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Prompt fission γ-rays from the reactions ^{252}Cf(SF) and ^{235}U(n$_{th}$, f) – new data

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Abstract. We present new spectral data of prompt γ-ray emission from the spontaneous fission of ^{252}Cf. This work was performed in direct response to an OECD/NEA high priority data request. We discuss the impact of our new data on evaluated nuclear data tables not only for this nuclide, but also for ^{238}U and ^{241}Pu, which are always produced in a reactor. Furthermore, we will show results from our investigation of prompt γ-ray emission from the reaction ^{235}U(n$_{th}$, f), measured in at the Centre for Energy Research of the Hungarian Academy of Sciences in Budapest, Hungary. Spectral data obtained with three different detectors are consistent and led to an uncertainty on total energy and multiplicity considerably smaller than requested by the OECD/NEA.

1. Introduction

Since four out of six of the impending Gen-IV reactors that have been selected by the Generation-IV International Forum (GIF) are fast reactors, an innovative core design is required to be able to handle the excessive heat deposit from the fission process [1]. In order to model these cores, a better understanding of the released heat from the common reactor isotopes is crucial. With the potential of more advanced nuclear reactors in the near future, a better understanding of the entire fission process is needed. Present knowledge regarding this heat deposit implies that approximately ten percent of the total energy released in fission is due to γ-rays, of which around forty percent of the heat originates from prompt fission γ-rays [2]. According to Refs. [1, 3] it is necessary to achieve an uncertainty of at most 7.5% in regard of the calculated γ-heating in order to adequately model these cores. However, with evaluated data the γ-heating is underestimated by up to 28% for the main reactor isotopes ^{235}U(n,f) and ^{239}Pu(n,f). Therefore, these two isotopes have been included in the OECD Nuclear Energy Agency high priority request list for prompt fission γ-rays data, in particular new values for γ-multiplicity and mean energy [4] are requested. The data in the evaluated data tables for both isotopes relies on results that were

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measured in the early 1970's [5, 6], and was recently confirmed by Kwan et al. [7]. Hence, it might be more likely that the underestimation comes from the isotopes ^{238}U(n,f) and ^{241}Pu(n,f), which are always produced in a reactor [8]. The evaluated data for those two isotopes exhibit exactly the same structure, with an individual scaling factor; and the same formula is also used for ^{252}Cf. Accordingly it seems that no experimental data has been used to evaluate neither of these three isotopes. Since the 1970's a lot has happened in regard of detector development, especially with the release of new lanthanide-halide scintillation detectors (see e.g. [9, 10] and ref. therein), as well as of data acquisition and signal processing technique's [11, 12]. Consequently, we wanted to take advantage of these advancements towards high-quality measurements of the γ-decay heat from the fission process. For an independent verification of the historical data, we performed an experiment on prompt γ-rays from neutron-induced fission of ^{235}U. To be able to accurately measure these isotopes we need to be certain of the quality of our experimental setup. Therefore, we started with studying the spontaneous fission of ^{252}Cf. Since this reaction has been measured both in the early 1970's by Verbinski et al. [5] and Pleasonton et al. [6], as well as very recently by Chyzh et al. with the DANCE spectrometer at Los Alamos National Laboratory [13], it serves as excellent proof of principle. The spectral data from our recent measurements are published in Refs. [14, 16] and are summarized here.

2. Experiment characteristics

In order to minimize the uncertainty in determining the γ-ray multiplicity and total released energy, three important detector characterisitics ought to be considered: (1) energy resolution, in order to determine the structure of the emission spectra with good precision, (2) intrinsic full peak efficiency, in order to decrease the uncertainty of the response function, and (3) timing resolution, in order to efficiently seperate prompt fission γ-rays from prompt neutrons by means of time of flight. We decided to use cerium-doped lanthanum-halide ($LaBr_3 : Ce$) and cerium-halide scintillation detectors ($CeBr_3$), which all have been carefully characterized at our institute ([10, 14] and Refs. therein).

2.1 Spontaneous fission of ^{252}Cf

A simplified ionization chamber (IC) loaded with a ^{252}Cf source served as fission trigger. The IC was built from very thin walls, i.e. 0.2 mm, to minimize the γ-ray background due to interaction with prompt fission neutrons and operated with P10-gas (mixture of 90% Ar and 10% CH_4, p = 1.2 bar) [15]. The californium was deposited on a polished stainless steel disc with 25 mm diameter. The source distributed on a spot of 10 mm diameter had an activity of (8600 ± 100) fissions/s and was mounted inside on the cathode, giving a signal for every fission event. Both scintillation detector were placed at a distance of 63 cm from the fission source. The distance between the detector and the source was determined with respect to the specific timing resolution of each detector in combination with the IC, i.e. $\sigma_{TOF} < 1.5$ ns, to make sure that there was a sufficient time difference between the prompt γ-peak and the fast neutron interactions. In this demonstration experiment the distance was a compromise to have optimum suppression of the prompt fission neutron component and count-rate. Details on experimental setup may be found in Ref. [14].

2.2 Thermal neutron-induced fission of ^{236}U*

Two experiments were performed at the 10 MW research reactor of KFKI Budapest, Hungary, in order to measure prompt fission γ-rays from the reaction $n_{th} + {}^{235}$U. In both, a uranium sample was exposed to the cold neutron beam and γ-rays were measured in coincidence with fission fragments, although different instrumentation was used, as described below and in Ref.[16] in more detail.

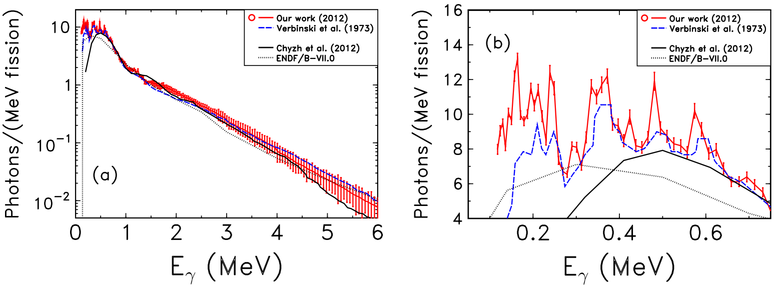

Figure 1. (a) The prompt fission γ-ray emission spectrum from this work taken with a 2 in. × 2 in. $LaBr_3$:Ce detector (full red line) is shown together with data from Refs. [5, 13] as well as from ENDF/B-VII for comparison. In the high energy range, all spectra agree rather well with each other, but the data from both ENDF/B-VII.0 [21] and Ref. [13] lacks structure in the low energy range. This is even more obvious in the lower part (b), which focusses on γ-ray energies below 0.8 MeV, and demonstrates on the other hand the very good reproduction of the historical data from Ref. [5] (see text for details and Ref. [14], from which the figure was taken).

In the first measurement campaign that took place during ten days of beam time in February/March 2010, photons were detected with a 3 in. ×3 in. coaxial $LaCl_3$:Ce scintillation detector, located at a distance of about 30 cm from a very thin ^{235}U target with an effective mass of 113 μg, mounted on a 34 $\mu g/cm^2$ thick polyimide backing. The sample itself was placed in the fission fragment spectrometer VERDI [17, 18]. The fast fission trigger was provided by a polycrystalline chemical vapor deposited (pCVD) diamond detector of size 1 cm × 1 cm, placed directly on top of the uranium sample (distance approximately 1 mm) [19].

The second experiment was carried out in June 2012 with about six days of actual beam time. An ultra-thin spectroscopic uranium sample (thickness 91.5 $\mu g/cm^2$) of effective mass 640 μg was placed inside a twin Frisch-grid ionization chamber, which delivered the fission trigger. The coincident measurement of photons was accomplished with two coaxial $LaBr_3$:Ce as well as two coaxial $CeBr_3$ scintillation detectors simultaneously, whose size (diameter × length) is 2 in. ×2 in. and 1 in. ×2 in., respectively.

In both measurements, pulse height and time-of-flight of the coincident events in each scintillation detector were recorded and stored in list mode. The first one gives, after proper calibration with different γ-sources, the energy deposited in the detector, while the second enables us to discard all events that arrived later in time than expected from a prompt γ-ray, e.g. due to neutron-induced reactions.

3. Prompt fission γ-ray spectral data

For deducing the emitted prompt fission γ-ray spectrum, the measured spectra have to be corrected with the response function of each detector. These response functions were determined by means of Monte Carlo simulations with the PENELOPE2011 computer code [20], folded with the energy resolution of the corresponding detectors. In Ref. [14], this as well as the actual extraction of the emission spectrum is described in detail. The unfolded prompt fission γ-ray spectra from the spontaneous fission of ^{252}Cf are shown in Figure 1. Only the spectral data obtained with the $LaBr_3$(Ce) detector is shown and compared to experimental data from [5] perfectly verifying the historical data. Even the distinct structure below $E_\gamma < 0.7$ MeV is well reproduced, and our measurements extends to somewhat lower energy. From Fig. 1 it is also evident, that the ENDF/B-VII.0 evaluation [21] deviates severely from our and the historic data. The recently released ENDF-/B-VII.1 evaluation still underestimates the total prompt γ-ray energy release by 9%. Chyzh et al. data [13] are consistent with our data, however, suffer from a much higher low-energy threshold. All characteristic prompt fission γ-ray spectral data are summarized in Table 1.

Table 1. Overview of results for the spontaneous fission of ^{252}Cf. The experimental results from this work for the prompt fission γ-ray multiplicity ν_γ, the average energy ϵ_γ and the total energy $E_{\gamma,tot}$ are averaged over both detectors employed in this work, and are compared to previously measured values from Refs. [5, 6, 13] as well as corresponding numbers from the evaluated nuclear data files in ENDF/B-VII.0 [21].

Results:	ν_γ (per fission)	ϵ_γ (MeV)	E_γ, tot (MeV)
This work	8.31 ± 0.09	0.80 ± 0.02	6.65 ± 0.10
Verbinski et al. [5]	7.80 ± 0.30	0.88 ± 0.04	6.84 ± 0.30
Pleasonton et al. [6]	8.32 ± 0.40	0.85 ± 0.06	7.06 ± 0.35
Chyzh et al. [13]	8.14 ± 0.4	0.94 ± 0.05	7.65 ± 0.55
ENDF/B-VII.0	7.48	0.76	5.71

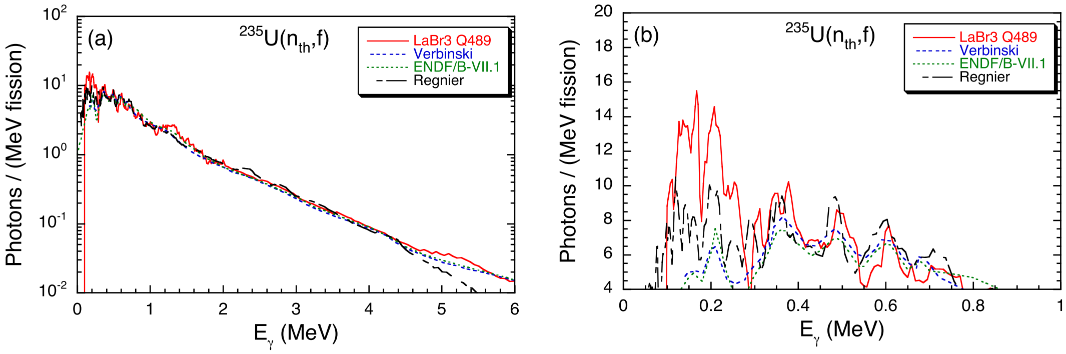

Figure 2. (a) The prompt fission γ-ray emission spectrum from this work taken with one of the 2 in. × 2 in. LaBr$_3$:Ce detectors (full red line) is shown together with experimental data from Ref. [5] (dashed blue line) and data from the evaluated library ENDF/B-VII.1 [23] (dotted green line). Recent results from model calculations [24] are shown as well (dashed-dotted black line). In the high energy range, all spectra agree very well with each other. (b) The low energy part of the spectrum corroborates this impression, since the historical data from Ref. [5] and from the recent ENDF/B-VII.1 evaluation [23] are well reproduced by our data. The best description, however, with respect to the yields at very low energies is given by the calculations [24, 26] (from Ref. [16]).

In Figure 2 the corresponding spectra are shown for the reaction ^{235}U(n$_{th}$, f). Again, we compare our experimental data with the historical data from Verbinski et al. [5], to the ENDF/B-VII.1 [23] evaluated spectrum and with recent model calculations performed by Regnier et al. [24, 26]. Again, our work confirms the historical data and also verify the observed structure in the spectrum below $E_\gamma < 0.8$ MeV. Here the evaluated data are in good agreement with the experimental ones, which indicates that experimental data were taken into consideration. Even more convincing to see is the excellent model description of our new experimental results. Even the distinct structure at $E_\gamma < 0.3$ MeV is described albeit not with the right intensity.

In Figure 3 we summarize all our results for the reaction ^{235}U(n$_{th}$, f), obtained with the different detectors. The mean value and the standard deviation of mean has been drawn from the data collected with the three most left detectors and are indicated by the full and dashed lines, respectively. Also shown are different historical data [5, 6, 22], the evaluated data from ENDF/B-VII.1 and two different model calculations [25, 26]. Looking at the total released γ-ray energy all experimental data compare quite well. Also the recent model calculations are quite successful to reproduce our experimental average photon multiplicity, mean photon energy per fission as well as the total released photon energy per fission. That the ENDF/B-VII.1 evaluated data are quite close to the experimental data indicating that the historical data had properly been taken up.

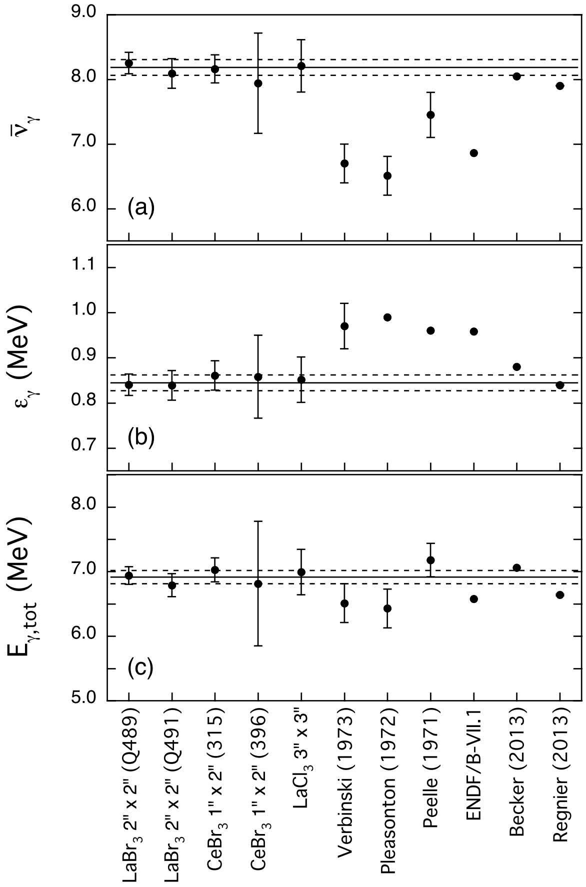

Figure 3. Overview of results for the measurement of prompt γ-ray emission for the neutron-induced fission of ^{235}U: (a) Average photon multiplicity, (b) mean photon energy per fission and (c) total released photon energy from this work are compared to experimental results from the early 1970s by Verbinski (1973) [5], Pleasonton (1972) [6], and Peelle (1971) [22], respectively, and values from ENDF/B-VII.1 [23] as well as results from recent Monte Carlo Hauser Feshbach calculations by Becker (2013) [25] and Regnier (2013) [26]. Values averaged over the results obtained with the first three detectors and their uncertainties are displayed as full drawn and dashed lines, respectively.

4. Conclusion and outlook

In this work we have presented the results from our first prompt fission γ-ray spectral measurements of the reactions ^{252}Cf(SF) and ^{235}U(n$_{th}$, f). Different lanthanide halide scintillation detectors were employed and have proven that they constitute a well-suited choice of instrumentation for this kind of investigations. The shapes of the measured spectra from two different detectors agree very well with each other and with previously published ones from the early 1970's [5, 6, 22]. PFGS characteristics were determined with high precision, which meets by far the request for an uncertainty of 7.5% at most with respect to the γ-heating in advanced nuclear reactor core simulations [1, 3, 27]. This achievement was possible due to the excellent agreement between the emission spectra obtained with each of our detectors, which allowed averaging the individual results in order to reduce the uncertainties.

We noticed that the data in the evaluated tables for ^{238}U and ^{241}Pu seems to have been obtained by simply applying a scaling factor to the evaluation of limited quality for ^{252}Cf. Since we have shown here that the evaluated data for ^{252}Cf are in conflict with existing experimental results, it might be reasonable to assume that at least some of the underestimation mentioned by Rimpault et al. [1, 3] is due to an unrealistic evaluation of prompt fission γ-ray data from ^{238}U and ^{241}Pu. In an upcoming experiment, dedicated to the measurement of PFGS from ^{241}Pu(n$_{th}$, f), we plan to use again several different lanthanide halide detectors simultaneously, like the ones reported about in this work.

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