Prompt fission neutron multiplicity in the ^{239}Pu(n, f) reaction and its energy dependence

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Abstract. Measuring prompt fission neutrons to high precision is an experimental challenge, especially for radioactive fissioning nuclides. However, accurate average multiplicities, $\bar{\nu}_p$, and kinetic energy distributions of prompt fission neutrons are essential for fundamental and applied nuclear physics. We present here a recent measurement of the ^{239}Pu (n,f) $\bar{\nu}_p$ as a function of the incident-neutron energy, over the range 1-700 MeV. The measurement was performed with a cutting-edge setup and an innovative technique, which allowed to minimize and account for the main sources of bias. An unprecedented precision was therefore achieved. Our data are compared to GEF predictions as well as to evaluated libraries. For the first time, at low energies, the ENDF/B-VIII.0 nuclear data evaluation is validated with an independent measurement and the evaluated uncertainty reduced by up to 60%. This work paves the way to precisely measure prompt fission neutron multiplicities on highly radioactive nuclei.

A full understanding of the nuclear fission process, a rich quantum phenomenon, is still a challenge both from theoretical and experimental points of view, despite its discovery being 80 years old. Worldwide efforts [1, 2, 3] aim at a deeper understanding of this phenomenon and support the development of new nuclear technologies for energy production. Precise measurements of observables over large energy ranges will set the most stringent constraints to theoretical models. In particular, relevant information on the total amount of the fissioning-system excitation energy transferred to the fragments, on its sharing between excitation and kinetic energies of each fragment, and on the fission fragment excitation energy due to its extreme deformation is provided by the number of emitted prompt fission neutrons, $\bar{\nu}_p$. Moreover, precise $\bar{\nu}_p$ data for the ^{235,238}U and ^{239}Pu nuclide are crucial ingredients to calculate criticality, efficiency, safety, and lifetime of next-generation nuclear reactors. To quantify the required accuracy, let us consider a Pu reactor assembly: a change in ^{239}Pu (n,f) $\bar{\nu}_p$ by 0.1% in a 100 keV energy range changes the computed criticality by about one third of the range between a controlled and an uncontrolled assembly [4, 5], i.e. about 100 pcm.

Theoretical models nowadays lack of the needed accuracy in $\bar{\nu}_p$ predictions. Therefore, nuclear data applications mainly rely on evaluated data, such as ENDF/B-VIII.0 and JEFF3.3 [2, 3].

To test their reliability for applications, integral experiments are used to validate libraries. The case of the ^{239}Pu (n,f) $\bar{\nu}_p$ is peculiar, as ENDF/B-VIII.0 ^{239}Pu (n,f) $\bar{\nu}_p$ were obtained from existing experimental data, but the evaluated data had to be adjusted [2] to obtain a good agreement between simulated and experimental criticality, k_{eff}. The relative difference between existing experimental and ENDF/B-VIII.0 $\bar{\nu}_p$ values in the fast neutron energy region, shown in Fig.1, is as high as 2% below 8 MeV, with data systematically lower than the most recent ENDF/B-VIII.0 evaluation. However, it should be noted that all the plotted data but Hopkins et al. [6] were obtained with the same experimental technique, i.e. using 4π scintillator tanks. This kind of measurement allows one to collect very high statistics and makes of them the most precise available ^{239}Pu (n,f) $\bar{\nu}_p$ values. However, the technique prevents the measurement of the emitted-neutron angular distribution. Moreover, as the emitted-neutron energy is also not experimentally measured, an average rather than an energy-dependent scintillator-detector efficiency is used in the data analysis.

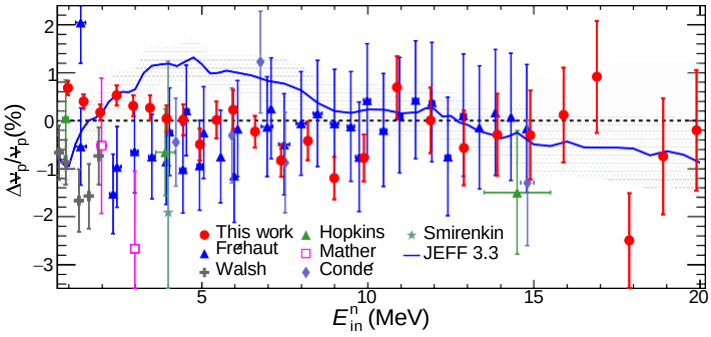

Figure 1. (Color online) (Color online) Relative difference between experimental and ENDF/B-VIII.0 $\bar{\nu}_p$ values from this work and previous experiments [6, 7, 8, 9, 10, 11]. The difference with the JEFF3.3 evaluation [3] is also shown.

In this work we report on high precision experimental data, obtained with a different technique, with the aims of i) providing an independent measurement ii) reducing the existing uncertainties and iii) correcting for all possible experimental bias. The double time-of-flight technique was used, for the first time for the highly radioactive ^{239}Pu isotope, at the high-intensity, pulsed and well-collimated white neutron source of the Weapons Nuclear Research facility [12, 13] of the Los Alamos Neutron Science Center at the Los Alamos National Laboratory. To detect neutrons emitted in fission events 54 EJ-309 [15] liquid scintillators from the Chi-Nu array [16] were coupled to a newly-developed, fast, high-efficiency, light-weight fission chamber [14]. The fission chamber was developed to achieve an improved discrimination between fission and α-decay events, despite their very different rates of about 15 events/s and 10 MBq/channel, respectively. A 95% fission-fragment detection efficiency is achieved. This feature allowed us to measure the whole fission-fragment angular and kinetic energy distributions and avoid the data bias due to an incomplete selection of the detected fragments. The highly-segmented Chi-Nu array allowed us to measure the neutron angular distribution, and therefore to precisely correct for the contribution of regions not covered by the detector. A detailed description of the experimental setup can be found in [14, 16, 17, 18]. The innovative setup and the high collected statistics allowed us to precisly reconstruct prompt fission neutron spectra (PFNS) for each incident-neutron energy E^n_{inh} from 0.7 to 700 MeV [17]. The measurement of PFNS allowed us to correct the data for an energy-dependent detector efficiency. The $\bar{\nu}_p$ values were extracted from the PFNS as discussed in Ref.[19].

The capability of properly estimating the sources of possible systematic bias is the main improvement with respect to previous measurements. Four different experimental biases were corrected for in the data: the limited detector angular coverage, the neutron detection energy range, the detector dead time and the presence of a slower incident neutron background (wrap-around) [20]. The procedures used to account for these biases are described in detail in Ref.[19].

Here we only recall the relative importance of these biases. The main correction to the $\bar{\nu}_p$ values for E^n_{in} up to 10 MeV comes from the limited angular coverage of the neutron detectors. It modifies the $\bar{\nu}_p$ value by up to 3%. The correction is stronger for forward and backward angles, explaining why it is relevant also for scintillator tank experiments. Below 200 MeV the wrap-around effect modifies the $\bar{\nu}_p$ value by up to 6%, with a maximum at 20 MeV. Above 200 MeV the neutron-detector dead time becomes predominant. The lower limit of the neutron detection energy range is set by the threshold for discriminating neutrons from γ-rays and, as expected, contributes more at the opening of the n-chance fission, where slower neutrons are emitted from the compound system before fission. The high-energy limit is related to the dynamic range of the electronics and corresponds emitted-neutron energies of ≈ 14 MeV. Neutrons above this energy are prediced by TALYS [21] to significantly modify the $\bar{\nu}_p$ value for E^n_{in} above 24 MeV. Therefore, $\bar{\nu}_p$ values for E^n_{in} above this energy should be considered as a lower limit.

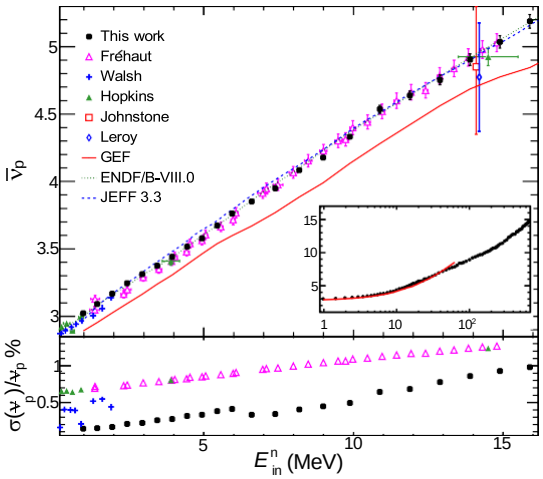

Figure 2. (Color online) Measured $\bar{\nu}_p$ and its uncertainty as a function of E^n_{in} up to 16 MeV – and over the whole studied E^n_{in} energy range, in the insert. Some data from previous experiments are shown [10, 11, 6, 22, 23]. Dotted, dashed and full lines are ENDF/B-VIII.0 and JEFF3.3 evaluations, respectively, and GEF predictions.

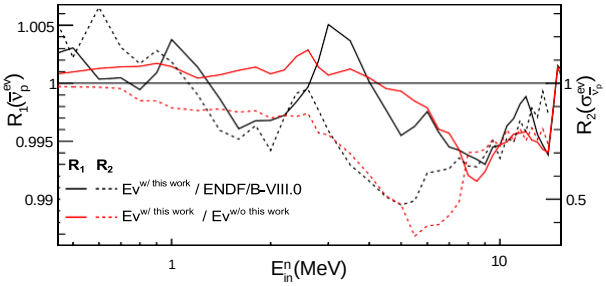

Figure 3. (Color online) Ratio of $\bar{\nu}_p$ evaluated mean value (R_1, full lines, left axis) and relative uncertainty (R_2, dashed lines, right axis) for different evaluations (see text).

The data after corrections are shown as a function of E^n_{in} in Figs. 1 and 2. As expected, our data constantly increase up to 700 MeV with no clear structure. Below about 14 MeV, $\bar{\nu}_p$ depends linearly on the neutron energy. It is worth noting that the obtained $\bar{\nu}_p$ total uncertainties vary from 0.15 to 1.3%, and, below 14 MeV E^n_{in} , are smaller than 1%. The bottom panel of Fig. 2 compares the achieved uncertainties to the most precise previous measurement: we observe that such low uncertainties on a broad energy range were never reached before, not even with different experimental techniques ([8, 9, 10, 11, 24, 25, 26, 27, 28, 29] and [6, 30, 31]). At low E^n_{in} , below 5 MeV, the averaged relative difference between our data and ENDF/B-VIII.0 values, shown in Fig. 1, is of 0.3%. The observed agreement with the ENDF/B-VIII.0 evaluation provides, for the first time, an independent validation of the evaluation itself.

Our data are compared to the semi-empirical model GEF [32] in Fig.2. They are reproduced by GEF predictions within 0.15 (4.5%) and 0.4 (8%) neutrons per fission below 8 MeV and over

the full energy range [1 − 25] MeV, respectively. The difference of 0.15 and 0.4 neutrons per fission corresponds, in the GEF model, to a "wrong" sharing between fission-fragment kinetic and excitation energies of about 1 and 2.8 MeV, respectively, to be compared to about 200 MeV released in fission, thus validating the implemented sharing model. It should be stressed that, although the model predictions can't be used in evaluations as they are too different from experimental data, GEF is not tuned to these experimental data.

Two new evaluations of ^{239}Pu $\bar{\nu}_p$ were performed using the same methodology, one with ($Ev^{w/\ this\ work}$) and the other without including our data ($Ev^{w/o\ this\ work}$). In the 1 to 15 MeV range, the inclusion of our data reduce the ENDF/B-VIII.0 and $Ev^{w/o\ this\ work}$ $\bar{\nu}_p$ evaluated relative uncertainty, $\sigma^{ev}_{\bar{\nu}_p}$, by up to 50% and 60%, respectively (see dashed lines in Fig.3). Moreover the $\bar{\nu}_p$ evaluated mean value, $\bar{\nu}^{ev}_p$ below 5 MeV is modified by less than 0.15% by our data. The importance of such agreement is due to the fact that the ENDF/B-VIII.0 $\bar{\nu}^{ev}_p$ values were obtained by an average over previous data measured all by the same technique, which is different from the one used here. Such validation of the ENDF/B-VIII.0 $\bar{\nu}^{ev}_p$ was missing up to now.

In conclusion, previously unattained accurate and precise new data on ^{239}Pu $\bar{\nu}_p$ obtained with the double time-of-flight technique and an innovative setup are reported, extending the range from 1 up to 700 MeV. Experimental systematic biases, which have limited the precision and accuracy of existing experimental results, are explicitly accounted for. Below 5 MeV the observed good agreement with the recent ENDF/B-VIII.0 evaluation validates, for the very first time, with an independent measurement, the evaluation itself. The impact of these data on a new evaluation performed here shows that they significantly reduce the uncertainty on evaluated nuclear-data libraries for the ^{239}Pu , a key isotope for nuclear energy applications. Reduced uncertainties lead to an increased predictive power of neutronics calculations.

The innovative setup and experimental technique fulfill the experimental challenge of precisely measuring prompt fission-neutron multiplicity on highly radioactive nuclei. These results pave the way to precisely investigate other high-activity actinide nuclei to provide key elements for the development of new technologies while contributing to a better understanding of the fission process.

Acknowledgments

This work was performed under the auspices of a cooperation agreement between CEA/DAM and DOE/NNSA on fundamental sciences and benefited from the use of the LANSCE accelerator facility. Work at LANL was carried out under the auspices of the NNSA of the U.S. Department of Energy under contract 89233218CNA000001. We gratefully acknowledge partial support of the Advanced Simulation and Computing program at LANL and the DOE Nuclear Criticality Safety Program, funded and managed by NNSA for the DOE.

References

[1] Schmidt K H and Jurado B 2018 *Reports on Progress in Physics* 81 106301
[2] Brown D A *et al.* 2018 *Nucl. Data Sheets* 148 1
[3] OECD/NEA JEFF3.3 Evaluated data Library - Neutron Data Tech. rep.
[4] Neudecker D *et al.* 2020 *Nuclear Data Sheets* 167 36 – 60
[5] Neudecker D *et al.* 2021 *Ann. Nucl. Energy* 159 108345
[6] Hopkins J and Diven B 1963 *Nucl. Phys.* 48 433 (EXFOR entry n. 12326:)
[7] Smirenkin G N, Bondarenko I I, Kutsaeva L S, Mishchenko K D, Prokhorova L I and Shemetenko B P 1958 *The Soviet Journal of Atomic Energy* 4 253 (EXFOR entry n. 40388)
[8] Mather D S, Fieldhouse P and Moat A 1965 *Nucl. Phys.* 66 149 and *A.W.R.E. Aldermaston Reports*, No.42/70, 1970. (EXFOR entries n. 20453 and 21135)
[9] Cond´e H, Hans´en J and Holmberg M 1968 *Jour. of Nucl. Ener.* 22 53 (EXFOR entry n. 20052)

[10] Fr´ehaut J, Mosinski G and Soleilhac M 1973 *EANDC Topical Conference* vol 154 p 67 (EXFOR entry n. 20490)
[11] Walsh R L and Boldeman J W 1971 *Jour. of Nucl. Ener.* 25 321 (EXFOR entries n. 30006 and 30046)
[12] Lisowski P W, Bowman C D, Russell G J and Wender S A 1990 *Nucl. Sci. Eng.* 106 208
[13] Lisowski P W and Schoenberg K F 2006 *Nucl. Instrum. Methods Phys. Res., Sect. A* 562 910
[14] Laurent B, Taieb J, B´elier G, Marini P and Morfouace P 2021 *Nucl. Instr. and Meth. in Phys. Res. A* 990 164966
[15] Eljen Technology https://eljentechnology.com
[16] Haight R C *et al.* 2012 *J. Inst.* 7 C03028
[17] Marini P *et al.* 2020 *Phys. Rev. C* 101 044614 and Refs. therein
[18] Taieb J, Laurent B, B´elier G, Sardet A and Varignon C 2016 *Nucl. Instrum. Methods Phys. Res., Sect. A* 833 1 French patent for the anode concept n. 1757033 deposited on July 25th, 2017
[19] Marini P *et al.* 2022 *Phys. Lett. B* 835 137513
[20] Los Alamos Neutron Science Center https://lansce.lanl.gov/facilities/time-of-flight.php
[21] Koning A, Hilaire S and Goriely S 2019 *TALYS-1.95 A nuclear reaction program, User Manual* www.talys.eu/documentation
[22] Leroy J 1960 *J. Phys. Radium* 21 617 (EXFOR entry n. 12397)
[23] Johnstone I 1956 *A.E.R.E. Harwell Reports, No. 1912* (EXFOR entry n. 21696)
[24] Diven B C and Hopkins J C 1962 *Proc. of the Seminar on Physics of Fast and Intermediate Reactors* (EXFOR entry n. 12337)
[25] Gwin R, Spencer R R and Ingle R W 1986 *Nucl. Sci. Eng.* 94 365 (EXFOR entry n. 13101)
[26] Jacob M 1958 *Centre d'Études Nucl´eaires report series* 652 17 (EXFOR entry n. 21484)
[27] Khokhlov Y A, Savin M V, Ludin V N and Lebedev B L 1976 *Yadernye Konstanty* vol 23 p 3 (EXFOR entry n. 40523)
[28] Savin M V, Khokhlov Y A, Zamjatnin Y S and Paramonova I N 1970 *Nuclear Data for Reactors Conf., Helsinki* vol 2 p 157 paper in Russian. (EXFOR entry n. 40058)
[29] Soleilhac M, Fr´ehaut J, Gauriau J and Mosinski G 1970 *Nuclear Data for Reactors Conf., Helsinki* vol 2 p 145 paper in French (EXFOR entry n. 20568)
[30] Kalashnikova V I, Lebedev V I and Spivak P E 1957 *Soviet Atomic Energy* 2 17 and Atomnaya Energiya 2, 18 (1957). (EXFOR entry n. 40356) URL http://dx.doi.org/10.1007/BF01480701
[31] Volodin K E, Kuznetsov V F, Nesterov V G, Nurpeisov B, Prokhorova L I, Turchin Y M and Smirenkin G N 1972 *Atomic Energy* 33 901 (EXFOR entry n. 40148)
[32] Schmidt K H, Jurado B, Amouroux C and Schmitt C 2016 *Nucl. Data Sheets* 131 107 – 221