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Prompt γ-ray spectral data from ^{252}Cf(SF), ^{235}U(n$_{th}$, f) and ^{241}Pu(n$_{th}$,f)

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Abstract

In this paper we talk about our work in developing a method to measure and analyze prompt fission γ-rays, as well as our results from the measurement of three different reactions, namely ^{252}Cf(SF), ^{235}U(n$_{th}$,f) and ^{241}Pu(n$_{th}$,f). The first two had, at the beginning of our measurements, not been examined since the early 1970s, and the last reaction had never been measured. Our results show a slight improvement over evaluated data-tables in comparison with benchmark experiments, but can still not explain all of the underestimation witnessed. Our results also show that new evaluations is needed, especially for ^{252}Cf(SF) and ^{241}Pu(n$_{th}$,f).

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Keywords: Prompt Fission γ-rays; total γ-ray energy; prompt γ-ray multiplicity; Generation IV; nuclear energy; lanthanum bromide; cerium bromide; scintillation detector; ^{252}Cf(SF); ^{235}U(n$_{th}$,f); ^{241}Pu(n$_{th}$,f);

1. Introduction

With the coming of newer and more efficient energy technology, the physics that drives this new technology has to be better understood. In this case it is about new nuclear reactor, so called Generation-IV reactors. Some of the models for these next generation reactors utilize fast instead of thermal neutrons, so there is a pressing need to understand more of the relation between energy release in fission and the incoming energy. For some parts of the energy release, this is very well known, e.g. neutron emission and fission fragments, but very little is known about the gamma emission. This is extremely important knowledge to safely model and build the shielding surrounding the new reactor cores. Therefore, OECD Nuclear Energy Agency (NEA) included gamma emission data for ^{235}U and ^{239}Pu in their High Priority Request List in 2006 (NEA, 2006). The last time these isotopes were fully investigated was in the early 1970s (Verbinski et al., 1973; Pleasonton et al., 1972; Peelle and Maienschein, 1971) but when that data is used together with benchmark experiments, they lead to an underestimation with up to 28% (Rimpault et al., 2006, 2012; Rimpault, 2006b), therefore it seems to be something lacking in the historical data.

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Table 1. Detector characteristics. This is the detectors we investigated, the one we decided to use in our array are marked with an asterisk.

Crystal	Size inch	Energy resolution ^{137}Cs	Peak efficiency ^{137}Cs	Timing resolution ^{60}Co	Intrinsic activity /cm^3/s
$LaCl_3$:Ce	1.5×1.5	4.00%	0.233(6)	352(5) ps	¿ 1.3
*$LaBr_3$:Ce	2×2	2.90%	0.340(5)	338(8) ps	¿ 0.23
*$CeBr_3$	1×2	4.40%	0.262(7)	400(7) ps	¡ 0.03
*$CeBr_3$	2×2	4.50%	—	—	¿ —
NaI	3×3	7.00%	0.15	3-5 ns	—

The DG JRC Unit for Standards for Nuclear Safety, Security and Safeguards in Geel decided to take up the challenge posted by NEA by building a detector array that in the end should be able to measure prompt fission γ-rays from any interesting fission isotope to the required precision. To succeed with this goal, we first needed to select detectors that could fulfill the task we had planned for them. The detector characteristics we need are good timing resolution, to be able to separate neutrons from γ-ray using the time of flight method, good energy resolution, to investigate the structure in the low energy part of the emission spectra as well as good efficiency, to minimize uncertainty and measurement time. With these characteristics in mind, we finally decided that the new lanthanide-halide detectors (Billnert et al., 2012; Oberstedt et al., 2013; Billnert et al., 2011; Lutter et al., 2013) would be a good compromise between the different attributes, and we bought in several kinds for further investigation. Once we had chosen the detectors most suitable for our needs, we needed to test if the setup we had in mind would actually give us good results, and therefore we started by measuring ^{252}Cf (Billnert et al., 2013). This isotope are a good start for several reasons, it has already been measured in the early 1970s, so we could immediately compare our results with something, ^{252}Cf is generally seen as the reference isotope when it comes to fission physics and last, it is spontaneous fission, so we can measure without taking up any valuable beam-time. Once we had determined that our setup is precise enough, we wanted to investigate one of the isotopes that was requested by NEA. Our choice fell on ^{235}U, so we took our detector array to the research reactor at KFKI in Budapest, Hungary to investigate the reaction ^{235}U(n$_{th}$,f) (Oberstedt et al., 2013b). This reaction was also measured in the early 1970, and therefore we could see immediately if the historical data was correct or if it needed an upgrade. After this measurement, we wanted to try something completely new, something that never been investigated before, and our choice fell on another isotope that exists in a reactor core, namely ^{241}Pu. We once again took our detector array to KFKI in Budapest, but this time we investigated the reaction ^{241}Pu(n$_{th}$,f) (Oberstedt et al., 2014).

2. Experimental setup

To be able to investigate prompt fission γ-rays, one of the major obstacles is to separate prompt γ-rays from neutrons and from neutron interactions in the surrounding material. To do this, we decided to use the time of flight method. In principal it is very easy, we just measure the time difference between the fission event and when an event is registered in our detector. Since nothing can travel faster than light, we know that the shortest time-span will be for prompt γ-rays. Because we want to utilize this method, we need detectors with good timing resolution, to be able to increase the geometrical efficiency but still separate γ-rays from neutrons. In previous experiments they used sodium-iodine (NaI) scintillation detectors, and, since our goal is to measure more precise than before, we most make sure that the detectors we choose have a better timing resolution than NaI detectors. Since we also notice some structure in the low energy range of the historical data, we wanted to be able to determine these structures more precisely in our measurements. Therefore, we need a better timing resolution than possible with a NaI detector as well. The detectors we investigated can be seen in Table 1, with their corresponding attributes for energy- and timing resolution as well as efficiency and intrinsic activity. We compare these results with that of a general NaI detector to see that our detectors are better at all three important characteristics. The detectors we finally choose are marked with an asterisk.
Now we have the the instruments to measure the emitting particles, but to be able to measure the time of flight, we also need to have a start signal. This will come from the fission event, and to measure this, we use a twin frisch-grid ionization chamber. To collect the data, we store the entire trace with the help of several digitizer cards, for the first to measurements, ^{252}Cf and ^{235}U, we used M3i digitizer cards from Spectrum (Spectrum, 2011), while in the last

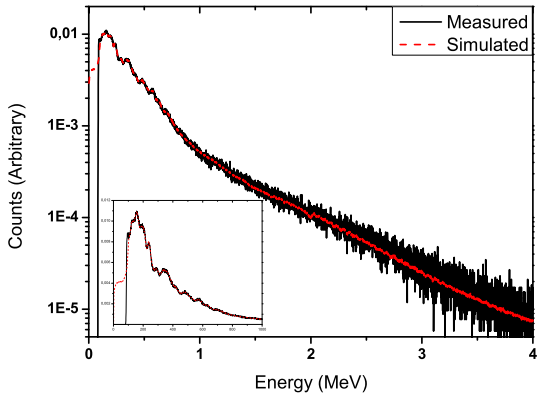

Fig. 1. Measured prompt fission γ-ray spectrum compared with the fitted simulated spectrum. The black solid line correspond to our measured prompt fission γ-ray spectrum and the dashed red line is our sum of the simulated spectra, fitted to the measured data.

measurement, ^{241}Pu we used cards from SPDevices (SPDevices, 2012). The traces were then treated using a ROOT based analysis program developed at JRC-Geel.

3. Response function

Now that we successfully measured fission γ-rays from a certain isotope, we need to determine the prompt fission γ-rays. As mentioned before, this will be done by the time of flight method, so we start by plotting our pulse-heights versus the time of flight. therefore we plot our pulse-heights versus the time of flight. Because of the excellent timing resolution of our detectors, the separation between prompt γ-rays and other interactions is very clear. Since it is the prompt γ-rays we are interested in here, we put a window over the time that corresponds to these, and take out the pulse-height spectrum. But since the random background is equally distributed over the entire spectra, we need to subtract this from our prompt part. This is done by putting a window on the background, scale it to the prompt part according to the number of time bins, and subtract the pulse-height spectrum from the prompt spectrum.
This spectrum now needs to be unfolded according to the response function of each detector. Since a photon of a certain energy makes several interactions inside the detector crystal, mainly Compton scattering and pair production, there is always a possibility that the photon only deposit a fraction of its energy before it escapes outside of the detector. This gives us the probability regarding how much energy a photon of a certain incoming energy will deposit in the detector, and this is called the response function of this energy. To determine this for the entire energy range, we use a Monte-Carlo code called PENELOPE (PENELOPE, 2011). With the help of this code, we simulate over 300 energies in the range of 100 keV up to 12 MeV, with an energy intervall determined by the detectors FWHM function. This simulation takes into account the detector geometry, the geometric efficiency as well as the different photon interactions that occurs int he crystal. The precision of this code has been verified using several calibration sources. We then introduce an individual scaling factor to each simulated energy, this scaling factor is applied the same over the entire simulated spectrum. After this we sum up all simulated energies, multiplied with the scaling factor, to get a contineous spectrum. Now we can manipulate this sum spectrum by changing the scaling factors, and to do that, we start with the highest measured energy, usually around 6-7 MeV, we set all scaling factors above this energy equal to zero, and then we choose a factor for our energy that forces our sum spectrum to the same intensity as our measured one, then we continue with the next highest energy, and do the same thing. We change all scaling factors until the sum spectrum reproduces the measured one (Fig. 1). Now we have what we need to get out the emission spectrum for this particular reaction, to determine it, we just multiply each scaling factor with the number of events we simulated to reach the spectrum, the results of this will be shown in the next chapter.

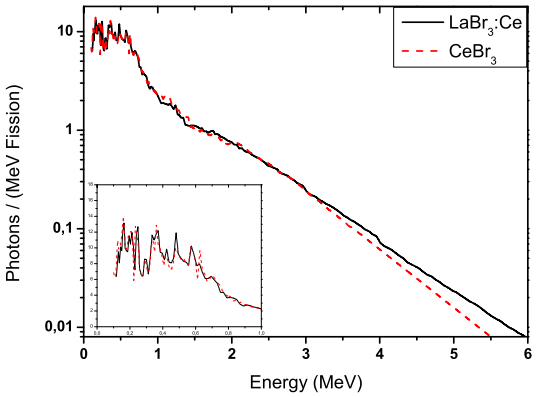

Fig. 2. Our results for both measurements of the spontaneous fission from ^{252}Cf. As can be seen, our detectors corresponds very well with each other. This gives us the confidence that our method and our detector is up to the task.

4. Results

Now that we know how to treat the data we collect, we can go on to the results. This section will be divided in three parts, one for each measurement. The structure of each segment will be more or less the same, were I first explain the difference of this measurement compared to the other two, as well as explaining where we measured and why. Then its time for the emission spectra and last some comparison with other peoples measurements.

4.1. ^{252}*Cf(SF*

First of is our first measurement, namely the measurement of prompt fission γ-rays from spontaneous fission of ^{252}Cf. This measurement was done at two separate occasions, with different detectors. To measure the fission trigger in this experiment, we did not use a twin-frisch ionization chamber like it the other measurements, but instead we used a light weight chamber, that only had a cathode for determining the fission trigger. The first measurement was done with a cerium doped Lanthanum-Bromide detector, and with analog acquisition technique. The second one we used our first Cerium-Bromide detector, as well as digital signal processing using the spectrum M3i cards.This measurement was performed to test our analysis method as well as to see if the detectors were up for the challenge. The results from both detectors can be seen in Fig. 2 and it is clear that they correspond very well with each other. That two different detectors, measured at two different times, reproduces each-others results so well gave us the confidence to claim that our method works, but to be absolutely certain, we had to compare our spectra with other measurements. This can be seen in Fig. 3, where we compare our spectra with Verbinski et al. (Verbinski et al., 1973) which was measured in 1973, the recent measurement made by Chyzh et al. (Chyzh et al., 2012) and the corresponding ENDF/B-VII.1 evaluation (ENDF, 2012). In the high energy region, we can see that all measured spectra corresponds well with each other, but there are some discrepancies in the low energy region. We measure overall a higher yield in this region than the other measurements, and we believe this comes from the fact that our detectors have a much lower threshold then both Verbinski et al. who used a NaI detector and Chyzh at al. that used an array of BaF3 detectors.

4.2. ^{235}*U(n_{th},f)*

The second experiment we preformed was to investigate the thermal neutron induced fission from the target ^{235}U. To do this we moved our setup to the research reactor at KFKI in Budapest. Since we had a greater understand of the process at this point, we decided to measure with 4 detectors at the same time, to lower our integral uncertainty. Once again all our detectors corresponded very well with each other. In Fig. 4 we can see how our results compare

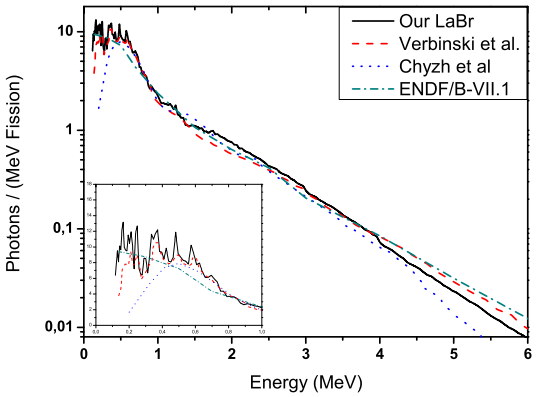

Fig. 3. ^{252}Cf(SF) result from our $LaBr_3$ detector compared with previous measurements as well as from ENDF/B-VII.1.

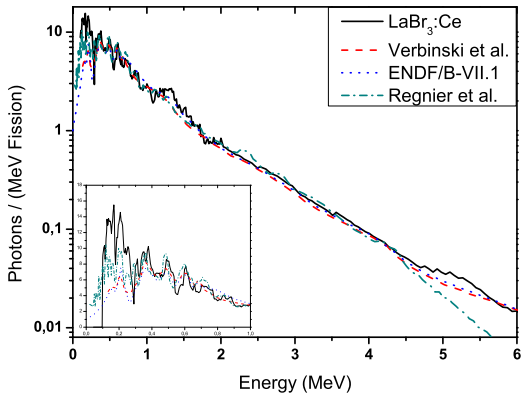

Fig. 4. ^{235}U(n_{th},f) result from our $LaBr_3$ detector compared with previous measurements as well as from ENDF/B-VII.1.

to the measurements preformed by Verbinski et al. (Verbinski et al., 1973), ENDF/B-VII.1 (ENDF, 2012) as well as a Monte Carlo simulation made by Regnier et al. (Regnier et al., 2012). In this graph we can see that the ENDF/B-VII.1 data corresponds to Verbinski et al. And both of them corresponds quite well to our measurements, which leads us to the conclusion that the underestimation mentioned in the introduction does not come from this isotope.

4.3. $^{241}Pu(n_{th},f)$

Since Verbinski et al. did such a good job measuring both ^{252}Cf as well as ^{235}U, we believe that the present data on ^{239}Pu is also valid, and therefore we need to find different reason for the underestimation. What we decided to measure was ^{241}Pu(n_{th},f), since this isotope is always present in the reactor fuel. To measure this, we once again took our setup to the research reactor in Budapest. We measured with 4 detectors, and the results from the measurements can be seen in Fig. 5. Since this isotope had never been measured before, the only data set we can compare it to is the evaluated data from ENDF/B-VII.1, and this can also be seen in Fig. 5.

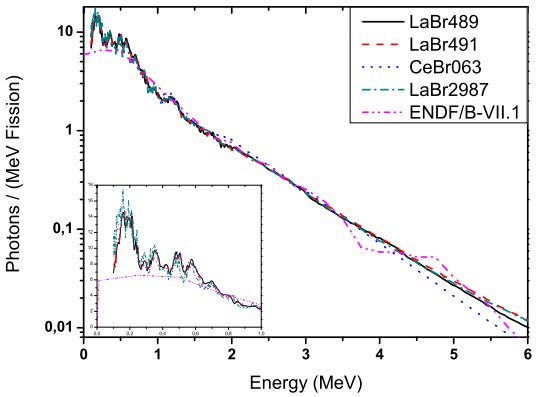

Fig. 5. ^{241}Pu(n$_{th}$,f) result from all our detectors, compared with ENDF/B-VII.1. As can be seen, our data corresponds extremely well with each other, but the evaluated data tables needs to be updated.

Table 2. Summary of our results compared to present ENDF/B-VII.1 data. The relative difference is how the evaluated data needs to change to correspond to our measurement.

Reaction	Dataset	Multiplicity /fission	γ-energy released MeV/Fission
^{252}Cf(SF)	Our measurement	8.29(6)	6.63(8)
	ENDF/B-VII.1	7.85	6.13
	Relative difference	+5.6%	+8.2%
^{235}U(n$_{th}$,f)	Our measurement	8.19(11)	6.92(9)
	ENDF/B-VII.1	6.86	6.58
	Relative difference	+19.3%	+5.2%
^{241}Pu(n$_{th}$,f)	Our measurement	8.21(9)	6.41(6)
	ENDF/B-VII.1	8.18	6.19
	Relative difference	+1.3%	+3.6%

5. Summary and Outlook

In Table 2 we can see our results for our three measurements, in regard of multiplicity and total energy, as well as a comparison with present ENDF/B-VII.1 data (ENDF, 2012). We also look at the relative difference between our data and the ENDF/B-VII.1 evaluations. As can be seen, we can not explain the underestimation on up to 28% mentioned in the introduction with our results. Therefore we have to look at other solutions, one is of course the measurement of ^{239}Pu, even though Verbinski et al. measured this together with the other isotopes. So our next plan is to take our setup to a suitable research facility to measure this isotope.

Another thing we are interested in is from which fission fragments certain γ-rays comes from. If we look at Fig. 6 we can see that a lot of the low energy peaks are at the same position, which tells us that they have to come from the same post fission isotope. Since the heavy part of the mass distribution is mainly the same for all fission isotopes, it is likely that the γ peaks come from this. A dedicated experiment to determine this is underway at IRMM right now.

Since we do not really believe that Verbinski et al. did any major mistake in the measurement of ^{239}Pu, our last explanation to understanding the underestimation comes in the form of neutrons with higher energy than thermal. Both we and Verbinski et al. measured thermal neutrons, and the ENDF/B-VII.1 data shows exactly the same spectrum for thermal and for 20 MeV inducing neutron energy. In a reactor there will of course be neutrons with higher energy produced, so we are involved in a few collaborations to determine the impact of these faster neutrons as well.

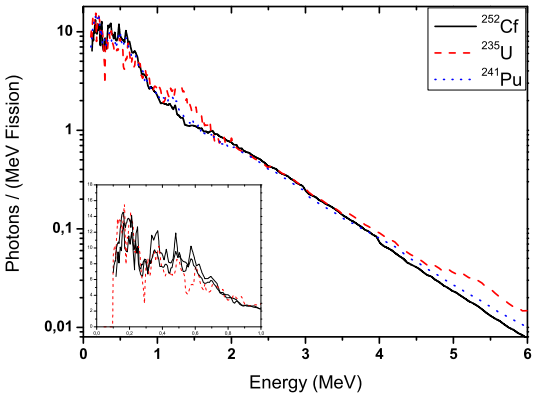

Fig. 6. Comparison between all our results. As can be seen, many of the peaks are located at the same energy. This needs to be investigated further.

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References

Billnert, R., Oberstedt, S., Andreotti, E., Hult, F.C., Marissens, G. , Oberstedt, A., Nucl. Instr. and Meth. A 647 (2011) 94.
Billnert, R. , Andreotti, E. , Hambsch, F.-J., Hult, M., Karlsson, J., Marissens, G., Oberstedt, A., Oberstedt, S., Phys. Procedia 31 (2012) 29.
Billnert, R., Hambsch, F.-J., Oberstedt, A. and Oberstedt, S., Phys. Rev C87, 024601 (2013).
Chyzh, A., Wu, C. Y., Kwan, E., Henderson, R. A., Gostic, J. M., Bredeweg, T. A., Haight, R. C., Hayes-Sterbenz, A. C., Jandel, M., ODonnell, J. M., and Ullmann, J. L., Phys. Rev. C 85, 021601 (2012).
ENDF/B-VII Evaluated Nuclear Data File, http://www.nndc.bnl.gov/exfor/endf00.jsp.
Lutter, G., Hult, M., Billnert, R., Oberstedt, A., Oberstedt, S., Andreotti, E., Marissens, G., Rosengard, U., Tzika, F., Nucl. Instr. and Meth. A 703 (2013) 158.
Nuclear Data High Priority Request List of the NEA (Req. ID: H.3, H.4), http://www.nea.fr/html/dbdata/hprl/hprlview.pl?ID=421 and http://www.nea.fr/html/dbdata/hprl/hprlview.pl?ID= 422.
Oberstedt, A., Billnert, R., Oberstedt, S., Nucl. Instr. And Meth. A 708 (2013) 7.
Oberstedt, A., Belgya, T., Billnert, R., Borcea, R., Bryś, T., Geerts, W., Göök, A., Hambsch, F.-J., Kis, Z., Martinez, T., Oberstedt, S., Szentmiklosi, L., Takacs, K. and Vidali, M., Phys. Rev. C87, 051602(R) (2013).
Oberstedt, S., Billnert, R., Belgya, T., Bryś, T., Geerts, W., Guerrero, C., Hambsch, F.-J., Kis, Z., Moens, A., Oberstedt, A., Sibbens, G., Szentmiklosi, L., Vanleeuw, D. and Vidali, M., High precision prompt spectral γ-ray data from the reaction ^{241}Pu(n$_{th}$, f), submitted for publication
Peelle, R.W. and Maienschein, F.C., Phys. Rev C3 (1971) 373.
http://www.oecd-nea.org/tools/abstract/detail/nea-1525.
Pleasonton, F., Ferguson, R.L. and Schmitt, H.W., Phys. Rev. C6 (1972) 1023.?
Regnier, D., Litaize, O. and Serot, O.,Physics Procedia, 31 (2012) 59.
Rimpault, G., Courcelle, A. and Blanchet, D., Comment to the HPRL: ID H.3 and H.4
Rimpault, G., Bernard, D., Blanchet, D., Vaglio-Gaudard, C., Ravaux, S., Santamaria, A., Phys. Procedia 31 (2012) 3.
Rimpault, G., Proc. Workshop on Nuclear Data Needs for Generation IV, April 2005 (Editor: P. Rullhusen) Antwerp, Belgium, World Scientic, ISBN 981-256-830-1 (2006) 46.
SP Device, http://spdevices.com/
Spectrum GmbH, http://spectrum-instrumentation.com/en
Verbinski, V.V., Weber, H. and Sund, R.E., Phys. Rev C7 (1973) 1173.