ENDF/B yields. The dashed lines correspond to similar ratios when only the fission yield for a single relevant nucleus was modified. (b) Same as (a), but for fast neutrons.

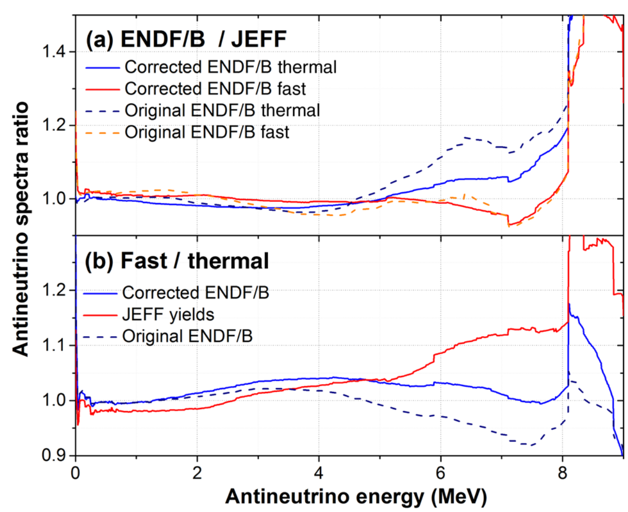

FIG. 3. (a) Ratio of ENDF/B-VII.1 to JEFF calculated ^{235}U(n,F) antineutrino spectra for thermal and fast neutron energies. (b) Ratio of fast to thermal ^{235}U(n,F) antineutrino spectra using corrected and original ENDF/B as well as JEFF yields.

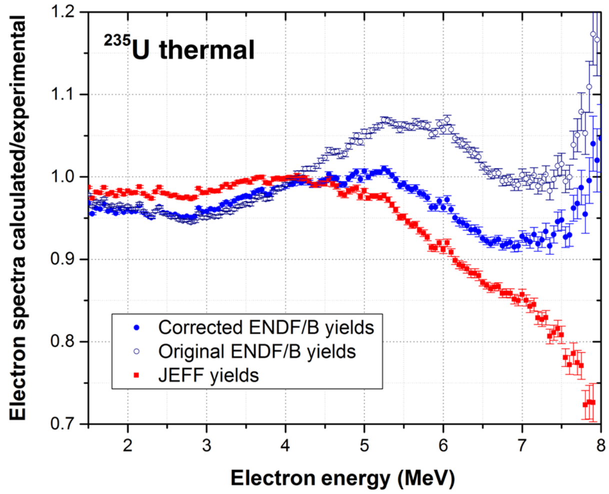

FIG. 4. Calculated thermal ^{235}U(n,F) electron spectrum divided by the experimental one [9], using the corrected and original ENDF/B-VII.1 yields as well as the JEFF yields. Only experimental uncertainties are included.

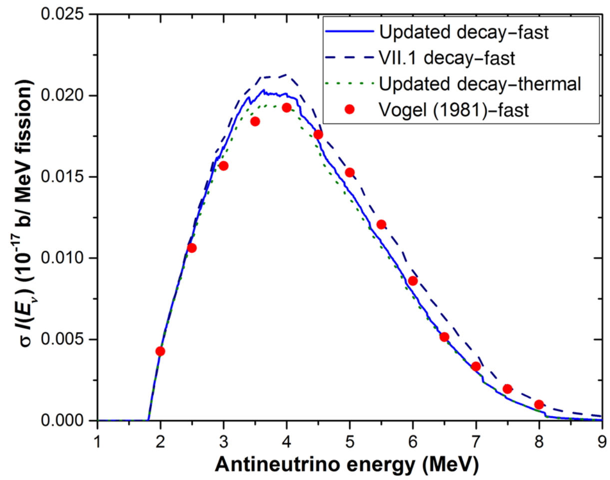

FIG. 5. ^{235}U(n,F) antineutrino spectra multiplied by the $\bar{\nu}+p\rightarrow n+e^{+}$ cross section for a combination of the ENDF/B yield and decay data choices: updated decay data with corrected fast (solid line) and thermal yields (dotted line), original ENDF/B-VII.1 decay data with corrected fast yields (dashed line), fast ENDF/B-V yields, and decay data (symbols) [12].

[13,16]; above its β-minus Q value, 8.095 MeV, the libraries differ greatly, reflecting that high Q values correspond to very neutron-rich nuclei where little to no experimental data exist. The fast to thermal ratio is plotted in Fig. 3(b); the use of corrected ENDF/B yields results in the fast spectrum being harder than the thermal one, in agreement with JEFF and with Ref. [12], which used ENDF/B-V yields where the ^{86}Ge yield was not anomalous.

Figure 4 shows the thermal ^{235}U calculated electron spectrum divided by the experimental one [9], using the original and corrected ENDF/B-VII.1 as well as the JEFF CFYs. Calculations using the corrected ENDF/B-VII.1 and JEFF CFYs agree reasonably well with the experimental values in the ∼3.5–5.5 MeV region. Everywhere else, these calculations underestimate the data, particularly for energies higher than 5.5 MeV, where the disagreement between the different fission yields results also increases. These calculations do not produce an excess of electrons anywhere in the spectrum. Using the original ENDF/B-VII.1 CFYs, on the other hand, produces an excess of electrons at 4–6 MeV, a feature which results mainly from the anomalous ^{86}Ge yield and is not a consequence of the underlying nuclear structure data.

The ^{235}U antineutrino spectrum folded with the $\bar{\nu}+p\rightarrow n+e^{+}$ cross section [42], $\sigma I(E_{\nu})$, is shown in Fig. 5. For fast neutrons, results are shown for three choices of yields and decay data: (a) updated ENDF/B-VII.1 decay and corrected ENDF/B-VII.1 yields, (b) ENDF/B-VII.1 decay and corrected ENDF/B-VII.1 yields, and (c) ENDF/B-V decay and yields [12]. The difference between (a) and (b) is due to the use of TAGS data, which leads to smaller $\sigma I(E_{\nu})$ values [14] due to "pandemonium" [43]. A comparison between (a) and (c) reflects the more precise data available today. Results using corrected thermal ENDF/B-VII.1 yields are also shown, leading to a ∼3% smaller integrated $\sigma I(E_{\nu})$, a result that may be important for HEU reactor experiments. As results from new TAGS measurements [17,44,45], beta spectra, and fission yields [46,47] become available, our ability to precisely calculate the antineutrino spectrum will improve further. Finally, we note that fission yields are available in ENDF/B or JEFF at two to four energy points, which is not enough to test the idea that nonthermal neutrons may cause the excess of antineutrinos at 5 MeV [18].

In summary, the ENDF/B-VII.1 ^{235}U thermal and fast fission yields were thoroughly reviewed and a number of corrections were applied, including a resolution of anomalous values as well as the use of updated isomeric ratios and decay probabilities. We explored the effect of these changes on the calculation of antineutrino spectra for thermal and fast ^{235}U fission. In the energy region of current interest, 5–7 MeV, the revision of the thermal ^{86}Ge yield and decay probabilities induces up to a 10% change in the calculated antineutrino spectrum. This result has major implications for prior calculations which found an excess of antineutrinos using the summation method in comparison to the conversion method. Our best summation calculations using either the corrected ENDF/B or JEFF CFYs with the updated decay data library produce a thermal electron spectrum that is mostly lower and never consistently exceeds the experimental one. The comparison between the thermal antineutrino spectra using two completely distinct fission yield libraries reveals differences of, at most, 6% up to 7 MeV, providing a low-fidelity estimate of the uncertainties introduced into summation calculations from the fission yield component. The fast antineutrino spectra calculated using these two sets of fission yields agree within 4% in the same energy region.

Work at Brookhaven National Laboratory was sponsored by the Office of Nuclear Physics, Office of Science of the U.S. Department of Energy under Contract No. DE-AC02-98CH10886. We are grateful to D. Brown and T. Kawano for useful comments, and to H-K. Schmidt for guidance with the GEF code.

[1] Y. Abe *et al.*, Phys. Rev. Lett. **108**, 131801 (2012).
[2] F. P. An *et al.*, Phys. Rev. Lett. **108**, 171803 (2012).
[3] J. K. Ahn *et al.* (RENO Collaboration), Phys. Rev. Lett. **108**, 191802 (2012).
[4] P. Huber, Phys. Rev. C **84**, 024617 (2011).
[5] Th. A. Mueller, D. Lhuillier, M. Fallot, A. Letourneau, S. Cormon, M. Fechner, L. Giot, T. Lasserre, J. Martino, G. Mention, A. Porta, and F. Yermia, Phys. Rev. C **83**, 054615 (2011).
[6] S.-H Seo (RENO Collaboration), AIP Conf. Proc. **1666**, 080002 (2015).
[7] F. P. An *et al.*, Phys. Rev. Lett. **116**, 061801 (2016).
[8] F. von Feilitzsch, A. A. Hahn, and K. Schreckenbach, Phys. Lett. **118B**, 162 (1982).
[9] K. Schreckenbach, G. Colvin, W. Gelletly, and F. Von Feilitzsch, Phys. Lett. **160B**, 325 (1985).
[10] A. A. Hahn, K. Schreckenbach, W. Gelletly, F. von Feilitzsch, G. Colvin, and B. Krusche, Phys. Lett. B **218**, 365 (1989).
[11] N. Haag, A. Gütlein, M. Hofmann, L. Oberauer, W. Potzel, K. Schreckenbach, and F. M. Wagner, Phys. Rev. Lett. **112**, 122501 (2014).
[12] P. Vogel, G. K. Schenter, F. M. Mann, and R. E. Schenter, Phys. Rev. C **24**, 1543 (1981).
[13] D. A. Dwyer and T. J. Langford, Phys. Rev. Lett. **114**, 012502 (2015).
[14] M. Fallot *et al.*, Phys. Rev. Lett. **109**, 202504 (2012).
[15] A. C. Hayes, J. L. Friar, G. T. Garvey, G. Jungman, and G. Jonkmans, Phys. Rev. Lett. **112**, 202501 (2014).
[16] A. A. Sonzogni, T. D. Johnson, and E. A. McCutchan, Phys. Rev. C **91**, 011301(R) (2015).
[17] A.-A. Zakari-Issoufou *et al.*, Phys. Rev. Lett. **115**, 102503 (2015).
[18] A. C. Hayes, J. L. Friar, G. T. Garvey, D. Ibeling, G. Jungman, T. Kawano, and R. W. Mills, Phys. Rev. D **92**, 033015 (2015).
[19] E. Christensen, P. Huber, P. Jaffke, and T. E. Shea, Phys. Rev. Lett. **113**, 042503 (2014).
[20] A. Algora *et al.*, Phys. Rev. Lett. **105**, 202501 (2010).
[21] M. E. Gooden *et al.*, Nucl. Data Sheets **131**, 319 (2016).
[22] G. Mention, M. Fechner, Th. Lasserre, Th. A. Mueller, D. Lhuillier, M. Cribier, and A. Letourneau, Phys. Rev. D **83**, 073006 (2011).
[23] J. Ashenfelter *et al.* (PROSPECT Collaboration), Nucl. Instrum. Methods Phys. Res., Sect. A **806**, 401 (2016).
[24] M. B. Chadwick *et al.*, Nucl. Data Sheets **112**, 2887 (2011).
[25] M. A. Kellett, O. Bersillon, and R. W. Mills, OECD Nuclear Energy Agency JEFF Report No. 20, 2009.
[26] K. Shibata *et al.*, J. Nucl. Sci. Technol. **48**, 1 (2011).
[27] T. Miyazono *et al.*, JAERI-CONF Report No. 97-005, 1997.
[28] W. B. Wilson *et al.*, Los Alamos National Laboratory Report No. LA-UR-07-8412, 2008.
[29] G. Chiba, M. Tsuji, and T. Narabayashi, Nucl. Data Sheets **118**, 401 (2014).
[30] K.-H. Schmidt, B. Jurado, C. Amouroux, and C. Schmitt, Nucl. Data Sheets **131**, 107 (2016).
[31] G. Rudstam, P. Aagaard, B. Ekström, E. Lund, H. Göktürk, and H. U. Zwicky, Radiochim. Acta **49**, 155 (1990).
[32] T. R. England and B. F. Rider, Los Alamos National Laboratory Report No. LA-UR-94-3106, 1994.
[33] M. Bernas *et al.*, Phys. Lett. B **331**, 19 (1994).
[34] M. Bernas *et al.*, Phys. Lett. B **415**, 111 (1997).
[35] D. G. Madland and T. R. England, Nucl. Sci. Eng. **64**, 859 (1977).
[36] A. Bail, O. Serot, L. Mathieu, O. Litaize, T. Materna, U. Köster, H. Faust, A. Letourneau, and S. Panebianco, Phys. Rev. C **84**, 034605 (2011).
[37] See http://www.nndc.bnl.gov/ensdf.
[38] R. C. Greenwood, R. G. Helmer, M. H. Putnam, and K. D. Watts, Nucl. Instrum. Methods Phys. Res., Sect. A **390**, 95 (1997).
[39] G. Rudstam, P. I. Johansson, O. Tengblad, P. Aagaard, and J. Eriksen, At. Data Nucl. Data Tables **45**, 239 (1990).
[40] T. Kawano, P. Moller, and W. B. Wilson, Phys. Rev. C **78**, 054601 (2008).
[41] L. A. Metz, J. I. Friese, E. C. Finn, L. R. Greenwood, R. F. Kephart, C. C. Hines, M. D. King, K. M. Henry, and D. E. Wall, J. Radioanal. Nucl. Chem. **296**, 763 (2013).
[42] A. Strumia and F. Vissani, Phys. Lett. B **564**, 42 (2003).
[43] J. C. Hardy, L. C. Carraz, B. Jonson, and P. G. Hansen, Phys. Lett. **71B**, 307 (1977).
[44] K. P. Rykaczewski, Nucl. Data Sheets **120**, 16 (2014).
[45] J. L. Tain *et al.*, Phys. Rev. Lett. **115**, 062502 (2015).
[46] K. Meierbachtol, F. Tovesson, C. W. Arnold, A. B. Laptev, T. A. Bredeweg, M. Jandel, R. O. Nelson, and M. C. White, Nucl. Data Sheets **119**, 389 (2014).
[47] H. Pentila *et al.*, Nucl. Data Sheets **119**, 334 (2014).