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Multiplicity of prompt fission neutron in the ^{239}Pu(n,f) reaction and its energy dependence

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Abstract. Accurate multiplicities of prompt fission neutrons emitted in neutron-induced fission on a large energy range are essential for fundamental and applied nuclear physics. Measuring them to high precision for radioactive fissioning nuclides is, however, an experimental challenge. In this work, we extract the average prompt-neutron multiplicity emitted in the $^{239}\mathrm{Pu}\,(n, f)$ reaction as a function of the incident-neutron energy, over the range 0.7-700 MeV. We used a novel technique, which allowed us to minimize and correct for the main sources of bias and thus achieve unprecedented precision. At low energies, our data validate, for the first time, the ENDF/B-VIII.0 nuclear data evaluation with an independent measurement and reduce the evaluated uncertainty by up to 60%. This work opens up the possibility of measuring, with high precision, prompt fission neutron multiplicities on highly radioactive nuclei relevant for energy production.

1 Introduction

The nuclear fission process was discovered about 80 years ago. A full understanding of this quantum process is, however, still a challenge both from experimental and theoretical points of view. The development of new nuclear technologies for energy production worldwide has boosted efforts [1–3] to better understand this phenomenon. Measurements of observables over large energy ranges are expected to constraint most theoretical models. In particular, the number of emitted prompt fission neutrons, $\overline{\nu}_p$ provide information on the total amount of the fissioning-system excitation energy transferred to the fragments and on its sharing between excitation and kinetic energies of each fragment. Moreover, precise $\overline{\nu}_p$ data for the ^{239}Pu and ^{235,238}U nuclides are mandatory to calculate efficiency, safety, criticality and lifetime of next-generation nuclear reactors. The required uncertainty for this quantity is less than 0.1%.

Theoretical models currently lack of the needed accuracy in $\overline{\nu}_p$ predictions. Nuclear data applications, therefore, mainly rely on evaluated data, such as ENDF/B-VIII.0 and JEFF3.3 [2, 3], which are validated against integral experiments.

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In the case of the ^{239}Pu (n,f) $\overline{\nu}_p$ ENDF/B-VIII.0 values 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}. However, the large majority of experimental data were obtained with the same experimental technique, i.e. using 4π scintillator tanks.

In this work we report on high precision experimental data, obtained with a different technique, the double time-of-flight technique, with the goals of a) reducing the existing uncertainties b) providing an independent measurement and c) correcting for all possible experimental bias.

The measurement was carried out at the high-intensity, pulsed and well-collimated white neutron source of the Weapons Nuclear Research facility [4, 5] of the Los Alamos Neutron Science Center at the Los Alamos National Laboratory. The 54 EJ-309 [6] liquid scintillators from the Chi-Nu array [7] were used to detect neutrons emitted in fission events, and coupled to a newly-developed, fast, high-efficiency, light-weight fission chamber [8]. The fission chamber characteristics 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 neutron angular distribution was measured with the segmented Chi-Nu array, thus we could correct for the contribution of regions not covered by the detector. A detailed description of the experimental setup can be found in [7–10]. Prompt fission neutron spectra (PFNS) for each incident-neutron energy E^n_{in}, from 0.7 to 700 MeV were precisely reconstructed as discussed in [9]. The main improvement with respect to previous measurements is the possibility of properly estimating the sources of possible systematic bias. The data were corrected for i) the limited detector angular coverage, ii) the neutron detection energy range, iii) the detector dead time and iv) the presence of a slower incident neutron background (wrap-around) [11]. The used procedures are described in detail in Ref.[12]. Here we only present the final results.

2 Results

The data after all corrections are shown in Fig. 1 as a function of E^n_{in}. As expected, our data constantly increase up to 700 MeV without any clear structure. Below about 14 MeV, $\overline{\nu}_p$ depends linearly on the neutron energy. The obtained $\overline{\nu}_p$ 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. 1 compares the achieved uncertainties to the most precise previous measurement: uncertainties as low as in this work on a broad energy range were never reached before, not even with different experimental techniques ([13, 14, 18–25] and [15, 26, 27]). At low E^n_{in}, below 5 MeV, the averaged relative difference between our data and ENDF/B-VIII.0 values is of the order of 0.3%. The very good 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 [28] in Fig.1. Our values are in agreement with 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 mentioned difference of 0.15 and 0.4 neutrons/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. Given the differences observed with GEF, the model predictions can’t be used in evaluations, however it should be stressed that GEF is not tuned to these experimental data.

Two new evaluations of ^{239}Pu $\overline{\nu}_p$ were performed using the same methodology, one with ($Ev^{w/\,this\,work}$) and the other without including our data ($Ev^{w/o\,this\,work}$) [12]. In the 1 to 15 MeV range, the inclusion of our data reduces the ENDF/B-VIII.0 and $Ev^{w/o\,this\,work}$ $\overline{\nu}_p$ evaluated relative uncertainty, $\sigma^{ev}_{\overline{\nu}_p}$, by up to 50% and 60%, respectively (see dashed lines in Fig.2).

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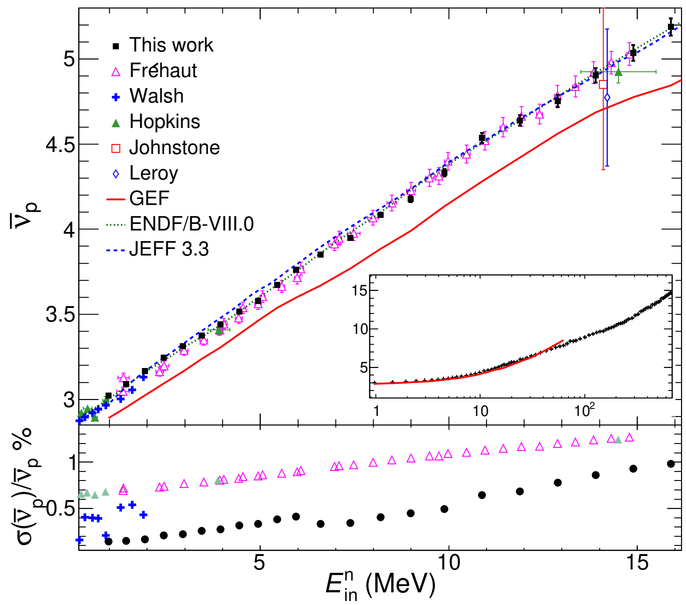

Figure 1. (Color online) Measured $\overline{\nu}_p$ and its uncertainty as a function of E^n_{in} up to 15 MeV – and over the whole studied E^n_{in} energy range, in the insert. Data from some previous experiments are shown [13–17]. Dotted, dashed and full lines are ENDF/B-VIII.0 and JEFF3.3 evaluations, respectively, and GEF predictions.

The $\overline{\nu}_p$ evaluated mean value, $\overline{\nu}_p^{ev}$ below 5 MeV is modified by less than 0.15% by our data. We recall that the ENDF/B-VIII.0 $\overline{\nu}_p^{ev}$ values were obtained by an average over previous data measured all by the same technique, which is different from the one used here. We therefore validate, for the first time, the ENDF/B-VIII.0 $\overline{\nu}_p^{ev}$ evaluation.

3 Conclusions

We have reported on previously unattained precise new data on ^{239}Pu $\overline{\nu}_p$ over the range from 0.7 up to 700 MeV obtained with the double time-of-flight technique and an innovative setup. 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 ENDF/B-VIII.0 evaluation validates, with an independent measurement, the evaluation itself, for the very first time. The new evaluation performed here shows the impact of our data: that they significantly reduce the uncertainty on evaluated nuclear-data libraries for the ^{239}Pu . Reduced uncertainties lead to an increased predictive power of neutronics calculations for a key isotope for nuclear energy applications.

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.

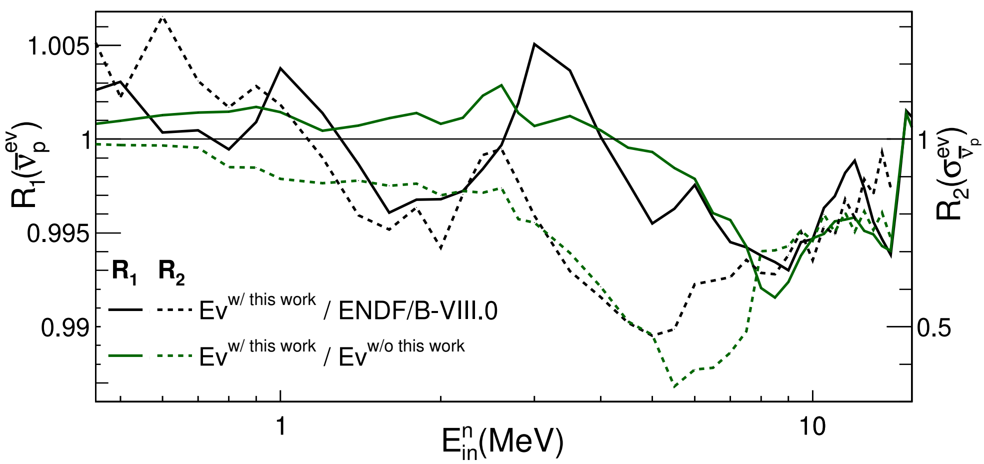

Figure 2. (Color online) Ratio of $\overline{\nu}_p$ evaluated mean value (R_1, full lines, left axis) and relative uncertainty (R_2, dashed lines, right axis) for different evaluations (see text).

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