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Measurement of (n,xnγ) reactions at high precision

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Abstract

The design of Generation IV nuclear reactors and the research of new fuel cycles require knowledge of cross-sections for different nuclear reactions. Our work is focused on the determination of the cross-sections of (n,xnγ) reactions occurring in these new reactors. The aim is to measure unknown cross-sections and to reduce uncertainty on present data relative to reactions and isotopes present in transmutation or regeneration processes.

The current study is relative to measuring ^{232}Th(n,n'γ) and ^{235}U(n,xnγ) reactions in the fast neutron energy domain (up to 20 MeV). The experiments are performed at GELINA (IRMM, Belgium) which delivers a pulsed, white neutron beam. The time characteristics of the beam enable us to measure neutron energies with the time of flight (TOF) technique. The neutron induced reactions (in this case inelastic scattering and (n,2n) reactions) are identified by online prompt γ spectroscopy with an experimental setup including 4 HPGe detectors. A double layered fission chamber is used to monitor the incident neutron flux.

As the precision is a key issue in these measurements, the detectors used in the experiment were studied extensively in order to reduce the uncertainties of the results to a minimum. Simulations and calibration experiments were realised and are presented.

Introduction

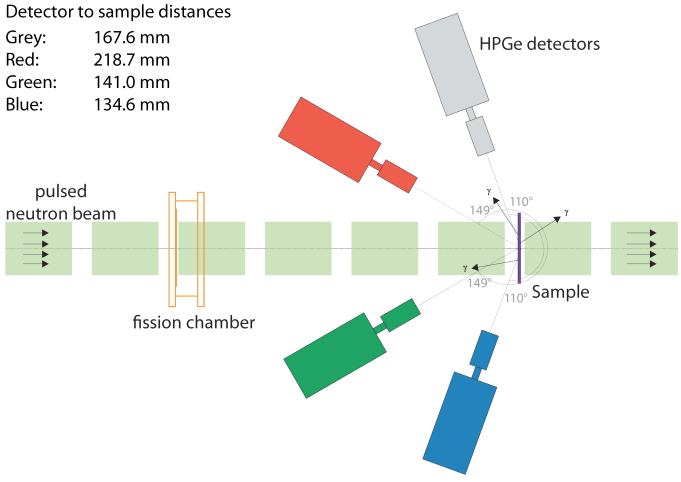
Knowledge of (n,xn) reactions is of crucial importance for the development of the new Generation IV nuclear reactors. These future systems will explore new energy domains and in order to optimise their design, the measurement of (n,xn) reaction cross-sections at a high level of precision is absolutely mandatory. Indeed, these reactions are an important energy loss mechanism, lead to neutron multiplication and production of radioactive isotopes.

The measurements are realised using the (n,xnγ) technique, for which a high precision experimental setup was developed and is presented here. It is meant to be used to probe reactions for which a high precision is required, such as for the ^{238}U(n,n') reaction cross-section, part of the NEA High Priority List [1]. The ultimate goal of developing these measurement techniques is to study (n,xn) reactions on ^{233}U, lacking experimental data, which is of upmost importance for the thorium cycle. For example, the ^{233}U(n,2n)^{232}U reaction leads in its decay to ^{208}Pb, emitter of a 2.6 MeV γ ray. Presence of such energetic photons has a major impact for the reactor core temperature, and therefore needs to be studied precisely.

Experimental setup

The experimental setup, shown in Figure 1, as well as the applied measurement techniques are treated in this section.

Figure 1: Experimental setup used at GELINA, FP16/30m



The (n,xnγ) technique

A sample enriched in ^{A}X isotopes is irradiated by a neutron beam inducing (n,xn) reactions, which produce $^{A-(x-1)}$X isotopes in excited states. Decay of these isotopes leads to emission of characteristic γ rays, witnessing a prior reaction. The intensity of these γ rays yields the cross-section of isotope production in a given excited state. The data can be used to validate theoretical codes, such as TALYS, which is able to predict (n,xnγ) reaction cross-sections (see *e.g.* ref. [2]).

The TOF technique

The experiments presented in this work were performed at the GELINA facility of the IRMM, Geel, Belgium, which produces a white, pulsed neutron beam using the (γ,xn) and (γ,F) reactions on a depleted uranium target. The energy domain of the produced neutrons ranges from a few keV up to several MeV [3,4].

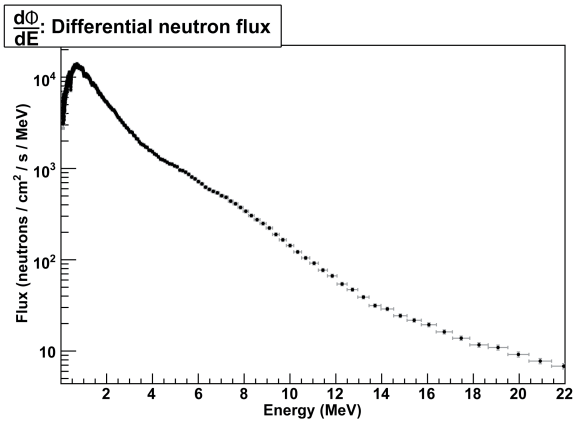
The TOF technique is used to determine the incident neutron energy and the presence of a γ flash enables us to calibrate the time spectra. In order to obtain the best compromise between time (and thus neutron energy) resolution and flux intensity (resolution of 1 MeV at E_n = 20 MeV) the setup was installed at 30 m distance from the neutron source.

Flux monitoring

The uncertainties on the incident neutron flux (Figure 2) influence strongly the quality of the cross-section measurements. For this reason special effort has been made to obtain very precise flux data.

A fission ionisation chamber with a uranium sample is used to determine the neutron flux. The deposit, highly enriched in ^{235}U (>99.5%), is very thin: 324 μg/cm^2 and was made of vacuum evaporated $^{235}UF_4$. 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), as explained later.

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



γ detection

4 High Purity Germanium (HPGe) detectors, referred to as *grey*, *red*, *green* and *blue* are used to observe the γ rays emitted by the created isotopes. These semiconductor counters are made of semi-planar crystals with depths ranging from 2 to 3 cm and surfaces between 10 and 28 cm^2. This type of detectors has been chosen as they allow for high energy resolution at low γ ray energy together with a good timing. In order to take into account the angular distribution of the emitted γ rays, they are placed at 110° and 149° with respect to the incident neutron beam. Backward angles were chosen to reduce dead time caused by the observation of events due to γ flash scattering, amounting up to 60% of the detections.

Efficiency of the fission chamber

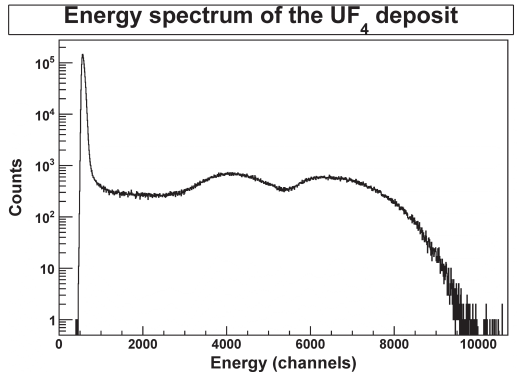
As mentioned previously, the precision of the (n,xnγ) measurements is of upmost importance. This section treats the efforts made on reducing the uncertainty caused by the fission chamber. Indeed, this detector presents one of the major sources of error on the final results. For this reason, an extensive work was realised on the determination of the fission chamber efficiency.

Determination of the fission yield

An energy spectrum for the UF_4 deposit acquired during (n,xnγ) measurements at the GELINA facility is shown in Figure 3. In the low energy domain of the energy spectra one can see a very strong contribution coming from the α particle signals, which are due to the radioactivity of the uranium

foils. To the right, in the higher energy ranges one can observe events due to fission products in a double-humped shape.

Figure 3: Energy spectrum for the fission chamber deposit UF4 in its initial configuration: the gap between the electrodes is 8 mm with reverse biasing (HV on the deposit)



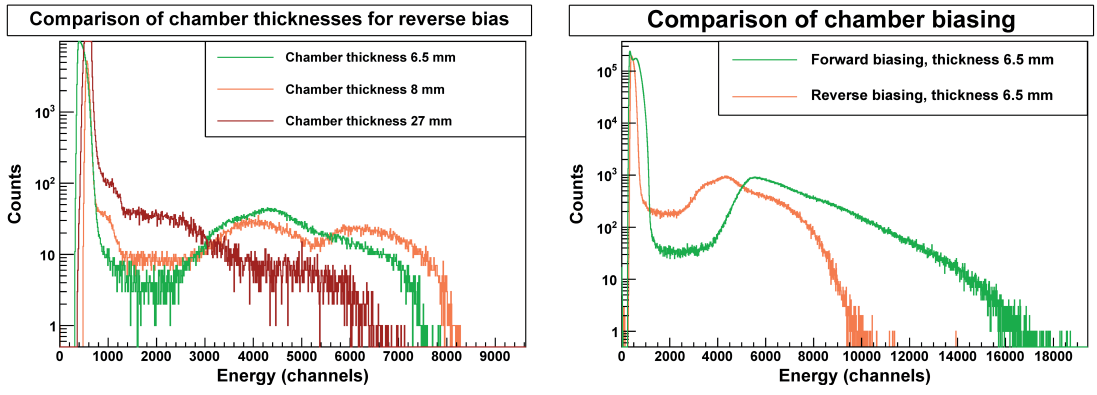
Separating the α particle signals from the fission events is a delicate process. In fact, the signal amplitudes generated both by the uranium decay and the neutron induced fissions have a respective upper limit value, *i.e.* their pulses are always weaker than a certain energy, but they do not have a lower limit. This means that the fission product signals share a part of their energy domain with the α particle signals which makes a strict separation between both types of signals very difficult. The only acceptable approach to achieve this goal is to apply an energy threshold to discard the α component and to correct for the portion of fission product signals lost.

Optimisation of the fission chamber configuration

As a threshold has to be applied to the energy spectra, it is important to configure the fission chamber in a way that as few events as possible are lost in this procedure. This means that we have to determine the configuration which leads to the best separation between α decays and fission products.

To achieve this, several measurements were performed. First, the thickness of the fission chamber was modified. Thicknesses of 6.5, 8 and 27 mm were chosen. The result is shown in Figure 4 (left). This comparison shows that the best configuration is to have a 6.5 mm gap between the anode and the cathode. Indeed, in this scenario the α particles are penalised with respect to the fission products. As their range is longer and their kinetic energy rather high, most of the energy loss occurs

Figure 4: Comparison of the different configuration possibilities of the fission chamber: (a) shows the effect of varying the effective volume of the chamber, (b) shows the effect of polarisation changes



in the Bragg peak, whereas fission products lose their energy almost continuously. Increasing the volume thus means detecting more α particles, and filling up the valley between both contributions.

In a second study the impact of biasing was analysed. Figure 4 (right) shows the difference obtained when applying a reverse bias (HV on the deposit) or a forward bias (HV on the opposite electrode). In fact, the signal left in the fission chamber depends on the charge created, and thus the energy deposited, but also on the distance to the collection electrode. For this reason, fission products stopping in the chamber before reaching the opposite electrode create a larger signal in the forward bias scenario than in the reverse bias case. It is therefore preferable to apply forward biasing.

Correction for signal loss

In order to correct for signals lost, we distinguish between two different types. First, there are fission products created in the deposit without sufficient energy to leave the foil creating no signal at all. Secondly there are fission products producing a weak signal which is lost as it falls below the threshold in the energy distributions.

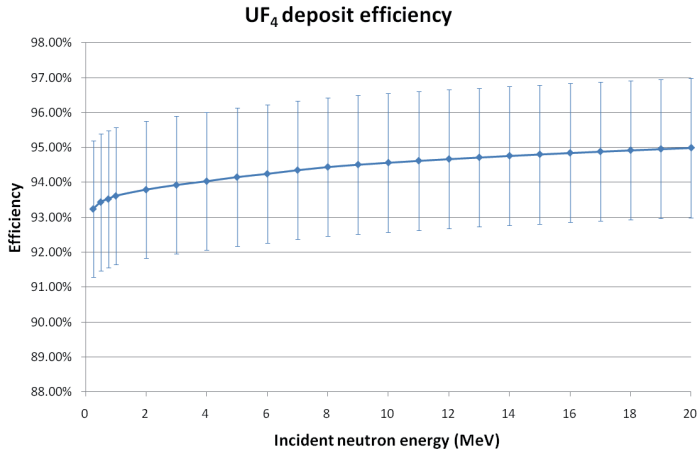
Whereas the first type can be corrected easily by applying well-known techniques [5,6], the second type cannot be corrected so easily. For this reason Geant4 [7] simulations were performed in order to reproduce the spectra and to show the behaviour of the fission products at low energy. The results in this region were however not satisfactory. Indeed, at very low energy, the energy loss of the fission products is not well known, which leads to a systematic difference between the simulated and measured spectra. Because of this, we cannot conclude how to extrapolate the fission product contribution towards low energies: A linear extrapolation to zero amplitude appears plausible, but there is no conclusive proof that this is adequate. In fact, for reverse biasing the simulations show that the expected loss is larger than would be inferred from a linear extrapolation while in contrast linear extrapolation is expected to be adequate for a forward bias. Again, given the imperfect nature of the simulations the situation is inconclusive.

Calibration measurements at PTB

As the previous correction strategies were not reliable, the fission chamber was calibrated using a monoenergetic (E = 8.4 MeV) neutron beam produced at the Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig. The beam was well characterised by different reference measurements of high precision, including a recoil proton telescope (RPT) and a liquid scintillation detector [8]. The neutrons were produced through D(d,n)^{3}He reactions. Time of flight and energy information were saved in list mode files, enabling time gating on the acquired data, to eliminate neutrons with wrong energy, *e.g.* born through break-up reactions in the deuterium target or slowed down through scattering in the experimental hall. For the measurements several foreground runs were performed to obtain statistics of at least 10000 events in the fission chamber. Two series of background measurements were made: the first by placing a shadow cone between the neutron source and the detector to evaluate the indirect component of fission events generated and a second by taking the deuterium gas out of the target and bombarding the empty target with deuterons to estimate the amount of neutrons created by other mechanisms than that of the direct reaction.

After corrections for acquisition dead time and air attenuation, these measurements enabled us to determine the efficiency to detect a fission event in the ionisation chamber to be (94.4 ± 2.1)%, when applying a threshold in the middle of the plateau separating α particles and the main fission product contribution. Using this value, measured at 8.4 MeV, as a reference together with the corrections for events lost due to absorption in the foil [5,6], we can calibrate our efficiency curve shown in Figure 5.

Figure 5: Final efficiency values as a function of incident neutron energy



Efficiency of the HPGe detectors

Another large source of uncertainties comes from the evaluation of the γ-detection efficiency of the HPGe detectors. This section presents the work that has been realised on the determination of this quantity.

Efficiency computing method

The absolute photopeak-efficiency of a germanium detector in the case of extended samples depends on several parameters, which have to be taken into account in the total efficiency calculation. Besides the position of the detector with respect to the γ-ray emitting sample and the energy of the studied γ ray, these parameters also include:

- the shape of the γ-ray emitting sample (in our case this corresponds to the beam diameter of the order of 5 cm)
- the geometry of the crystal inside the detector
- the self-absorption $k_{E\gamma}$ of the γ rays inside the sample

Calibrations realised with a point source only are not sufficient in our case, as the studied sample is not point-like, *i.e.* the geometrical distributions of the emitted γ rays cannot be calculated at a satisfactory level. Another difficulty is introduced by the absorption of the γ rays inside the sample which is an important factor as the density and thickness of the used samples are rather high.

The best way to consider all these parameters is to measure the efficiency with calibrated sources and to compare these to Monte-Carlo simulations of the detector [9]. The procedure for this method is realised in several steps: First, the geometry of the crystal is determined and entered into a Geant4 simulation code. Then calibrated sources (point-like and extended) are placed at different spots of the sample position. The obtained results are compared to the simulated spectra. The differences in the yields are used to determine the dead layers of the crystal and to calibrate the simulation input. Once the simulation parameters are fixed, the second step consists of simulating the studied sample. For this purpose the geometry of the sample is entered into the simulation code and γ rays of the energies of interest are shot randomly within the sample. The ratio between the number of events in the photo-peak and the number of γ rays simulated is the absolute peak-efficiency of the detector.

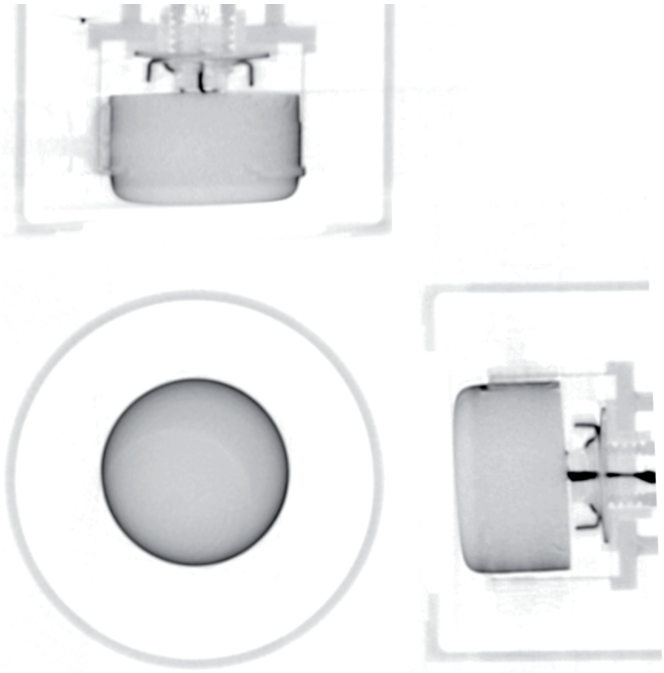
Microfocus computer tomography of the detectors

As the precision of the constructor's specifications of the detector geometry was not reliable enough to perform accurate simulations, the internal crystal dimensions had to be verified. A non-destructive way to accomplish precise crystal measurements is to expose the detector to a radiative source. This was realised at the *Microfocus Computer Tomography* device of the *Department of Metallurgy and Materials Engineering* (MTM) at the *Katholieke Universiteit Leuven* (KUL).

A polychromatic X-ray source (Philips HOMX 161) operated at a voltage of 125 kV and a current of 0.28 mA, filtered by a 2 mm aluminium and a 1 mm copper shield [10], was used for the experiment. The data acquisition was realised through a CCD camera (Adimec MX12P) delivering 12 bits grey scale images at a resolution of 1024 × 1024 pixels, resulting in a voxel size of 110–150 µm. The detectors were placed on a rotating table and pictures were taken ever. 0.5°. The data were processed with the AEA Tomohawk software for 3D reconstruction.

With this configuration we were able to visualise the internal geometry of the detector. For the chosen beam energy, the detector cap was transparent, whereas the high density germanium crystal was not penetrated. Figure 6 shows the results of this experiment for the green detector. In this picture we can clearly recognise the germanium crystal held by an aluminium structure at the centre, surrounded by the detector cap. Knowing the precise dimensions of the cap, we were able to calibrate the experimental data and to determine the size of the crystal.

Figure 6: µ-ct analysis of the green detector, showing different cuts of the reconstructed data



For the small crystals of the green and blue detectors we found that the diameter of the crystal was indeed a little smaller as the manufacturer specified. Moreover, the difficulties encountered in simulating the green detector could be explained by a wrong crystal to detector entrance window distance, which is in fact 3 mm longer than expected. The values determined here are in good agreement with the simulation results obtained by Geant4. Further investigations with the manufacturer also led to better specifications of the red and grey detectors, for which an agreement was found in the simulations.

Efficiency results

The efficiencies were simulated with the best fitted detector geometries and dead zone sizes obtained from the discussions above, and measurements were performed using different sources. The final experiment to simulation ratios of our calibration measurements are shown in Figure 7. In total 7 measurement series have been performed. A ^{152}Eu point source was placed at the centre position of the sample, and then displaced by 12 mm to the right, left, up and down. Two further measurements were realised with a ^{133}Ba point source and an extended ^{152}Eu source of 50 mm diameter at the centre position to check the previous results.

Figure 7: Experiment to simulation ratios for the efficiency calibrations for different source positions (see legends) of the four detectors

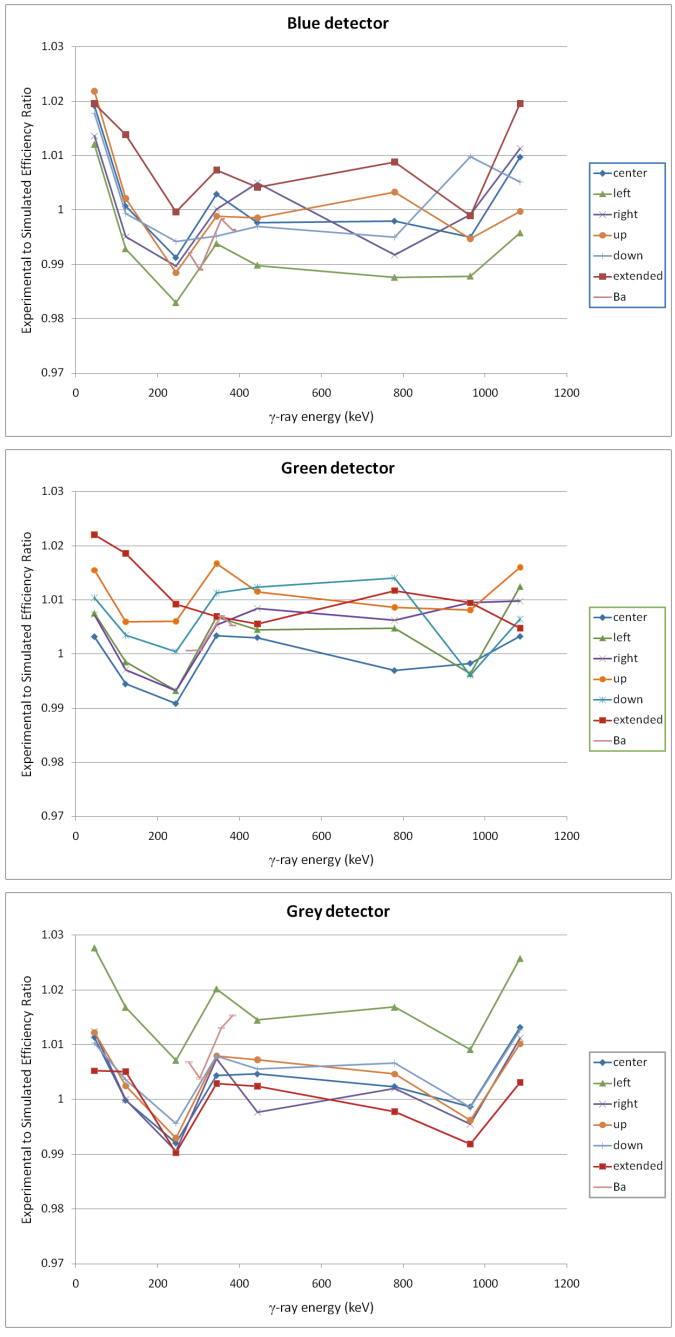
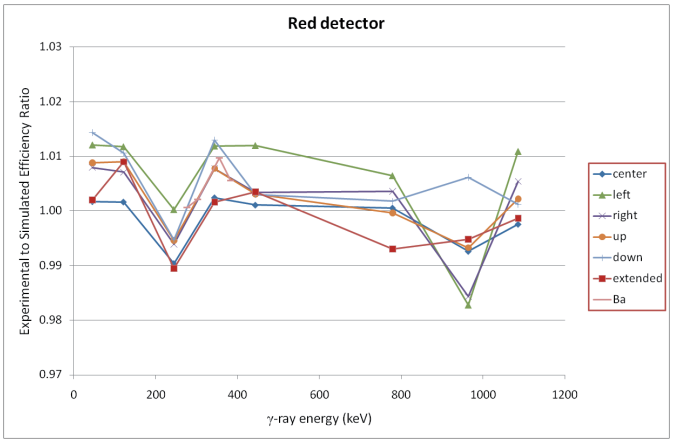


Figure 7: Experiment to simulation ratios for the efficiency calibrations for different source positions (see legends) of the four detectors *(continued)*



One can observe that the values obtained during the different measurement series all lie within a range of ± 2% with respect to the simulated values for the intermediate energy range (from 121.78 to 778.90 keV). For the low energy values, mainly the first point at 46 keV, the experimental values are differing a little more from the simulated ones. Indeed this region is more sensitive to dead zones and the rounding parameter at the front of the crystal as well as on the absorption of the detector window. The results for this domain are however sufficient for our needs.

The deviation between data and calculation in the intermediate energy domain can be explained by uncertainties on branching ratios and also by the fact that the simulations did not account for complete energy spectra. In fact, only the energies of interest were simulated, neglecting smaller energies rays due to background radiation. This can lead to small errors when subtracting the rays from the background. Such a particular deviation could me observed for the 244.7 keV ray, which is systematically lower experimentally than in the simulated spectra.

The differences between the measured series are mainly due to the position precision of the source. This can be observed best for the green line, corresponding to the "left" position of the source. For the detectors grey and red it is overestimated experimentally, whereas the detectors blue and green are showing an underestimated value. In fact, the grey and red detectors are positioned to the right-hand side (RHS) of the experimental setup, as described in Figure 1, and the blue and green ones to the left-hand side (LHS). As the experimental values are higher for the RHS placed detectors, this means that the source was placed too much to the right. This is confirmed by the fact that the outer detectors, *i.e.* the grey and the blue one, see a larger difference. Indeed as they are located at 110° they have a grazing angle to the target plane and a variation of the source position is seen more strongly as for the other detectors. Simulations show that a variation of 1% on this ratio of the grazing angle detectors corresponds to approximately 1 mm displacement.

Taking into account the variation of the different points simulated at different energies as well as the systematic uncertainties coming from the calibration sources used, this procedure enabled us to determine the γ efficiencies at uncertainties ranging between 2 and 3% in the energy domain of interest.

Conclusions

The aim of this work was to analyse all the different components used in the experimental setup to reduce the error to a minimum. This was achieved through an extensive analysis of the detection efficiencies of the used counters. For the fission chamber we were able to reduce the uncertainties on the efficiency to 2.1% and the Microfocus Computer Tomography helped to obtain uncertainties on the γ-detection efficiencies ranging between 2 and 3% depending on the photon energy.

With these efforts, the current experimental setup is able to measure (n,xnγ) reaction cross-sections at uncertainties of 5-6%, coming from the before mentioned efficiency uncertainties as well as from other errors due to target specifications and yield extraction. It has already been used to probe (n,n'γ) and (n,2nγ) reactions on ^{235}U and (n,n'γ), (n,2nγ) and (n,3nγ) reactions on ^{232}Th [11].

Acknowledgements

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