Nuclear Measurements, Evaluations and Applications – NEMEA-6

Workshop Proceedings

25-28 October 2010
Krakow, Poland

Co-organisers

Joint Research Centre
The Henryk Niewodniczanski Institute of Nuclear Physics
Institute for Reference Materials and Measurements

NUCLEAR ENERGY AGENCY
Organisation for Economic Co-operation and Development

Measurement of (n,xnγ) reactions of interest for new nuclear reactors

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Abstract

Our presented research is focused on cross-section measurements of (n,xnγ) reactions in the framework of Generation IV nuclear reactors studies. Indeed the development of new fast reactors or the investigations concerning new fuel cycles require the improvement of nuclear databases over a wide range of energies, nuclei and reactions. One of the challenges of new measurements, in this field, is the accuracy level that they can reach and which is required by the Nuclear Data High Priority List produced by the NEA.

Our collaboration has developed an experimental set up based on the prompt gamma ray spectroscopy method, using the GELINA facility of IRMM at Geel (Belgium), which produces a pulsed, white neutron beam. The results concerning ^{232}Th(n,xn γ) and ^{235}U(n,xn γ) reactions cross-sections measurement, in the fast neutron energy domain (up to 20 MeV), are presented and compared with existing experimental data but also with theoretical TALYS calculations.

All these investigations are performed in the framework of ANDES program (7th framework program, EURATOM).

Introduction

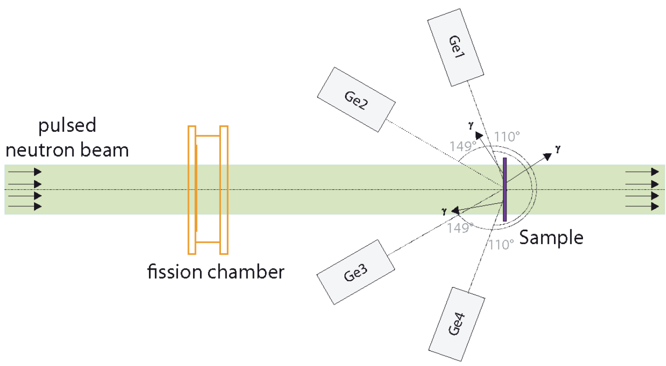
Precise knowledge of (n,xn γ) reactions is a key issue in present day's reactor development studies. Indeed the new Generation IV nuclear reactors explore new energy domains, and imply reaction rates unknown or badly known at this stage. For the design of these new systems, the (n,xn) reactions have 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. From a theoretical point of view, in the case of fissionable targets, the prediction of (n,xn) reactions implies a good knowledge of fission parameters as a strong competition exists between neutron emission and fission in the studied nuclei. Moreover (n,xn γ) reactions allow to test, validate or improve theoretical codes, such as TALYS[1] (used in this work) by providing information and constraints on a wide range of nuclear structure parameters such as branching ratio, level densities, spin distributions…

The presented work is performed using the (n,xn γ) technique, already used for stable isotopes as ^{208}Pb [2], for which a high precision experimental setup was designed. It has already been used to measure (n,xn γ) reactions on isotopes such as ^{235}U, ^{232}Th and ^{nat}W. In this paper, results on ^{235}U(n,xn γ) for x=1,2, and on ^{232}Th(n,xn γ) for x=1,2,3 are presented and compared to TALYS[1] calculations.

Experimental setup

This section treats the applied measurement techniques as well as the experimental setup, shown in Figure 1.

Figure 1: Sketch of the experimental setup used at GELINA, FP16/30m



The (n,xnγ) technique

This method consists in detecting the γ radiation from the decay of the excited nucleus created by the (n,xn) reaction using High-Purity germanium (HPGe) detectors. They yield the level population cross-section of the nucleus in a given excited state.

The TOF technique

The experiment is realized at GELINA, a facility at IRMM, Belgium [3,4]. GELINA produces a white, pulsed neutron beam using the (γ,xn) and (γ,F) reactions on a depleted uranium target which leads to an incident flux spectrum from a few keV up to several MeV.

The pulsed beam enables energy separation of the incident neutrons using a time spectrum, which can be calibrated thanks to the presence of a γ-flash. The experimental setup is located 30 m away from the neutron source. The data acquisition resolution being 10 ns, this flight path is the best compromise between time resolution and flux intensity, allowing a resolution of 1 MeV at neutron energy of 20 MeV.

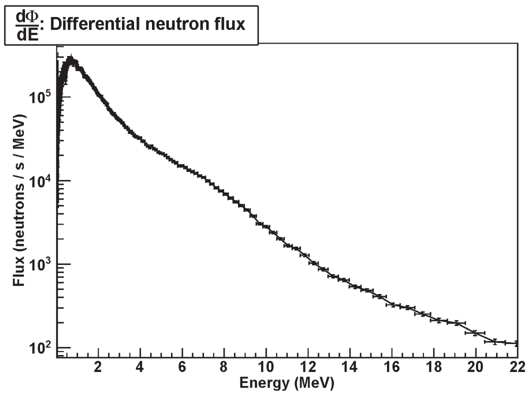
Data acquisition

The signals arising from the detectors are processed by TNT2[1] cards developed at IPHC. Signals are processed online in parallel in two different channels, one determining the event time by applying the Constant Fraction Discriminator (CFD) method and one calculating the γ-ray energy of the incident events using the Jordanov-knoll [5,6] signal treatment method. The events are stored in list mode files, where the energy is encoded on 14 bits and the time resolution is 10 ns.

Flux monitoring

Precision of cross-section measurements depends very strongly on the uncertainties of the incident neutron flux which is illustrated in Figure 2, thus it is of upmost importance to have very precise flux determination.

Figure 2: Differential neutron flux measured at FP16/30m at GELINA



The flux is measured using a ^{235}U fission chamber. The vacuum evaporated $^{235}UF_4$ deposit is highly enriched in ^{235}U (>99.5%) and very thin (324 μg/cm^2 of ^{235}U). The effective thickness of the fission chamber was chosen between 6 and 7 mm, as this leads to the best ratio of fission fragment energy loss (signal) and radioactivity α particle energy loss (background noise).

After extensive studies for the optimization of the fission chamber configuration and a high precision calibrating measurement performed at the Physikalisch-Technische Bundesanstalt (PTB), Braunschweig, Germany, uncertainties of 2.1% on the efficiency of the detector have been reached. See the paper of J.-C. Thiry elsewhere in these proceedings for more details.

Gamma-ray detection

The γ-rays, emitted from the produced isotopes, are detected using four high purity germanium (HPGe) counters, made of planar crystals with depths ranging from 2 to 3 cm and surfaces dimensioned between 10 and 28 cm^2. The detectors are optimized for high resolution detection at low energies (resolution of 0.7 keV at 122 keV). They are placed at angles of 110° and 149° which allows the angular dependence to be taken into account. Backward angles were chosen to reduce dead time caused by the observation of events due to γ-flash scattering.

1. TNT : Treatment for NTof.

Data analysis

Differential cross-sections

The differential production cross-section for a γ transition of interest at a given angle θ_i and energy E_i can be expressed as:

$$\frac{d\sigma}{d\Omega}(\theta_i, E_i) = \frac{1}{4\pi}\frac{n_{GE}(\theta_i, E_i)}{n_{FC}(E_i)}\frac{\varepsilon_{FC}\,\sigma_{U,f}(E_i)}{\varepsilon_{GE}(E_i)}\frac{\varsigma_{FC}}{\varsigma_{sple}}\frac{s_{FC}}{s_{sple}} \qquad (1)$$

where n_{GE} and n_{FC} represent the dead time corrected numbers of detections for a given ray in the Ge energy spectrum and for the fission chamber high energy spectrum respectively, ε_{GE} and ε_{FC} the germanium detector's and the fission chamber's efficiency, $\sigma_{U,f}$ the ^{235}U fission cross-section, ς_{FC} and ς_{sple} the areal densities of the uranium layer in the fission chamber and the sample, s_{FC} and s_{sple} the surfaces of the uranium layer in the fission chamber and the sample.

Angle integration

The quantity of interest is the total reaction cross-section which requires integration of equation (1). One can show that the differential cross-section can be expressed as a finite sum over even degree Legendre polynomials [7,8]:

$$\frac{d\sigma}{d\Omega}(\theta) = \frac{\sigma_{tot}}{4\pi}\cdot\sum_{i=0}^{\infty}\alpha_i P_i(\cos\theta) \qquad (2)$$

where σ_{tot} is the total angle integrated cross-section, and the α_i are coefficients ($\alpha_0 = 1$) depending on the angular momentum of the initial and final state J_i, J_f and the transition multipolarity L [7,8]. As the highest order Legendre polynomial in the decay distribution has order ≤2L and ≤2J_i, this infinite summation can be limited to M terms, where M=min{2L,2J_i}.

Usually the sum can be limited to even Legendre polynomials up to the order of 6 as the contribution of higher-order polynomials is small. Under this assumption the integrated cross-section can be obtained in very good approximation from measurements at two angles where the value of the fourth-order Legendre polynomial P_4 is zero according to:

$$\sigma_{tot} \approx 4\pi\left[w_1^*\frac{d\sigma}{d\Omega}(\theta_1^*) + w_2^*\frac{d\sigma}{d\Omega}(\theta_2^*)\right] \qquad (3)$$

with θ_1^* = (30.6°or 149.4°), θ_2^* = (70.1° or 109.9°), w_1^* = 0.3479 and w_2^* = 0.6521 [7,8].

Results

In this paper, the results for two different measurement sets of (n,xn γ) cross-sections on ^{235}U and ^{232}Th are presented and compared to the prediction of the TALYS code [1].

Samples and running time

The two samples used in these experiments have the following characteristics (see Table 1):

Table 1: sample characteristics and measuring time

	Purity (%)	Total mass (g)	Surface (cm²)	Thickness (mm)	Running time (h)
^{235}U	93.18 ± 0.031	37.43 ± 0.01	113.173 ± 0.070	0.211 ± 0.006	1248
^{232}Th	99.5	11.9939 ± 0.0001	36.463 ± 0.195	0.302 ± 0.004	375

Cross-sections

Averaged cross-sections were derived from the time-of-flight spectra for neutron energy bins of suitable sizes. The integral cross-sections were summed according to Eq. (3) using the results obtained with the four HPGe detectors from both angles 110° and 149°. The cross-sections are not corrected for internal conversion. The total uncertainties vary from 5 to 7%.

Table 2 summarizes the γ transitions observed in various nuclei created by (n,n') and (n,2n) reactions on ^{235}U and (n,n'), (n,2n) and (n,3n) reactions on ^{232}Th.

Table 2: g transitions observed corresponding to ^{235}U(n,n'), ^{235}U(n,2n) and ^{232}Th(n,n'), ^{232}Th(n,2n), ^{232}Th(n,3n) reactions

Target	Reaction	Gamma Energy (keV)	Initial level	Final level
^{235}U	n,n'	129.3	$5/2^+$	$7/2^-$
	n,2n	152.7	6^+	4^+
	n,2n	200.9	8^+	6^+
	n,2n	244.2	10^+	8^+

Target	Reaction	Gamma Energy (keV)	Initial level	Final level
^{232}Th	n,n'	49.4	2^+	0^+
	n,n'	112.75	4^+	2^+
	n,n'	171.1	6^+	4^+
	n,n'	223.7	8^+	6^+
	n,n'	550.4	5^-	6^+
	n,n'	612.3	3^-	4^+
	n,n'	665	1^-	2^+
	n,n'	714.2	1^-	0^+
	n,n'	774.1	2^+	0^+
	n,n'	681.1	0^+	2^+
	n,n'	735.9	2^+	2^+
	n,n'	780.2	3^+	2^+
	n,2n	185.7	$5/2^-$	$5/2^+$
	n,3n	182.5	6^+	4^+

The ^{235}U isotope

Figure 3 shows the results obtained for the (n,n' γ) and (n,2n γ) reactions on ^{235}U compared to the only existing experimental data [9] and TALYS calculations [10].

The used TALYS code was very well optimized for the fission cross-section of the isotope of interest and its descendants [10] as shown in Figure 4.

One can remark that the total cross-sections for the (n,n') and (n,2n) reactions are quite well predicted by the TALYS code. But, nevertheless, the agreement with our experimental data is not so good for (n,xn γ) reactions. In the case of the 129.3 keV γ-transition in ^{235}U, the discrepancies between experimental data and TALYS are rather small but the shape is not well reproduced in the 1 – 6 MeV neutron energy range. Concerning the (n,2n γ) cross-sections, the behaviour of the experimental data is well predicted by the code but as one can see, in Figure 3 (b, c and d), a factor of 0.455 exist between theory and experiment. For the 152.7 keV γ-transition in ^{234}U created by the ^{235}U(n,2n) reaction, experimental data from [9] exist but they have been normalized to theoretical predictions by a factor whose value is not specified in the paper.

Figure 3: Total γ-production cross-sections due to a/ the ^{235}U(n,n') reaction for the 129.3 keV transition (state 5/2^{+} -> 7/2^{-}) and due to ^{232}U(n,2n) reaction for the b/ the 152.7 keV transition (state 6^{+} -> 4^{+}) c/ the 200.97 keV transition (state 8^{+} -> 6^{+}) and d/ the 244.2 keV transition (state 10^{+} -> 8^{+}). The results are compared to TALYS predictions and experimental data if they exist. Our data points are connected by a solid line.

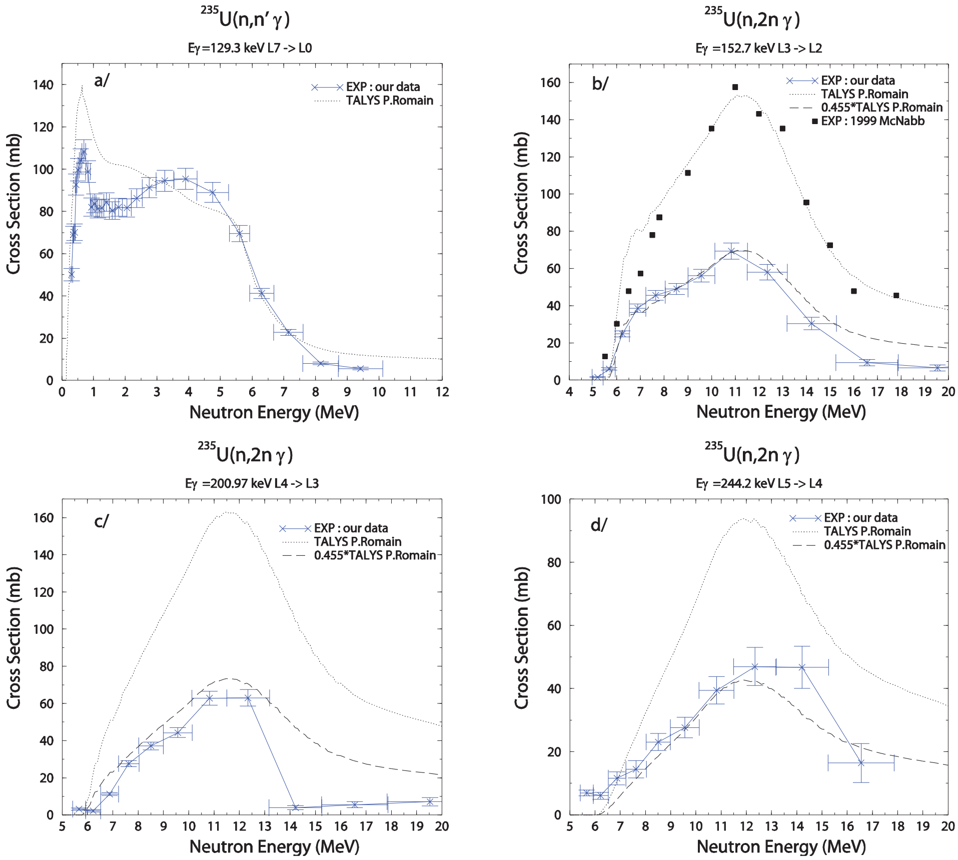
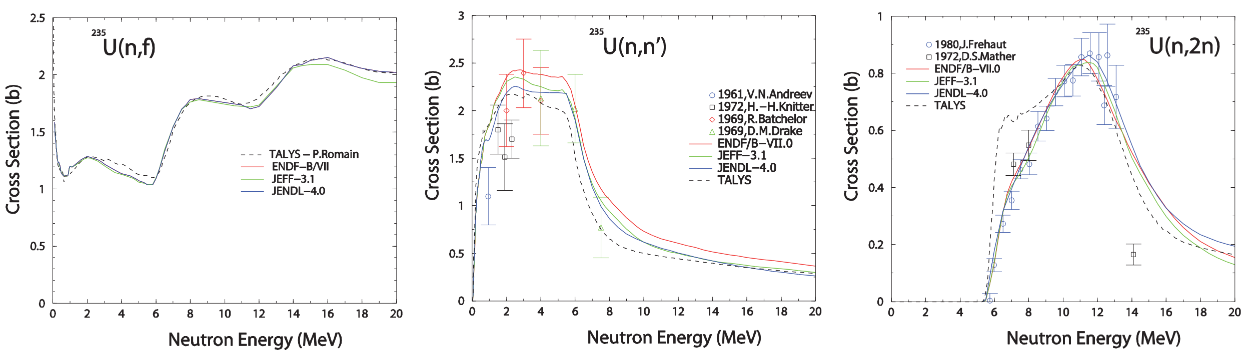


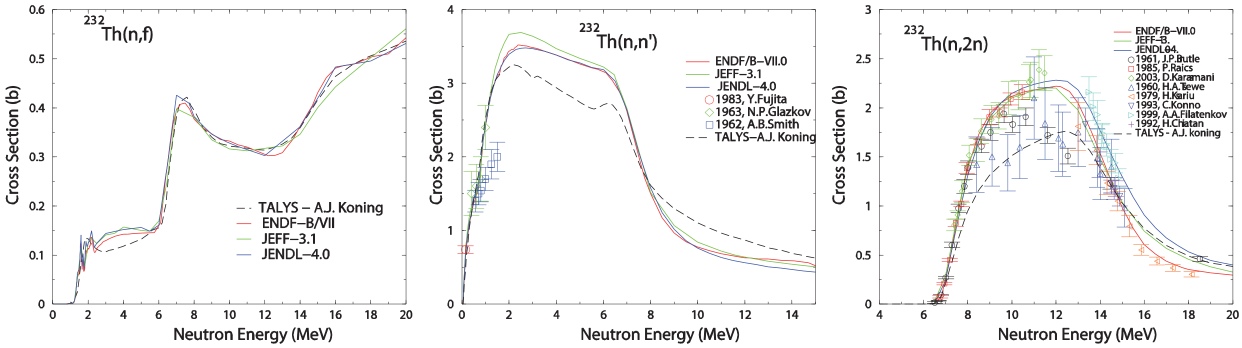
Figure 4: TALYS total cross-section predictions for the (n,f), (n,n') and (n,2n) reactions on ^{235}U compared to evaluated databases and experimental data (EXFOR)



The ^{232}Th isotope

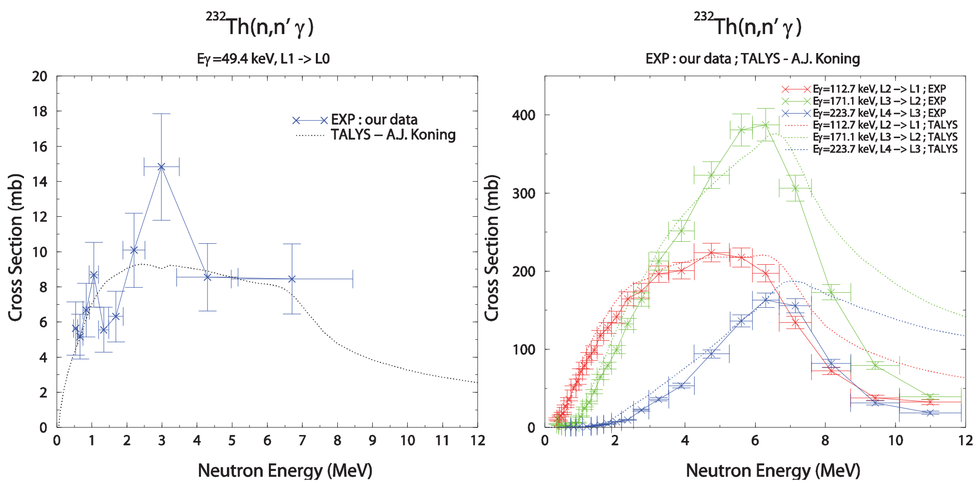
The total (n,n') reaction cross-section has been already measured in the range 0- 2 MeV while the (n,2n) reaction has been well studied as shown in Figure 5. One can see also that the TALYS calculations of the fission process in ^{232}Th, made by A.J. Koning, is in good agreement with the evaluated data. On the contrary, the theoretical predictions of the total (n,n') and (n,2n) cross-sections seem to be underestimated in comparison with experimental and evaluated data.

Figure 5: TALYS total cross-section predictions for the (n,f), (n,n') and (n,2n) reactions on ^{232}Th compared to evaluated databases and experimental data (EXFOR)



Concerning our (n,n' γ) cross-section measurements, one can note in Figure 6 that the TALYS calculations are in good agreement with the experimental data except above roughly 8 MeV where the decrease of the excitation function is overestimated by the code. It has to be mentioned that the uncertainties of the 49.4 keV γ-transition cross-section are rather large due to the low statistic in this peak (highly converted).

Figure 6: Total g-production cross-sections due to ^{232}Th(n,n') reaction for the 49.369 keV transition (left) (state 2^+ -> 0^+) and the 112.75 keV transition (state 4^+ -> 2^+), the 171.2 keV transition (state 6^+ -> 4^+) and the 223.6 keV transition (state 8^+ -> 6^+) (right). The results (connected by a solid line) are compared to TALYS predictions (no experimental data exist in this gamma energy range).



Concerning higher γ energy transitions (Figure 7) coming from the deexitation of the other exited bands to the states of the ground state band, the agreement between TALYS calculations and experimental data is worse than for γ-transitions between states belonging to the groundstate band. In this case, we were able to compare our data, in the 0 – 3 MeV neutron energy range, with existing experimental ones from [11] and the agreement is very good.

For the (n,2n γ) and (n,3n γ) reactions, we have, for the moment, obtained the cross-sections for the 185.7 keV γ-transition in ^{231}Th and for the 182.5 keV γ-transition in ^{230}Th. In the last case, the uncertainties are rather large because of the combined effect of the lower cross-sections and the low neutron incident flux (Figure 2). As for the results obtained with the ^{235}U isotope, those reactions (with x>1 and neutron energies higher than ~ 8 MeV) are not well predicted by TALYS. More precisely, the behaviour of the excitation functions is well reproduced but for the (n,2n γ) reaction the TALYS code underestimates the cross-section by a factor of 2.58 and for (n,3n γ) reaction, the calculated cross-section is overestimated by a factor of 0.72.

Discussion

These exclusive measurements are very useful to test the predictive power of theoretical calculations. As we have seen before, it is not sufficient for a model code to calculate correctly the total reaction cross-section of a process to predict with the same confidence level the γ production cross-sections. Indeed, the code has to take in consideration a lot of structure parameters like branching ratio, level densities, spin distributions etc… And even on an extensively studied nucleus like ^{235}U, the TALYS code is not yet able to predict the (n,xn γ) reaction cross-section correctly. These kinds of experimental data are thus very important and allow the improvement of the knowledge of these parameters.

There are indications that the semi-classical exciton model simply fails to properly describe the spin distribution that accompanies the pre-equilibrium process. Large deviations, in the right direction, from the exciton model are found [12,13] when a quantum-mechanical multi-step direct model is used instead of the exciton model. The correct spin dependence of the underlying DWBA cross-section is then taken into account [14], as opposed to a posteriori assigning a spin distribution to the cross-sections. It is expected that theoretical agreement will be improved if so-called FKK or other multi-step direct models are included.

Conclusion and perspectives

After a consequent work to reduce the uncertainties of these measurements, we are currently able to measure (n,xn γ) cross-sections of a precision ranging from 5 to 7%. The new data on ^{235}U and ^{232}Th presented in this paper, will thus complete the knowledge of γ production in (n,xn) reactions. These results will be subject to further investigations from experimental as well as from theoretical point of view.

In the framework of ANDES project (7th Framework program), future measurement campaigns will be devoted to the measurement of ^{238}U(n,xn γ) cross-sections. Indeed, some experimental data already exist but are not in agreement with each other as it can be seen in [12]. Nevertheless, with our experimental set-up, we will be able to provide cross-sections with reduced uncertainties to complete the knowledge of these reactions.

Acknowledgements

The authors thank the team of the GELINA facility for the preparation of the neutron beam and for their strong support day after day. This work was partially supported by the Integrated Project for European Transmutation (EUROTRANS) and by the transnational access schemes Neutron data measurements at IRMM (NUDAME) and European facility for innovative reactor and transmutation data (EUFRAT).

Figure 7: Total g-production cross-sections (preliminary results) due to the ^{232}Th(n,n') reaction for g-transitions from few exciting band to the ground state band compared to J.H.Dave et al. [11] and TALYS predictions. As the existing data have been measured only up to 3 MeV, the comparison is done only in this neutron energy range. Our experimental data are connected by a solid line.

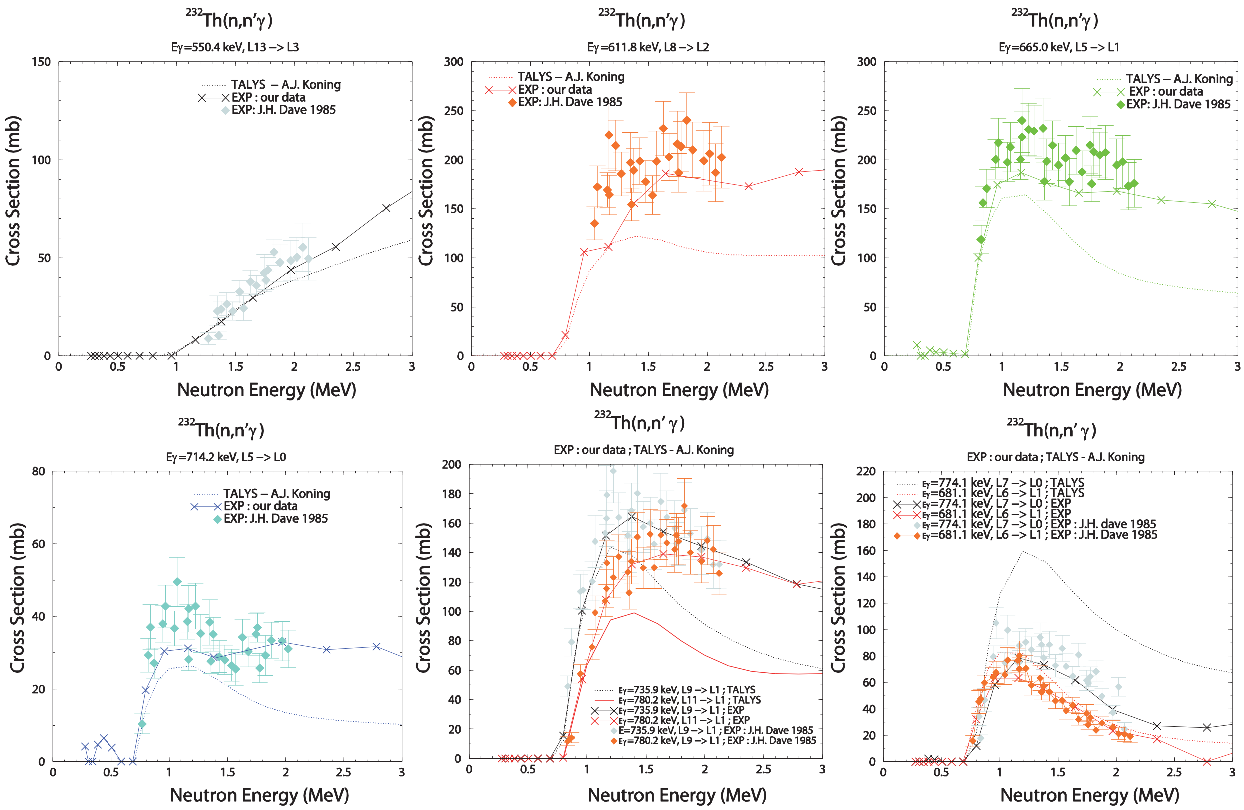
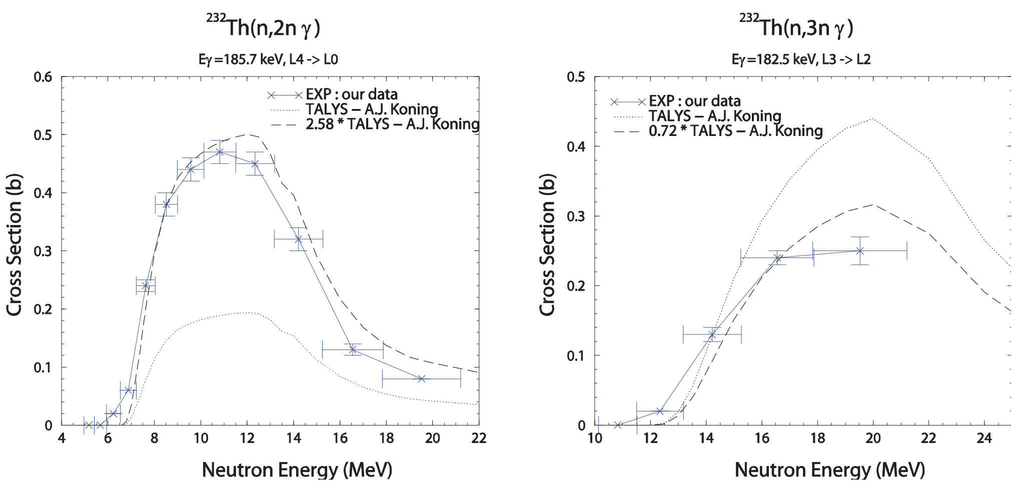


Figure 8: Total g-production cross-sections due to the ^{232}Th(n,2n) reaction for the 185.7 keV transition (left) (from state 5/2- -> 5/2^{+}) and due to the ^{232}Th(n,3n) reaction for the 182.5 keV transition (right) (from state 6^{+} -> 4^{+}). Our experimental data are connected by a solid line.



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