EPJ Web of Conferences **42**, 01005 (2013)
DOI: 10.1051/epjconf/20134201005

(n,xn γ) reaction cross section measurements for (n,xn) reaction studies

Maëlle Kerveno[1,a], Antoine Bacquias[1], Catalin Borcea[2], Philippe Dessagne[1], Jean-Claude Drohé[3], Nikolay Nankov[3], Markus Nyman[3], Alexandru Negret[2], Arjan Plompen[3], Chariklia Rouki[3], Gérard Rudolf[1], Mihai Stanoiu[2], and Jean-Claude Thiry[1]

[1] CNRS, Université de Strasbourg, UMR7178, IPHC, 23 rue du Loess 67037 Strasbourg, France
[2] Horia Hulubei National Institute of Physics and Nuclear Engineering , Str. Reactorului 30, 077125, Bucharest Magurele, Romania
[3] European Commission, Joint Research Centre, Institute for Reference Materials and Measurements, Retieseweg 111, B-2440 Geel, Belgium

Abstract. In the context of improvement of nuclear data bases for future nuclear reactor researches, we study (n,xn) reactions experimentally with the help of the (n,xn γ) technique. The experiments are performed at the GELINA facility which delivers a pulsed, white neutron beam. Several measurement campaigns have been performed on ^{235}U, ^{232}Th, ^{182,183,184,186}W and ^{238}U isotopes. A compilation of all these experimental cross sections (mostly still preliminary) compared to theoretical predictions will be discussed.

1 Introduction

Precise knowledge of (n,xn γ) reactions is a key issue in present day reactor development studies. Indeed the new Generation IV nuclear reactors explore new energy domains, and imply reaction rates unknown or badly known at this stage. For the design of these new systems, the (n,xn) reactions have to be well described by simulation codes as they are an important energy loss mechanism and as they lead to neutron multiplication and production of radioactive isotopes. The case of the inelastic scattering on ^{238}U is an interesting illustration as, in the EXFOR data base, we find around forty references for the ^{238}U(n,n') reaction cross section but only eight concerning the total cross section. The experimental data cover mainly only the low neutron energy part and discrepancies exist between the data but also between evaluations. Finally the uncertainty on the ^{238}U(n,n') cross section is close to 20%. Following the studies made by Salvatores et al. [1] and A.Santamarina et al. [2] this uncertainty impacts too strongly the accuracy of different core parameter calculations like the k_{eff}, the radial power distribution and also the β_{eff} calculations of pressurized water and fast reactors. So the recommendations for ^{238}U(n,n') cross section uncertainty is to obtain 10% for PWR studies and 5% for SFR studies. Among the three possible experimental methods to study (n,xn) reactions, our collaboration has chosen to use the prompt γ-ray spectroscopy which provides (n,xn γ) cross sections. The presented work is thus performed using this technique coupled to time of flight measurements, for which a high precision experimental setup called GRAPhEME, was designed. It has already been used to measure (n,xn γ) reactions on isotopes such as ^{235}U, ^{238}U, ^{232}Th and ^{182,183,184,186}W. This paper is a review of preliminary results obtained during the different GRAPhEME measurement campaigns compared to TALYS-1.2 calculations [4]. We also discuss the validity of the (n,xn γ) technique for (n,xn) reactions studies.

---

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

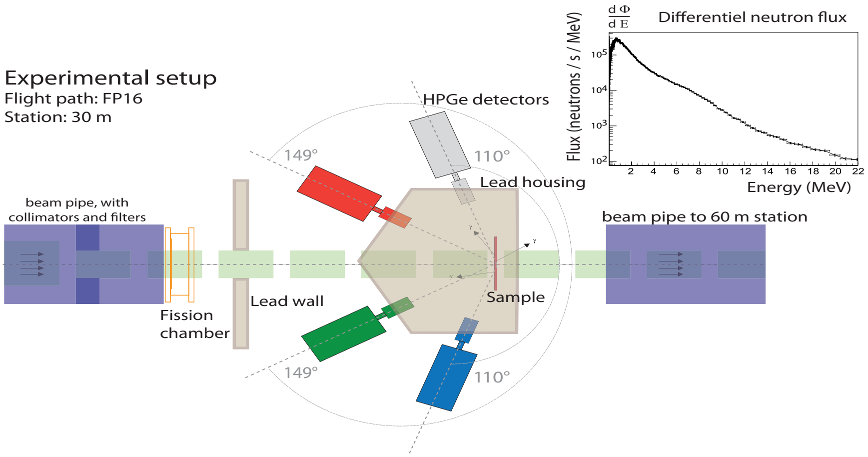

Fig. 1. Experimental setup GRAPhEME and differential neutron flux measured at FP16/30m at GELINA.

2 Experimental set-up and data analysis

The (n,xn γ) technique consists in detecting the γ radiations from the decay of the excited nucleus created by the (n,xn) reaction using Planar High-Purity germanium (HPGe) detectors. Thus we can determine the γ production cross section.

The GRAPhEME set-up (GeRmanium array for Actinides PrEcise MEasurements) has been developped and is installed at GELINA, a facility at IRMM, Belgium [5]. GELINA produces a white pulsed neutron beam characterized by an incident flux spectrum from a few keV up to several MeV [6] (see Fig. 1). This experimental set-up shown in Fig. 1, is described in detail in [3, 7–9].

For a given γ-transition, averaged cross-sections were derived from the time-of-flight spectra for neutron energy bins of suitable sizes. The obtained cross sections are not corrected for internal conversion. Our efforts were focused on the uncertainty minimization of all parameters. Up to now we can reach cross section uncertainties from 5 to 7% for neutron energies up to 9 MeV and 20% in the high neutron energy range.

3 Results

3.1 ^{235}U measurement - preliminary results

The first results presented concern the ^{235}U measurement campaigns [3] (Fig. 2). Cross sections have been obtained after 1466 hours of beam and with a 37.43 g sample. For this case, we were able to determine one (n,n' γ) and three (n,2n γ) transitions cross sections. Concerning the (n,n') measurement, the low statistics for each γ-transition coupled to possible peak contamination (fission product decay for example) explain the difficulty to obtain the cross section for more than one transition. TALYS-1.2 calculations have been performed by P. Romain from CEA Bruyères-le-Châtel. Three calculations test the sensitivity of the prediction to the nuclear model inputs. Two optical model potentials have been used: a specific global one, the CGMR (Coupled Global Morillon Romain), and the Soukhovitkii one, coupled to finely tuned phenomenological inputs or to microscopic inputs (cf. [3] and references inside). In the data base only two (n,xn γ) cross section references are available [11, 10]. For the (n,n' γ) transition, our data agree with those of Younes *et al.* at high neutron energies but the three TALYS-1.2 calculations under-estimate the cross section especially for neutron energy above 1 MeV. For the (n,2n γ) channel, our maximum cross section is lower than that of Younes *et al.* except for the 244 keV γ-transition where the agreement is good. The TALYS-1.2 calculations all overestimate the cross sections, nevertheless the pheno-CGMR gives the best prediction.

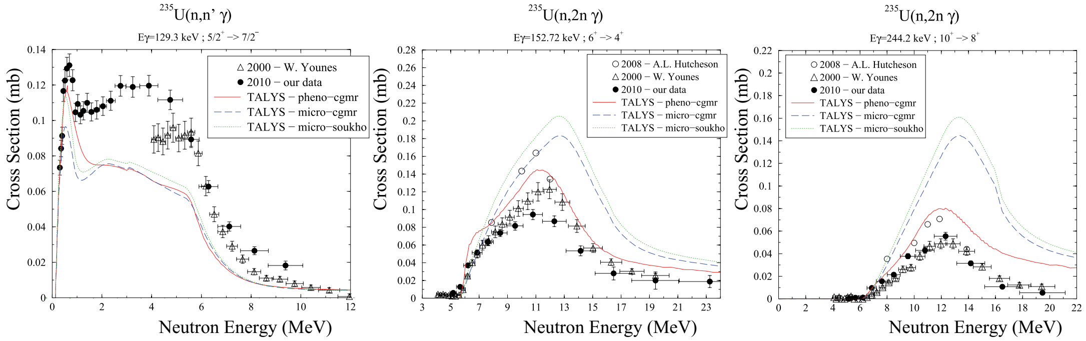

Fig. 2. Total γ-production cross sections due to the ^{235}U(n,n') reaction for the 129.3 keV transition (left) and due to ^{235}U(n,2n) reaction for the 152.7 keV transition (middle) and the 244.2 keV transition (right). The results are compared to TALYS-1.2 predictions and existing experimental data.

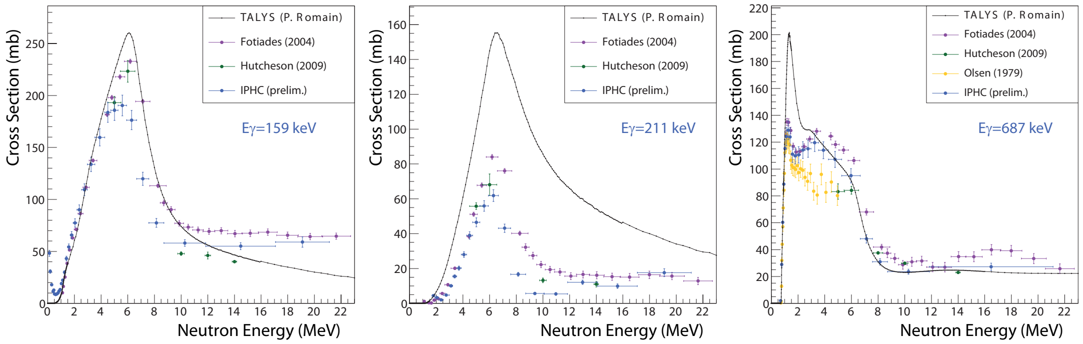

Fig. 3. Total γ-production cross sections due to the ^{238}U(n,n') reaction for the 159 keV transition (left) and the 211 keV transition (middle) in the ground state band and for the 687 keV transition (right). The results are compared to TALYS-1.2 predictions and existing experimental data.

3.2 ^{238}U measurement - preliminary results

For this measurement, data were accumulated during 1200 hours with a sample of 10 g. Unlike the case of ^{235}U we are able to identify about 30 γ-transitions and we have determined 20 preliminary cross sections. It has to be noticed that we are able to extract also cross sections for the transition from the first inelastic level at 45 keV to the ground state [12] but with limited statistical and systematical accuracy due to the high conversion coefficient and sample sel attenuation for this γ transition. Our experimental data are compared to TALYS-1.2 calculations made by P. Romain (CEA). In EXFOR data base, four (n,n' γ) measurements are available [14, 13, 15, 16].

Fig. 3 shows three examples of obtained γ-transitions cross sections. The agreement with Fotiades *et al.* is roughly good even if our values are a bit lower. The shape is well reproduced by TALYS-1.2 except in some cases where a direct component appears, like for the 687 keV γ-transition where this component is overestimated by the code. Quantitatively agreement varies with the γ-transition.

3.3 ^{232}Th measurement - preliminary results

For the thorium measurement campaign, the 12 g sample was in the beam during 375 hours and we are able to observe many γ-transitions and especially the deexcitation of the first level at 49 keV. A lot of measurements exist in the EXFOR data base and particularly for the (n,2n) reaction which can be measured by the activation technique. Nevertheless only one (n,n' γ) reference from Dave et al. [17] exists and for which the measurement has been performed for neutron energy up to 2 MeV. For

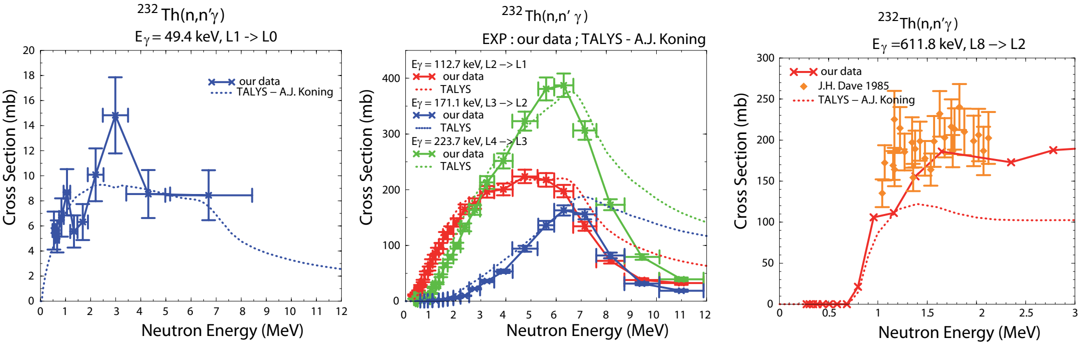

Fig. 4. Total γ-production cross sections due to the ^{232}Th(n,n') reaction for the four γ-transitions in the ground state band (left and middle) and for the 611.8 keV transition (right.) The results are compared to TALYS-1.2 predictions and existing experimental data.

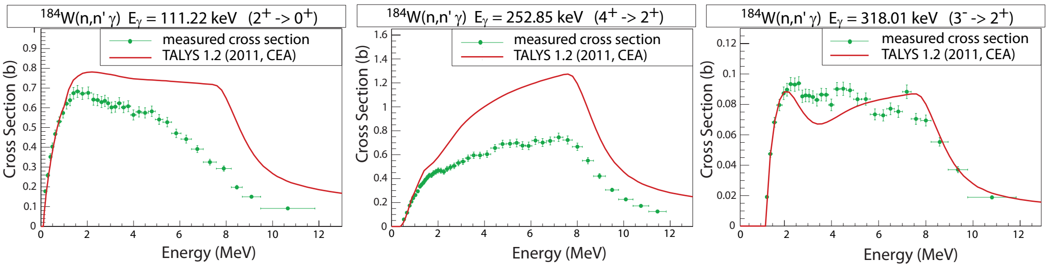

Fig. 5. Total γ-production cross sections due to the ^{184}W(n,n') reaction for the 111 keV transition (left) and the 252.85 keV transition (middle) in the ground state band and for the 318 keV transition (right). The results are compared to TALYS-1.2 predictions

thorium, A. Koning from NRG, Petten performed the TALYS-1.2 calculations. A selection of the very preliminary results of (n,n' γ) cross sections is shown in Fig. 4. Our measured cross sections are in agreement with those of Dave *et al.*. The cross section's shapes and amplitudes are well described by TALYS-1.2, but only up to 7 MeV and for γ-transitions in the ground state band.

3.4 ^{184}W measurement - preliminary results

Measurements on four tungsten isotopes (^{182,183,184,186}W) have been performed during 300 or 500 hours, depending on the sample. A lot of γ-transitions can be analysed and compared to TALYS-1.2 predictions made by P. Romain (CEA). In the EXFOR data base, only few (n,xn) cross section measurements exist but nothing concerning (n,xn γ) cross sections.

Fig. 5 shows three (n,n' γ) cross sections on ^{184}W. The cross section's shapes are well reproduced by TALYS-1.2 but discrepancies on amplitude, sometimes large, exist and depend on the γ-transition. The situation is roughly the same for the four measured isotopes. One thing brought out by this preliminary analysis is the crucial role of branching ratio description as significant differences appear on the TALYS-1.2 cross sections depending on the data bases used.

4 Discussion

From the (n,xn γ) measurements the total (n,xn) cross section can be deduced since the total inelastic cross section is the sum of the cross section carried by all transitions that directly decay to the ground state. Unfortunately for this kind of experiment, all these direct ground state feeding can not be observed. Nevertheless the total cross section can be obtained by considering other transitions if their branching ratios are known. In practice, the deduced (n,xn) cross section is thus frequently only a lower limit of the total cross section and spectroscopic parameters and theoretical model predictions should be used to determine the total (n,xn) cross section. To avoid a strong model dependence of the method, experimentalists should thus provide accurate measurements with well known and quantified uncertainty sources for a maximum of γ-transitions. Efforts should also be done to measure those feeding the ground state and especially from the first excited level. From the theoreticians and evaluators point of view, for fissionable nuclei, a good description of the fission process is a important prior knowledge for a good (n,xn) calculation [9]. As explained before, a good knowledge of spectroscopic parameters is also very important. And finally improvement of models which describe the pre equilibrium process has to be considered.

5 Conclusion

With the GRAPhEME set-up we provide (n,xn γ) cross section as accurate as possible. Several analyses are still in progress and the covariance matrix determination of our data sets will also be conducted. All this work is performed with the collaboration of our theoretician and evaluator colleagues. In the near future, this kind of measurement on a more active target, ^{233}U, will be carried out. The knowledge of the ^{233}U(n,2n) cross section is indeed very important for the thorium cycle study. For this purpose, a segmented (with 36 pixels) Germanium detector will be added to the GRAPhEME set-up. At last, our collaboration will also focus its efforts on the measurement of branching ratios and of highly converted level de-excitation.

References

1. M. Salvatores and R. Jacqmin, *Uncertainty and target accuracy assessment for innovative systems using recent covariance data evaluations* (Nuclear Science-NEA/WPEC-26, Volume 26, NEA N^o.6410, 2008)
2. A. Santamarina *et al.*, private communication
3. M.Kerveno *et al.*, submitted soon
4. A. J. Koning, S. Hilaire, M.C. Duijvestijn, *TALYS-1.0, Proceedings of the International Conference on Nuclear Data for Science and Technology - ND2007* (EDP Sciences, Paris, France, 2008) 211-214
5. W. Mondelaers and P. Schillebeeckx, Notiziario neutroni e luci di sincrotrone **11**, (2006) 19-25
6. D. Ene *et al.*, Nucl. Instrum. Methods Phys. Res. A **618**, (2010) 54-68
7. J. C. Thiry *et al.*, Journal of the Korean Physical Society, **59**, issue 2, (2010) 1880-1883
8. J. C. Thiry *et al.*, NEMEA-6 workshop, online proceedings : http://www.oecd-nea.org/science/wpec/nemea6/ (2010)
9. M. Kerveno *et al.*, NEMEA-6 workshop, online proceedings : http://www.oecd-nea.org/science/wpec/nemea6/ (2010)
10. A. Hutcheson, PhD thesis : *Neutron-Induced Partial Gamma-Ray Cross-Section Measurements on Uranium*, (Department of Physics in the Graduate School of Duke University, 2008)
11. W. Younes *et al.*, *The ^{235}U(n,2n γ) Yrast Partial Gamma-Ray Cross Sections: A Report on the 1998 - 1999 GEANIE Data and Analysis Techniques*, (U.S. Department of Energy, Laurence Livermore National laboratory,UCRL-ID-140313, 2000)
12. A. Bacquias *et al.*, *Study of (n,xn γ) reactions on 235,238U* , 13th International Conference on Nuclear Reaction Mechanisms, Varenna (2012).(to be published)
13. N. Fotiades *et al.*, Phys. Rev. C **69**, (2004), 24601
14. A. Hutcheson *et al.*, Phys. Rev. C **80**, (2009), 14603
15. F. Voss, *γ-ray production cross sections in inelastic neutron scattering on ^{238}U*, (Kernforschungszentrum Karlsruhe Reports, KFK2379, 1976)
16. D. K. Olsen *et al.*, Measurement of ^{238}U(n,n'γ) cross sections, (International Conference on Nuclear Cross Sections for Technology, 1979), 677
17. J. H. Dave *et al.*, Nuclear Science and Engineering **91**, (1985), 187