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Measurement of partial (n, n'γ) reaction cross-sections on highly radioactive nuclei of interest for energy production.

François Claeys^{1,2,*}, Philippe Dessagne¹, Maëlle Kerveno¹, Cyrille De Saint Jean^{3,4}, Catalin Borcea⁵, Marian Boromiza⁵, Roberto Capote⁶, Nicolas Dari Bako¹, Marc Dupuis^{3,4}, Greg Henning¹, Stéphane Hilaire^{3,4}, Alexandru Negret⁵, Gilles Noguère², Markus Nyman⁷, Adina Olacel⁵, and Arjan Plompen⁸

¹Université de Strasbourg, CNRS, IPHC/DRS UMR 7178, 23 rue du Loess 67037 Strasbourg, France

²CEA, DES, IRESNE, DER, SPRC, LEPh, Cadarache, F-13108 Saint-Paul-lez-Durance, France

³CEA, DAM, DIF, F-91297 Arpajon, France

⁴Université Paris-Saclay, CEA, Laboratoire Matière sous Conditions Extrêmes, 91680 Bruyères-Le-Châtel, France

⁵Horia Hulubei National Institute for Physics and Nuclear Engineering, 077125 Bucharest, Romania

⁶Nuclear Data Section, International Atomic Energy Agency, Wagramer Strasse, A-1400 Vienna, Austria

⁷Department of Chemistry, P.O. 55, 00014 University of Helsinki, Finland

⁸European Commission, Joint Research Centre, Retieseweg 111, B-2440 Geel, Belgium

Abstract. In the context of the development of Gen. IV nuclear reactors, the GIF (Generation IV. International Forum) has selected six innovative technologies. Among them, one can highlight the concept of breeding for $^{232}\text{Th}/^{233}\text{U}$ and $^{238}\text{U}/^{239}\text{Pu}$ fuel cycles. But those nuclei, crucial for such cycles, suffer from a lack of precise knowledge (nuclear structure, reaction cross sections). In particular, it has been demonstrated that neutron inelastic scattering reaction cross sections are not known with sufficient precision for the isotopes ^{238}U and ^{239}Pu , and not known at all experimentally for ^{233}U . In order to perform simulations of innovative reactor cores for the development of those technologies, the knowledge of the reaction cross section has to be improved which implies that new measurements have to be done. The GRAPhEME (GeRmanium array for Actinides PRecise MEasurements) experimental setup, developed by the IPHC laboratory from CNRS and installed at the EC-JRC-Geel GELINA facility is a powerful tool to answer this need [1, 2]. Combining the prompt γ-ray spectroscopy and the time-of-flight methods, it measures partial (n, n'γ) reaction cross sections. This paper reports on the improvements made on the GRAPhEME setup and data analysis methodology to tackle the challenge of (n, n'γ) cross section measurements on high activity actinides. Results obtained so far on ^{233}U are presented compared to TALYS calculations.

1 Introduction

In the context of the development of Generation IV nuclear reactors, innovative breeding $^{232}\text{Th}/^{233}\text{U}$ and $^{238}\text{U}/^{239}\text{Pu}$ fuel cycles are closely studied [3]. Having an accurate knowledge of neutron scattering on those nuclei is mandatory to meet industrial needs. In particular, the neutron inelastic scattering reaction plays a key role in the slowing down of neutrons in a nuclear reactor core. But fissile ^{233}U and ^{239}Pu nuclei suffer from a lack of precise experimental data, which explains notable discrepancies between evaluations shown for ^{233}U target in Figure 1. The increase of the cross section at low energy varies from evaluated data to another and the plateau (between 2 and 6 MeV) varies in terms of shape or amplitude. Moreover, in the case of the ^{233}U nucleus, there is no experimental data at all, neither for the total (n, n') reaction cross section, nor for partial (n, n'γ) reaction cross sections. This paper presents a methodology developed for (n, n'γ) reaction cross section determination for highly radioactive nuclei,

based on the GRAPhEME experimental setup installed at the EC-JRC-Geel GELINA facility. Section 2 presents the experimental setup. Section 3 gives main features of the undertaken data analysis methodology. First preliminary results of $^{233}\text{U}(n, n'\gamma)$ will be shown and discussed in section 4. They will be compared with theoretical predictions from the nuclear reaction code TALYS. Finally, section 5 summarizes this work and gives an outlook on the next experiment with GRAPhEME.

2 GRAPhEME at GELINA

To answer the need of precise experimental data, the DNR (Données Nucléaires pour les Réacteurs) team from IPHC developed the GRAPhEME experimental setup. It is installed at the GELINA (Geel Electron LINear Accelerator) facility, operated by the EC-JRC Geel [4]. It combines two experimental methods: the prompt γ-ray spectroscopy and the time-of-flight technique in order to produce experimental (n, n'γ) reaction cross section data [5].

*e-mail: francois.claeys@iphc.cnrs.fr

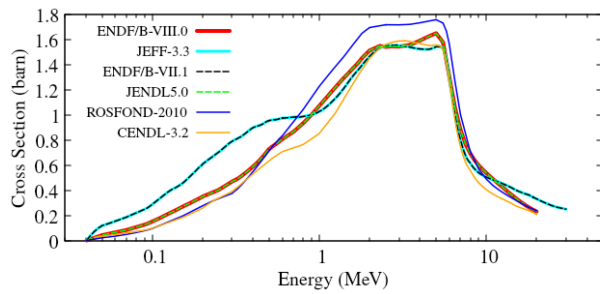


Figure 1. (color online) Comparison between evaluated nuclear data for the $^{233}\text{U}(n, n')$ total inelastic cross section.

2.1 GELINA

The GELINA facility is an electron linear accelerator producing 1 ns bunches of electrons of energies between 70 and 140 MeV at a repetition rate of 800 Hz [6]. They are sent on the primary target, a rotating disk of depleted Uranium. By the *Bremsstrahlung* effect, they emit photons that induced (γ, f) and (γ, n) reactions on nuclei of the primary target. The latter reactions produce neutrons with an energy between a few tens of keV up to 20 MeV, with a maximum neutron flux at 30 m from the primary target (where GRAPhEME is installed) of around $1.10^4 \text{ neutrons.keV}^{-1}.\text{cm}^{-2}.\text{s}^{-1}$ for a incident neutron energy of $E_n \in [0.6, 1] \text{ MeV}$.

2.2 GRAPhEME

Since the genesis of the GRAPhEME project, experimental $(n, xn\gamma)$ reaction cross sections for several nuclei have been obtained, namely $^{235}, ^{238}\text{U}$, ^{232}Th and $^{nat.182,183,184,186}\text{W}$ nuclei [7]. The experimental campaign presented here is the very first experiment for which the studied nucleus is highly radioactive. To deal with the very high counting rate of this experiment, a segmented (in 36 pixels) HPGe (High Purity Germanium) detector has been added to GRAPhEME. Each pixel, due to their small area, lowers significantly the counting rate and thus, their energy resolution is very good: $\text{FWHM}(E_\gamma = 59,54 \text{ keV}) = 0.724 \text{ keV}$ and $\text{FWHM}(E_\gamma = 1014,6 \text{ keV}) = 1.317 \text{ keV}$. These features allow a fine identification of γ transitions. Figure 2 shows a portion of radioactivity spectrum obtained from the segmented HPGe detector and a classical one. One can see the good improvement in terms of energy resolution for the segmented detector.

To measure the neutron flux, a Fission Chamber (FC) is used. It is made out of a highly enriched in ($^{235}\text{U} > 99.5\%$) UF_4 deposit of an arial density of $323.757 (1740) \mu\text{g.cm}^{-2}$. A Cu-Cd-Pb shielding protects the target from background contamination. Finally, 13 TNT2 cards (100 MHz sampling frequency) are used for data acquisition [8]. This experiment ran from 2014 to 2017, representing a total acquisition time of 4794 hours.

Incident neutrons produced by GELINA may interact with the ^{233}U target and induce nuclear reactions $((n, f), (n, \gamma)$ or (n, xn) for instance), leaving the residual nucleus

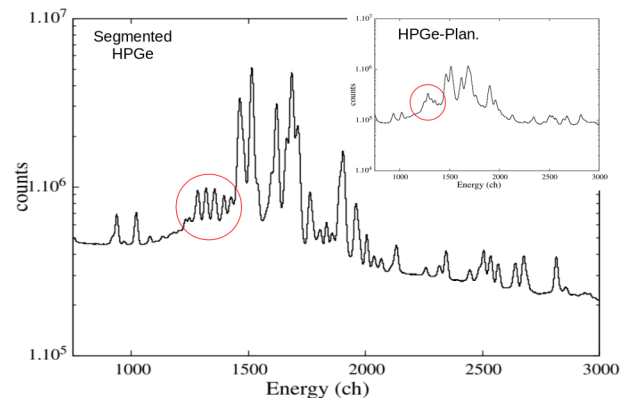


Figure 2. (color online) Radioactivity spectra ($E_\gamma \in [33, 170] \text{ keV}$) from the segmented HPGe detector and a classical HPGe detector.

in an excited state. GRAPhEME detects γ -rays that are emitted after such reactions. For each event (in the FC or HPGe detectors), the acquisition system reads and stores the detection time of γ -rays and their energy. The software *ROOT* [9] is used to read data and bi-dimensional time-energy diagrams are constructed. With time and energy projections of those diagrams, one can extract quantities of interest from time spectra and energy spectra.

3 Data analysis methodology

3.1 Angular differential cross section determination

For each detector, placed at a specific angle with respect to the neutron beam (110° and 150°), a differential angular cross section can be derived as follows:

$$\frac{d\sigma}{d\Omega}(\theta_i, E_n) = \frac{1}{4\pi} \frac{1}{N_{233\text{U}}} \frac{n_\gamma^{\text{DET}}(E_n)}{\epsilon^{\text{DET}}(1 - \tau_{\text{PU}}^{\text{DET}})} \frac{\epsilon^{\text{FC}}}{N_n(E_n)(1 - \alpha_{\text{air}})},$$

where $N_{233\text{U}}$ is the number of target nuclei, $n_\gamma^{\text{DET}}(E_n)$ the area under the γ peak of interest, $N_n(E_n)$ the number of detected neutrons, $\tau_{\text{PU}}^{\text{DET}}$ the detector's pile-up rate, ϵ^{FC} the fission chamber efficiency, ϵ^{DET} the detector efficiency and α_{air} a correction factor due to air absorption of neutrons.

$N_{233\text{U}}$ is determined by studying radioactivity γ spectra. From γ transition of ^{229}Th and using the radioactive decay law, we can obtain the number of ^{233}U nuclei in the target. This gives $N_{233\text{U}} = 1.327(60) \times 10^{21} \text{ nuclei.cm}^{-2}$, which corresponds to a mass of $m_{233\text{U}} = 7.60(35) \text{ g}$. To extract the $n_\gamma^{\text{DET}}(E_n)$, one constructs γ spectra from different energy windows in the neutron inelastic scattering energy range (*i.e.* $E_n \in [0.1; 10] \text{ MeV}$). The dedicated software *gf3* [10], especially developed for γ spectroscopy with Ge detectors is then used to obtain the quantity of interest. In the framework of this data analysis, a specific care has been taken for the extraction of the γ transitions of interest. The radioactivity component has been subtracted

from spectra and the meticulous work on peaks identification helped in the fitting procedure of remaining γ -rays of interest and from fission products.

3.2 Semi Monte Carlo method to determine integrated (n, n' γ) cross section

From the differential cross section $\frac{d\sigma}{d\Omega}$, the Gaussian quadrature method is used to determine the integrated (n, n' γ) cross section as a finite and weighted sum of differential cross sections as the following:

$$\sigma(E_n) = 2\pi \sum_{i=1}^2 \omega_i \frac{d\sigma}{d\Omega}(\theta_i, E_n),$$

where $\theta_1 = 150^\circ$, $\theta_2 = 110^\circ$, $\omega_1 = 0.69571$ and $\omega_2 = 1.30429$. Up to now, the cross section and its uncertainty were computed with a deterministic approach. However, this method implies strong assumptions. In particular, it does not take into account correlations and covariances between parameters. To meet all these issues, we developed a semi Monte-Carlo (sMC) tool, implemented for the first time in the data analysis methodology with GRAPhEME data. It was developed in *python3* along with G. Henning and its principle is the following. One realises N random draws of each parameter x of the cross section from a Gaussian distribution. For each set of parameters, a cross section is calculated and stored in a histogram. At the end of the N calculations, the obtained distribution is fitted with a Gaussian function. Its central value is thus the value of the cross section and its standard deviation the associated uncertainty. To test the validity of this method, a comparison between deterministic and semi Monte-Carlo calculations has been made. Looking at results variation between the two methods, one can see that there is a good agreement in term of cross section (less than 0.2% variation). However, some discrepancies arise in term of uncertainties. Variations on the cross section uncertainty $\Delta\sigma$ goes from 0.11% up to 27%. Especially, strong uncertainties variation are observed at low neutron energy. This can be explained by the low statistics of γ transitions near the threshold energy which implies a high dispersion of data in the sMC calculation that increase the width of the distribution [11].

With the data analysis that was described before, for the first time, $^{233}\text{U}(n, n'\gamma)$ reaction cross sections data were obtained for 12 γ transitions.

4 Results and Discussions

Table 1 sums up structure data from the ENSDF data base [12] for ^{233}U γ transitions for which a cross section has been computed.

As mentioned previously, since there is no experimental data available for this nucleus, we will only compare our results with theoretical predictions obtained with the nuclear reaction code TALYS-1.95 [13]. As a first and preliminary approach, TALYS-1.95 have been run with parameters adjustment based on *best* inputs from TALYS

data, optical model parameters from [14], level density parameters and level scheme (taking into account the first 30 levels) from the TENDL data base [16] and the pre-equilibrium prescription [15]. With this input, the total cross section (n, tot) is rather well reproduced but it is not the case for fission (n, f) and radiative capture (n, γ) ones which will have impact on the inelastic (n, n') and (n, n' γ) cross sections as shown on Figure 3. Since preliminary results are presented here, Figure 3 does not show all transitions but only some of interest for the discussion.

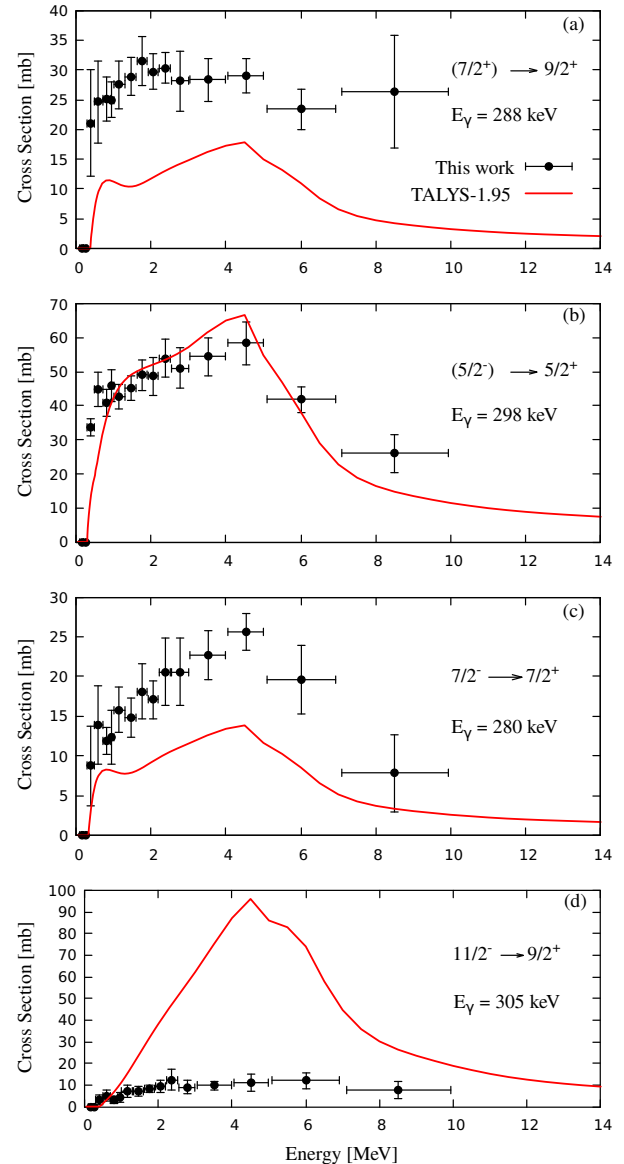


Figure 3. (color online) Experimental $^{233}\text{U}(n, n'\gamma)$ reaction cross sections (black dots) for a selection of characteristic analysed γ transitions compared to TALYS-1.95 calculations (red lines).

For the transition $(\frac{7}{2}^+) \rightarrow \frac{9}{2}^+$ (Figure 3a), One can observe an increase of the cross section at high neutron energy, which is characteristic of polluted transitions. Identification work highlighted γ transitions coming from fission products that could explain this increase of the cross section.

Table 1. Sum up of analysed ^{233}U γ transitions for which a $(n, n'\gamma)$ reaction cross section has been extracted. For each transition, the energy of the transition and initial and final state spin/parity and energy are given.

E_γ (keV)	Initial state		Final state		E_γ (keV)	Initial state		Final state	
	J^π (\hbar)	E_i (keV)	J^π (\hbar)	E_f (keV)		J^π (\hbar)	E_i (keV)	J^π (\hbar)	E_f (keV)
261.4 (2)	$(5/2^-)$	301.94 (9)	$7/2^+$	40.351 (7)	305.4 (2)	$11/2^-$	397.55 (21)	$9/2^+$	92.15 (4)
280.58 (5)	$7/2^-$	320.77 (5)	$7/2^+$	40.351 (7)	311.901 (10)	$3/2^+$	311.906 (6)	$5/2^+$	0.0
288.33 (10)	$(7/2^+)$	380.38 (8)	$9/2^+$	92.15 (4)	313.34 (20)	$9/2^-$	353.78 (12)	$7/2^+$	40.351 (7)
298.81 (2)	$(5/2^-)$	298.815 (10)	$5/2^+$	0.0	340.477 (10)	$5/2^+$	340.378 (6)	$5/2^+$	0.0
300.128 (10)	$5/2^+$	340.378 (6)	$7/2^+$	40.351 (7)	375.407 (10)	$3/2^+$	415.761 (7)	$7/2^+$	40.351 (7)
301.99 (10)	$(5/2^-)$	301.94 (9)	$5/2^+$	0.0	415.764 (10)	$3/2^+$	415.761 (7)	$5/2^+$	0.0

Beside, various discrepancies arise between experimental data and theoretical predictions in terms of $(n, n'\gamma)$ reaction cross section amplitude and shape. Let us consider three cases of particular interest. For the transition $(\frac{5}{2}^-) \rightarrow \frac{5}{2}^+$ (Figure 3b), with γ -ray energy $E_\gamma = 298$ keV, one sees a good agreement between theoretical predictions and experimental data. Aside from the rapid increase of the cross section after the threshold energy which is more pronounced in the experimental case, both shape and amplitude of the cross section are well reproduced. In the case of transition $\frac{7}{2}^- \rightarrow \frac{7}{2}^+$ (Figure 3c), with energy $E_\gamma = 280$ keV, one notices that predictions reproduce the $(n, n'\gamma)$ reaction cross section shape well, but not its amplitude which could be explained by a wrong branching ratio used in the nuclear structure file. The latter case *i.e.* transition $\frac{11}{2}^- \rightarrow \frac{9}{2}^+$ (Figure 3d), with a γ -ray energy $E_\gamma = 305$ keV, shows discrepancies in terms of both shape and amplitude of the cross section.

We can conclude that TALYS calculations are not able to reproduce experimental cross sections. This is why a better and meticulous optimization of parameters is needed to improve theoretical predictions. A first step of this optimization will be to improve the modelling of radiative capture and fission cross sections by tuning the level density parameters. A second action tool is the adjustment of deformation parameters for which fission cross section is sensitive. Once fission and radiative capture well described by TALYS, discussions about the description of $(n, n'\gamma)$ cross sections will be more relevant. This work will be, as this measurement was, a challenge because this nucleus is not well known, in particular in terms of nuclear structure and only a few experimental data are available to constrain the models.

5 Conclusion and Outlooks

The prompt γ -ray spectroscopy, combined with a time-of-flight installation is a very good method for studying $(n, n'\gamma)$ cross section. Even though it was a very challenging experimental work $^{233}\text{U}(n, n'\gamma)$ reaction cross section data were measured for the very first time. In total, 12 γ transitions have been studied. A deeper investigation of the theoretical description with TALYS is planned. To complete this study, a comparison with another nuclear reaction code, namely EMPIRE [17] is foreseen. In particular, R. Capote [18] worked on the modelling of the fission

reaction cross section for ^{233}U . It will be now interesting to compare both calculations.

With this work on ^{233}U , we have demonstrated that $(n, n'\gamma)$ cross section measurements of highly radioactive nuclei is possible with GRAPHEME. The next challenge is now the measurement of the $^{239}\text{Pu}(n, n'\gamma)$ cross section [7].

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