# (n,xn$\gamma$) cross sections on actinides versus reaction code calculations

Maëlle Kerveno[1,a], Antoine Bacquias[1], Francesca Belloni[6], Catalin Borcea[2], Roberto Capote[3], Philippe Dessagne[1], Marc Dupuis[4], Greg Henning[1], Stéphane Hilaire[4], Toshihiko Kawano[5], Nicolas Nankov[6], Alexandru Negret[2], Markus Nyman[6], Eliot Party[1], Arjan Plompen[6], Pascal Romain[4], Charoula Rouki[6], Gérard Rudolf[1], and Mihai Stanoiu[2]

[1] Université de Strasbourg, CNRS, IPHC UMR 7178, Strasbourg, France
[2] Horia Hulubei National Institute of Physics and Nuclear Engineering, Bucharest-Magurele, Romania
[3] IAEA Nuclear Data Section, Vienna, Austria
[4] CEA, DAM, DIF, Arpajon, France
[5] Los Alamos National Laboratory, Los Alamos, New Mexico, USA
[6] European Commission, Joint Research Centre, Geel, Belgium

**Abstract.** The experimental setup GRAPhEME (GeRmanium array for Actinides PrEcise MEasurements) has been used at GELINA (EC-JRC, Geel, Belgium) to perform (n,xn $\gamma$) cross sections measurements. GRAPhEME has been especially designed to take into account the specific difficulties generated by the use of actinides samples. This work takes place in the context of new nuclear data measurements for nuclear reactor applications. Considering the very tight accuracy requested for new experimental data, special care has been paid to quantify as accurately as possible all the uncertainties from the instruments and the analysis procedure. From the precise (n,xn $\gamma$) cross sections produced with GRAPhEME, the use of model calculations is required to obtain (n,xn) cross sections. Beyond the measurements, extensive work on theoretical models is necessary to achieve a better evaluation of the (n,xn) processes. In this paper, we will discuss the final step of the $^{238}$U data analysis and present some recent results obtained on $^{232}$Th compared to TALYS modellings. A new measurement campaign on $^{233}$U has started recently, a first assessment of the recorded data will be presented.

## 1. Introduction

As actinides are key components of nuclear reactors, a better knowledge of their behavior under neutron flux is required [1]. This is true both for actinides involved in present fuel ($^{235}$U, $^{238}$U, minor actinides) and for those involved in new generation cycles like the thorium cycle ($^{232}$Th, $^{233}$U). Reaction (n,xn) and especially inelastic reactions (slowing down processes), are of importance in the reactor core as the value of their cross sections impacts, for example, the radial power or key neutronic parameters like $k_{eff}$. This impact has been demonstrated in sensitivity studies (see Ref. [2] for the $^{238}$U case) and has been highlighted by the introduction of requests in the High Priority list of the Nuclear Energy Agency [3]. The target accuracy demand is very tight (few percent) and represents a real challenge from the experimental point of view. To take up this challenge our collaboration has chosen the prompt gamma-ray spectroscopy coupled to time of flight measurements to provide complete sets of (n,xn $\gamma$) cross sections on various nuclei [4]. This method allows to get round the neutron detection difficulties, but requires the use of reliable nuclear structure information and also, in many cases, theoretical models to deduce the (n,xn) cross sections from the measured (n,xn $\gamma$) ones. The GRAPhEME setup (GeRmanium array for Actinides PrEcise MEasurements) has been developed at the white neutron beam facility GELINA (GEel LINear Accelerator) operated by the European Commission - Joint Research Center at Geel in Belgium [5,6]. Up to now, four measurements campaigns have been managed on actinides. The first one was carried out with a $^{235}$U sample [7] followed by $^{232}$Th [8] and $^{238}$U [9] samples. Currently, a $^{233}$U sample is on the beam, monitored by an updated version of GRAPhEME which includes a segmented Germanium detector [10]. An overall description of the installation is given in the second section and more details can be found in Ref. [4,7]. In the third section, the status of the final analysis of the $^{238}$U is discussed and new preliminary results on thorium compared to TALYS calculations are presented. The fourth section highlights the updates of the GRAPhEME setup illustrated by first assessments of the recorded data on $^{233}$U. Conclusions and perspectives end the paper.

## 2. Experimental method

The GRAPhEME setup is located at 30 m from the GELINA neutron source (few eV to 20 MeV) which is very well adapted for (n, n′) and (n,2n) reaction studies. With four planar High Purity Germanium detectors in its initial version, GRAPhEME is able to observe gammas coming from the de-excitation of the nucleus created by the (n,xn) process. A fission chamber allows the neutron

[a] e-mail: maelle.kerveno@iphc.cnrs.fr

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flux determination [4,10]. To register events, a digital acquisition system [11] is used. The samples weight a dozen of grams and are a few tenth of millimeters thick. As accurate data are a real issue in this field a special care is taken during the analysis procedure to determine precisely all the parameters involved in the cross section formula (events numbers, detection efficiencies, atoms number in the samples, pile up and dead time corrections, neutron attenuation in the air).

## 3. Preliminary results on $^{238}$U and $^{232}$Th

Several measurements campaigns, which represent almost 2700 h of beam time, were performed between 2010 and 2013 with a $^{nat}$U sample. Preliminary results coming from a set of collected data in 2012 (1200 h), have been obtained for 36 (n,n$'\gamma$) transitions. As these data have been presented at several occasions [9,12,13], the present section will focus on the last work dedicated to the finalization of the analysis in view of the final publication. First, an extensive study of the radioactivity spectra obtained during all the measurements campaigns allowed us to determine the number of $^{238}$U atoms in the sample with an uncertainty which won't exceed 3%. Another issue concerns the observed transitions which show a behavior suggesting a contamination by other reaction channels (fission product, capture, n,2-3n). A special treatment is thus required to extract the exact cross section. This procedure includes constructing an optimized set of data with very stable gain and $\gamma$-energy resolution. Indeed a slight drift of the gain or a degradation of the resolution are always possible in such long measurements. Then a complex adjustment of the $\gamma$-energy distribution is performed. This special treatment is needed for about a third of the analyzed $\gamma$-transitions. The low energy transitions (lower than 120 keV) require an additional correction for the non linearity of the energy-time response of the electronics in this low amplitude signals region (the walk effect). Another important part of the work concerns the theoretical modelling of the cross sections. This is done, in close collaboration, by our theoretician colleagues of CEA-DAM, IAEA and LANL in the frame of the French national program NEEDS (Nucléaire, Energie, Environnement, Déchets, Société) driven by CNRS. A new evaluation of the neutron inelastic cross section on $^{238}$U (IAEA, CIELO b46) [14–16] has been produced but no optimization of the exclusive (n,n$'\gamma$) channels was yet included. For this optimization, we have seen first the importance of a good knowledge of branching ratios, which is not so obvious for $^{238}$U. Beyond this observation, several advances have been achieved with the use of microscopic calculations (QRPA) [17,18]. For example, for the transitions in the rotational band, the microscopic description of the E1 and M1 strength functions and of pre-equilibrium cross sections has notably increased the predictive power of TALYS-1.82 [19]. It has been also shown that the number of discrete levels considered in the calculation and the coupling from continuum to discrete levels have a non-negligible impact on the inter-band transition cross sections and thus have to be studied in depth in the case of $^{238}$U. All this work will be detailed in a future publication.

Thorium is the fertile nucleus of the new fuel cycle generation Th/U. This fuel is an alternative to uranium as it can be used in both current or future breeder systems, it creates less minor actinides than the uranium fuel, it is abundant in the earth [20], it is proliferation-resistant... The state of nuclear data relative to this cycle is not at the level of the one relative to the uranium cycle. New data are thus required to study in an efficient way all the possible options. The inelastic process on $^{232}$Th and $^{233}$U is poorly known especially for $^{233}$U where no experimental data exist. Figure 1 illustrates the status of experimental data base for $^{232}$Th(n,xn) reactions and exhibits the small number of experiments on neutron inelastic process on thorium. In our experiment, the thorium sample (m ≈ 12.6 g, thickness ≈ 0.3 mm) was irradiated at GELINA during several months and a data set of 800 hours of beam time could be analyzed. In a first step analysis, 46 $\gamma$-ray transitions in $^{232}$Th have been identified in the $\gamma$ spectrum but also 12 in $^{231}$Th and 4 in $^{230}$Th.

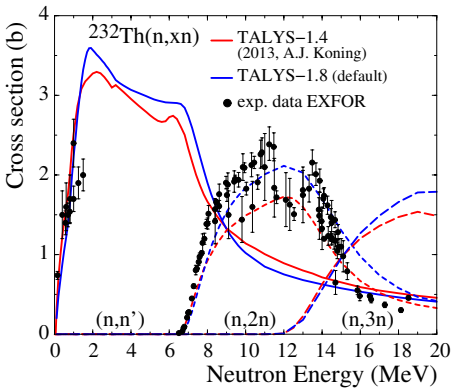


**Figure 1.** Inelastic, (n,2n), (n,3n) experimental reaction cross sections on thorium compared to TALYS calculations.

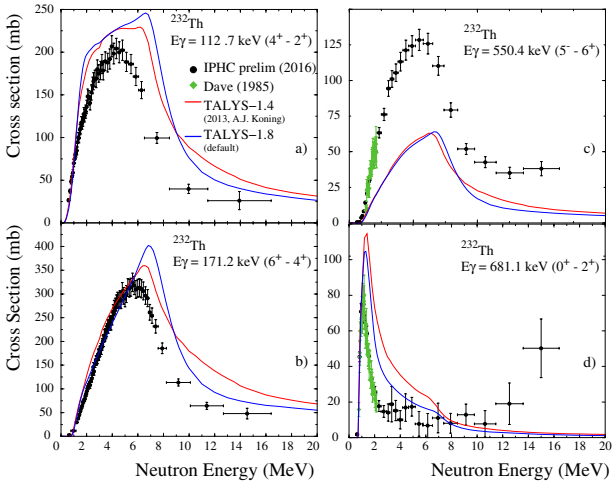


**Figure 2.** Preliminary $\gamma$-transition cross sections in $^{232}$Th compared with experimental data [21] and TALYS calculations.

Preliminary cross sections for four $\gamma$-transitions (2 in the rotational band (a, b) and 2 intra-band transitions (c, d)) are presented in Fig. 2 compared to the only previous experimental data from Dave et al. [21] and with two versions of TALYS calculations. One notices that the agreement of our data with the Dave et al. ones is excellent.

Concerning the modelling, a first calculation was performed by A.J. Koning in 2013 with optimized input

parameters for the version 1.4 of TALYS, in the frame on a work on $^{232}$Th(n,5n $\gamma$) cross sections measurements [22]. The second calculation is a default input parameters calculation with TALYS-1.8 performed by the authors. A general comment is that the predicted (n,n$'\gamma$) cross sections are not very different between the two versions of TALYS. One finds the same behavior for experimental cross sections versus model predictions as the one found for $^{235}$U or $^{238}$U (TALYS over estimation of the high spin $\gamma$ transitions in rotational band and over and under estimations in inter-band transitions). Nevertheless one can remark that the maximum of the TALYS cross sections is shifted to high neutron energy. This shift is may be partly due to the too slow rise of the (n,2n) cross section as shown in Fig. 1. Consequently, with the large set of new data we will provide on thorium, the same theoretical work done for $^{238}$U should be done on this nucleus with expected improvements on the modelling.

## 4. Perspectives on $^{233}$U

Another key nucleus in the thorium fuel cycle is obviously the fissile $^{233}$U for which no experimental data exist for the inelastic process and only one spectrum-averaged measurement [23] at about 1.5 MeV for the (n,2n) reaction. This is nevertheless not surprising taking into account, first the difficulty to obtain a usable $^{233}$U sample and second, the activity of this nucleus. Our collaboration managed to solve the first difficulty and obtained a $^{233}$U sample (m $\approx$ 8.3 g, diameter $\approx$ 30 mm, $\sim$0.64 mm thick with an activity of $\sim$3 GBq) prepared by the target laboratory of EC-JRC, Geel. To solve the second point (high radioactivity) but also to reduce the impact of the $\gamma$-flash, our collaboration added a segmented germanium detector [10] to GRAPhEME in 2014. The Ge crystal of the detector is 54 mm by 54 mm, 20 mm thick segmented into 6 by 6 grid with squared pixels 6.66 mm wide. Its efficiency has been precisely characterized with both source measurements and GEANT4 simulations. With this kind of segmented detector one issue is the good determination of the electric cross talk between pixels and the $\gamma$-ray scattering from one pixel to its neighbors. The analysis procedure is currently being developed to take into account these effects and ensure that the $\gamma$-energy deposit recorded by a pixel is not perturbed. After a first measurement of about 700 h in 2014–2015, a second campaign is presently running since the beginning of 2016 (after the renovation of the GELINA flight paths) adding-up to a total of 1500 h. Preliminary analysis shown that in a pixel, the pile-up is fifteen times less than in a single crystal as shown in Fig. 3.

The $\gamma$-energy resolution in a pixel is very good and better to the one of the best planar crystal of GRAPhEME. Figure 4 compares a portion of the $\gamma$-energy spectra (867 h of beam time) for a planar Germanium and for a pixel of the segmented one, with time windows corresponding to radioactivity and (n,n$'$) energy region. Some peaks due to $\gamma$-transition in $^{233}$U ((n,n$'$) reactions) are visible but the intense radioactivity makes the background very dense therefore much more beam time is required to be able to extract cross sections.

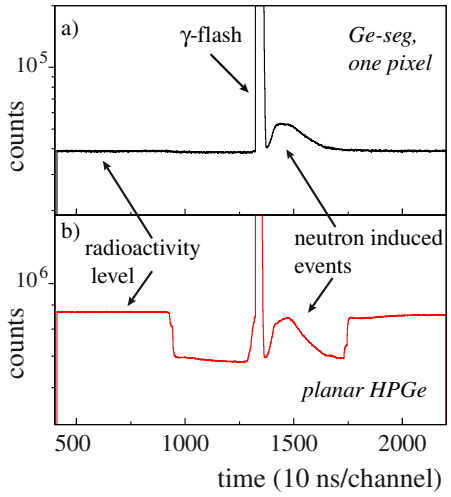


**Figure 3.** Time spectra for a single HPGe crystal b) and in a pixel of the segmented HPGe a). The hollow before and after the $\gamma$-flash peak, clearly visible in b), is due to pile-up events on $\gamma$-flash. Without the beam, this rate is 32% in the single HPGe and only 2% in one pixel of the segmented detector, with the beam these rates come to 36.7% and 2.5% respectively.

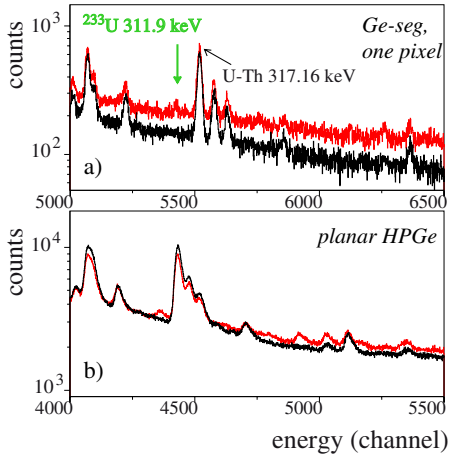


**Figure 4.** Portion of the $\gamma$-energy spectra for one pixel of the segmented detector a) and in a single HPGe b). The black spectra are the radioactivity ones and the reds correspond to a time window in the (n,n$'$) energy region.

## 5. Conclusion

The GRAPhEME setup is used at the GELINA facility to measure accurate (n,xn $\gamma$) cross sections on actinides. Consolidated data on $^{238}$U will be available in the coming months and have been used to test and improve predictive power of theoretical codes allowing a better knowledge of the neutron inelastic scattering process. The same work with data collected on a thorium sample is under progress. Currently, with an updated version of the GRAPhEME setup, a measurement campaign on $^{233}$U is running aiming to provide first measurement of (n,n$'$ $\gamma$) cross sections.

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