Recent capture cross-sections validation on ^{232}Th from 0.1 eV to 40 keV and self-shielding effect evaluation

L. Perrot, A. Billebaud, R. Brissot, A. Giorni, D. Heuer, J.M. Loiseaux, O. Meplan, J.B. Viano

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RECENT ^{232}TH CAPTURE CROSS SECTION VALIDATION ON FROM 0.1 EV TO 40 KEV AND SELF-SHIELDING EFFECT EVALUATION

L.perrot, A.Billebaud, R.Brissot, A.Giorni, D.Heuer, J.M.Loiseaux, O.Meplan and J.B.Viano

Institut des Sciences Nucléaires / CNRS-IN2P3 / UJF
51 avenue des Martyrs 38026 Grenoble - France

Abstract

Research on accelerator driven systems (ADS), related new fuels and their ability for nuclear waste incineration has led to a revival of interest in nuclear cross sections of many nuclides over a large energy range. Discrepancies observed between nuclear data bases require new measurements in several cases. A complete measurement of such cross sections including resonance resolution consists of an extensive beam time experiment associated to a long analysis. With a slowing down spectrometer associated to a pulsed neutron source, it is possible to determine good cross section profile in an energy range from 0.1eV to 40keV by making use of a slowing-down time lead spectrometer associated to a pulsed neutron source. These measurements performed at ISN (Grenoble) with the neutron source GENEPI requires only small quantities of matter (as small as 0.1g) and about one day of beam per target.

We present measured cross section profiles and an experimental study of self-shielding effect. A CeF_3 scintillator coupled with a photomultiplier detects gamma rays from neutron capture in the studied target. The neutron flux is also measured with a ^{233}U fission detector and a ^{3}He detector placed symmetrically relative to the PM in relation to the neutron source. Absolute flux values are given by activation of Au and W foils. The cross section profiles can then be deduced from the target capture rate and are compared with very detailed MCNP code simulations, which reproduce the experimental set-up and provide also capture rates and neutron flux. A good agreement between experimental and simulated profiles for well-known cross sections like Au for different thicknesses is found in our energy range, and therefore validates the method and the taking into account of self-shielding effects.

The method is then applied to ^{232}Th which is of main interest for new fuel cycle studies, and is complementary to higher energy measurements made by D.Karamanis et al. [1]. Results obtained for three target thicknesses will be compared with the simulations based on different data bases. Special attention will be paid to the region of unresolved resonances (>100eV).

I Introduction

At the dawn of XXI century, energy is a crucial problem to study. By 2050, it is predicted that the energy demand will double. In the same time, some green house effect emission scenarios predict approximately the same increase in this effect. The nuclear energy contribution to the total energy

production in the world represents about 6%. Nuclear development and utilization can be one of the possible responses to the energy demand increase and the green house effect limitation.

However, it is necessary to minimize the radioactive waste production as actinides and long-lived fission products and this can be obtained by using the ^{232}Th-^{233}U fuel cycle, in an accelerator driven system or critical reactors.

But a good prediction for this new way of producing energy is strongly dependent on the neutronic properties of materials and more particularly the ^{232}Th capture cross section.

In the present work, it is proposed an experimental method that allows a validation of available data bases. A lead slowing-down spectrometer coupled with a neutron pulsed generator allows to measure reaction rates (n,γ) or (n,f) over a wide energy range from 0.1eV to 40keV for different thicknesses of targets. Experimental data are then compared to precise simulation calculation using ENDF/BVI, JEF2.2 and JENDL3.2 data bases.

The Gold results for which the capture cross section is well known, provides a validation of the method for three different thicknesses. Tantalum, Indium and Thorium data for three thicknesses (for self-shielding effects studies) are presented. The accuracy of the validation method is estimated to be around 5%. Form 0.1eV to 300eV, it is shown that predictions of reaction rates using the different data bases agree among themselves and with the experimental data. From 300eV up to 40keV the discrepancies between data bases predictions can be as large as ±20%. The experimental data allow to either indicate the best data bases to be used, or a need to remeasure the cross section in a certain energy range. For the ^{232}Th, experimental capture rate data agree with the predictions of ENDF/BVI and JEF2.2 bases within 5%.

II The experimental set-up

The neutron source:

This accelerator has been especially designed for neutronic experiments being carried out in the nuclear reactor MASURCA located at the CEA Cadarache Center (France). In fact, for these experiments the neutron pulse length must be of the order of the neutron lifetime in a fast reactor. The pulse intensity must be as big as possible.

The GENEPI (GEnérateur de NEutrons Pulsés Intense) produces fast pulses. The pulse duration is typically 1.0 μs. Deuterons are produced by a duoplasmatron source, especially studied for a pulsed use. The frequency can vary from a few Hz up to 5000 Hz. The deuterons are accelerated to the maximal energy of 250 keV. The maximum peak intensity is 50mA. The deuterons are focused through a five meter long tube (glove finger) onto a deuterium or tritium target. The nuclear reactions D(d,n)^{3}He or T(d,n)^{4}He produce neutrons with energy 2.67 MeV and 14.0 MeV respectively. This accelerator can produce in the case of a tritium target 5.0 10^{6} neutrons/4π per pulse [8].

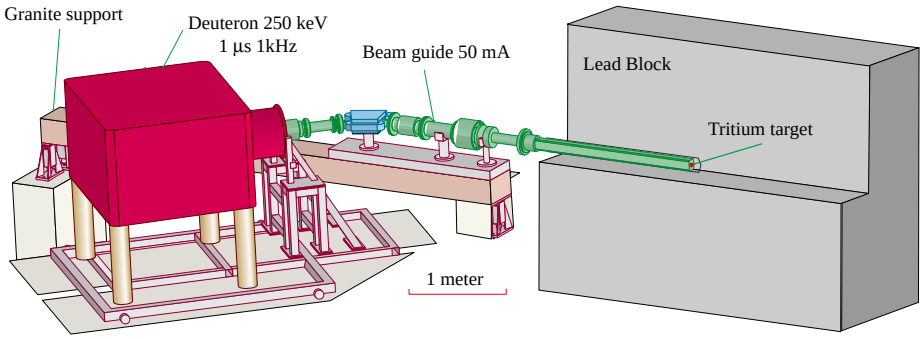
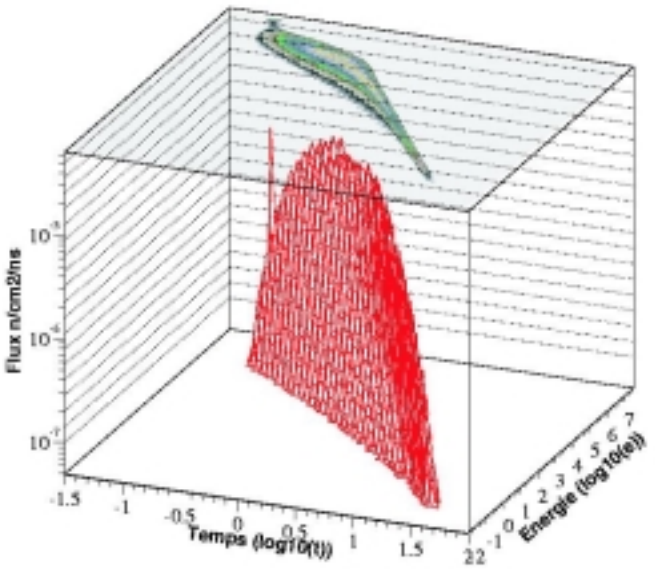


Figure 1: The GENEPI accelerator and the slowing-down time spectrometer

The Slowing down time spectrometer:

The slowing-down time spectrometer is an assembly of 46.45 tons of lead with a cubic symmetry in a relation to the beam axis of GENEPI. The neutron production takes places in the center of the lead block. This block is made up of 8 blocks with dimensions 80 x 80 x 80 cm^3. Each block has two channels (10 x 10 cm^2 in section) parallel to the beam axis. They are used both for handling of the block and for detectors insertion. In this last case, the dimensions of the holes were reduced to 5 x 5 cm^2. Pure lead (99.99%) was chosen to ensure that impurities have negligible effect on the neutron flux. Impurities in lead are less than 5 ppm and are principally Silver, Bismuth, Cadmium, Copper, Antimony, Tellurium. The lead block is shielded with a Cadmium foil to capture the neutron that escape from the lead block and are backscattered by concrete walls, these can perturb the time-energy correlation.

In a slowing-down time lead spectrometer, there is a correlation between the neutron time of flight in the block and its kinetic energy. The scattering mean free path of a neutron in the lead medium, λ_s = 2.76 cm, is about constant over the energy range 0.1 eV to a few tens of keV. The relation between the slowing-down time and the neutron mean energy can be written in the form [2]:



$$\overline{E} = \frac{K}{(t + t_0)^2} \qquad K = 166 \pm 1 \text{keV}\mu s^2 \qquad t_0 = 0.5\mu s$$

Figure 2: The energy-time correlation from MCNP calculation.

The K parameter value, which is a function of the neutron masse m_n, the scattering mean free path λ_s and the medium properties, has been experimentally determinated and well understood by MCNP calculation. The quantity t_0 can be considered as a time correction owing to the fact that the initial neutron is not created with infinite energy but with energy E_0 = 14 MeV and with velocity v_0, it suffers inelastic or (n,2n) reactions before being slowed down only by elastic scattering [4].

Detectors:

i) ***Neutrons source monitoring***:

The reaction on the target produces associated charged particles: α in the case of T(d,n)α reaction and proton in the D(d,p)T reaction which occurs about as often as the D(d,n)^{3}He reaction. The charged particle is emitted at 180° with respect to the neutron. For 0° neutrons, the charged particle goes upstream the incident beam, being focused in the glove finger and is bent by the GENEPI magnet. Two silicon detectors are placed side by side in the vacuum of the magnet chamber. They detect both the α and the p associated to the T(d,n)α and D(d,p)T reactions, since they have the same magnetic rigidity. One of the detectors is covered with a 10 μm Aluminium foil to stop the α particle. We can thus measure through α detection the relative source of 14 MeV neutrons and through p detection of 2.5 MeV neutrons due to D^+ implantation in the T target.

ii) ***Neutron spectrum normalization:***

The detector used for neutron spectrum normalization is a classical proportional ^{3}He-gas counter belonging to a series of counters developed at ISN Grenoble for fast neutrons spectroscopy in reactor. We detect the p and/or t produced by the reaction ^{3}He(n,p)T, Q = 764 keV. It collects the energy deposited by the products of the exothermic reaction. The effective zone is a 6 cm long cylinder of 1 cm in radius. The counter is filled with 70 mbar of ^{3}He, 3.3 bars of Argon and 2.5 mbar of CO_2 (quencher gas). The detector is described in figure 3. It is placed at a symmetrical position relative to the (n,γ) detector in relation to the beam axis.

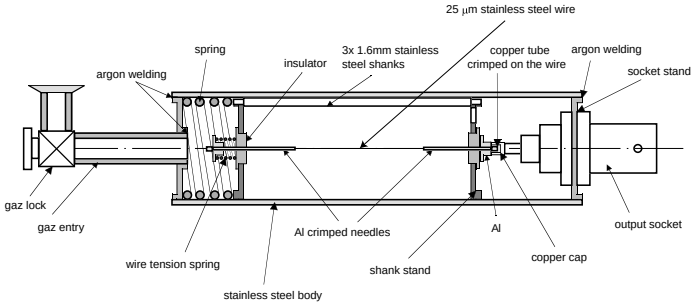


Figure 3: Mechanical structure of the ^{3}He gas proportional counter detector

iii) ***Neutron flux measurements***:

- Absolute calibration:

The integral flux is measured by activation of Nickel foils. These foils are put against the GENEPI target. The dimensions of the foils are 5 mm in radius and 0.5 mm in thickness. 6 hours of irradiation with a beam intensity of 66 μA are enough to reach saturation. The following reactions are used: ^{58}Ni(n,2n)^{57}Ni with a threshold 13 MeV, ^{58}Ni(n,np)^{57}Co with a threshold 13 MeV. These activated foils are then counted in the low background laboratory at ISN.

- ^{233}U fission detector:

The GENEPI neutron pulse, generated at time zero by the reaction T(d,n)α in the lead block centre region, gives at position <r> a neutron flux ϕ(E,t, <r>) which is measured with a detection system using the exothermic reaction ^{233}U(n,fission). The reaction rate versus time is proportional to the quantity ϕ(E,t, <r>)σ(E). Assuming the cross section σ(E) is known, the measurement of the reaction rate gives an experimental access to the quantity ϕ(E,t, <r>). The fission fragments produced in the reaction ^{233}U(n,fission) (Q = 180MeV) are collected by a silicon detector. The ^{233}U target of 200 μg/cm^2 is electro-deposited pure ^{233}U on a 200 μm thick aluminium foil. This small detection device is enclosed in a lead container. Two small charge-preamplifier are connected to the detectors inside the steel cylinder [4].

iv) ***Capture rate reaction measurement***:

A scintillator coupled with a Photomultiplier is used for sample (n,γ) reaction rate measurements. The PM is an XP1911 type from Philips [5]. It was chosen for its small dimensions (ϕ = 19 mm). Teflon has been chosen for the embase material, to avoid Hydrogen and the subsequent neutron energy degradation. The PM gain variation has been minimized with an adequate decoupling capacitance. A CeF_3 scintillator has been chosen for its quick response time (30 ns) and for its low neutron capture cross section. The detection system and the sample are embedded in a lead box, in order to have a good reproducibility of the detection geometry. Every two samples, the background has been systematically measured in order to check the stability of the PM gain. The beam intensity was adjusted to have a low dead time for each sample (0.1 evts/pulse during the first 10 μs). This detector and its lead box are shown in figure 4.

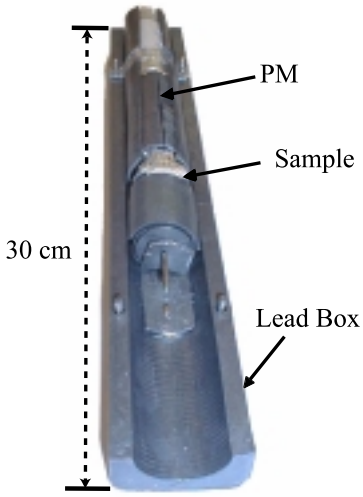


Figure 4: Picture of the PM in its lead box

III Experimental results:

The detector signals are recorded with a timing module referenced to the neutron source pulse with a 100 ns precision. For each detector, time spectra are built, giving the number of events as a function of the time of flight of the associated neutron.

Flux measurements:

The ^{233}U fission rate $\sigma_f\phi(t)$ is obtained with the silicium detector as described above. The α emission due to ^{233}U disintegration introduces a background in the fission rate time spectrum. Fortunately the energy deposition of fission products and α particles in the silicium can easily be separated allowing to build a pure fission rate time spectrum. Assuming the same efficiency for α and fission products detection and knowing the neutron production (Ni foil activation, 1.7 10^6 neutron per pulse), the fission rate $\sigma_{(n,f)}\phi(t)$ can be normalized per source neutron. In figure 5 the time spectrum exhibits a peak at 300 µs corresponding to the well-known 1.7 eV ^{233}U fission resonance.

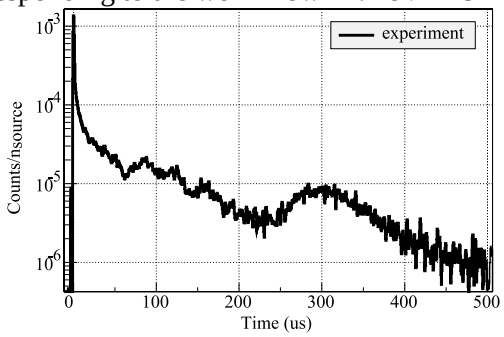


Figure 5: Timing spectra of the 233U detector: 200µg/cm2

Neutron flux monitoring:

For these measurements we used a ^3He gas detector. The figure 6 presents a typical time spectrum obtained with this counter. The (n,p) cross sections is particularly smooth in the 10^{-4} eV to 1.0 MeV energy range and elastic cross section is negligible below 100 keV. Therefore we are confident that the flux is the same in these two holes, one of them is used for capture rate measurements. The figure 6 shows the good agreement between two measurements made in symmetrical channels.

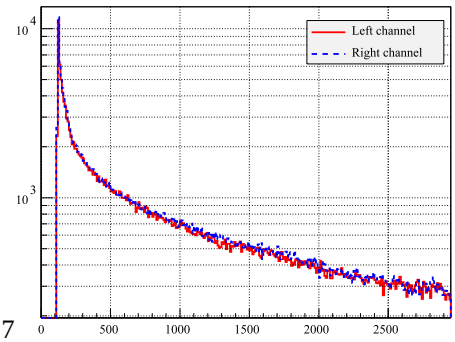


Figure 6: Time spectrum of ^3He gas detector in two measurement holes of lead block

(n,γ) experiments:

Background study:

The photomultiplier with the scintillator and the sample embedded in the lead housing, are inserted into a channel of the lead block. This detection system is very sensitive to gamma rays emitted by neutron captures in the surrounding materials. Therefore background measurements and a good understanding of its structure are necessary.

The figure 7 shows a background measurement. The general exponential dependence is due to the decrease of the neutron flux associated with the scattering process in the lead block. Superimposed structures can be seen which are due to neutron capture resonances in various nuclides. This background has been simulated taking into account all the elements contained in the detection system itself (CeF_3 scintillator, PM) and in the lead impurities, with their proportions as free parameters. Measured and simulated spectra over a 300 µs time range are shown in figure 7.

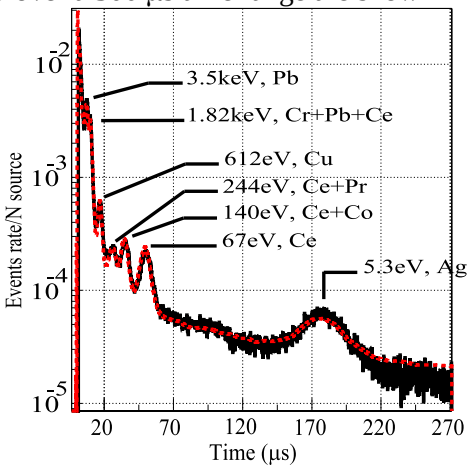


Figure 7: Experimental (full line) and simulated reconstructed (dotted line) background time spectra with energy of the resonances and identification of associated nuclides

Results for gold and thorium targets:

Figure 8 shows the normalized reaction rate for Thorium and Gold. Due to a larger cross section Gold spectrum is less affected by the background. The large structure which appears at 180 µs correspond to the 27000 barns well-known 4.9 eV resonance. In the case of Thorium a high radioactivity level is observed above 140 µs. The peak at 85 µs is due to the 21.8 eV and 23.5 eV unresolved resonances.

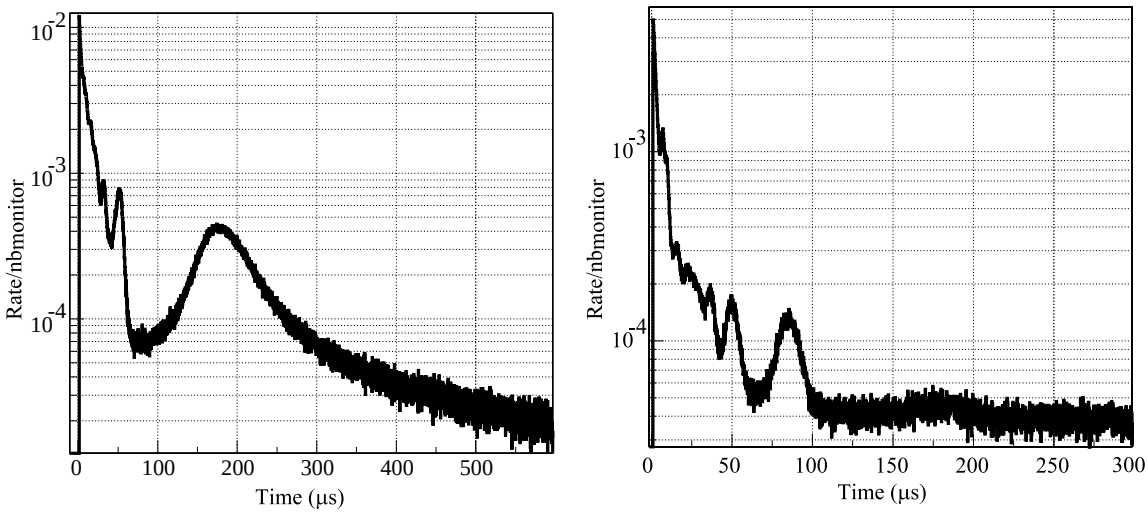


Figure 8: Capture rates for Gold (left) and Thorium (right)

IV Analysis:

Background subtraction method:

All spectra are normalized to the counting rate of the ^{3}He reference monitor. Due to the self-shielding effects in the target, the background subtraction cannot be directly made. We proceed in three steps. Both activation and natural radioactivity induce a constant counting rate over the whole time range. This constant level for background and target can be measured when the neutron flux becomes negligible i.e. for time bigger than 2ms. The first step consists of subtracting this level for each spectrum. In a second step, the measured background must be corrected as it is higher than its real contribution in the presence of the target, the γ-rays due to captures in the surrounding materials being slightly absorbed in the target. This correction is evaluated according to the density and the thickness of the target. The last step consists of taking into account the neutron flux perturbation induced by the target. This correction factor is obtained by the ratio of simulation performed with and without target.

This corrected background is subtracted from the spectrum obtained in the first step. It must be noticed that the two last corrections are second order effects.

Monte Carlo simulation:

We use the MCNP/4B code for simulations [7]. This code allows a very detailed description of the experimental set-up: detectors, generator components, lead block and concrete walls. The reaction rates $\sigma(n,\gamma)\phi(t)$ are calculated that can be directly compared with experimental data. Three different data bases have been used: ENDF/BVI, JENDL3.2 and JEF2.2.

Simulation and experimental results:

Simulation provides capture rate per source neutron as a function of time. Both the simulated and experimental time spectra are converted into energy spectra by mean of the time-energy correlation described in a previous section. The simulation to experiment ratios are calculated for each energy bin, and are presented in figures 9 to 12. We present results for 4 different targets: Au in order to validate the procedure, Tantalum, Indium the Thorium target results which are of main interest for new fuel cycle studies.

Gold:

First of all, in order to validate the procedure, we used Gold for which capture cross-section is well known.

The figure 9 shows results for 1250 µm, 500 µm and 125 µm Gold targets. A good agreement is found from 0,2 keV to 40 keV with ENDF/BVI and JEF2.2 capture cross section data bases. However, a noticeable discrepancy is observed between 2 keV to 6 keV, which remains unexplained.

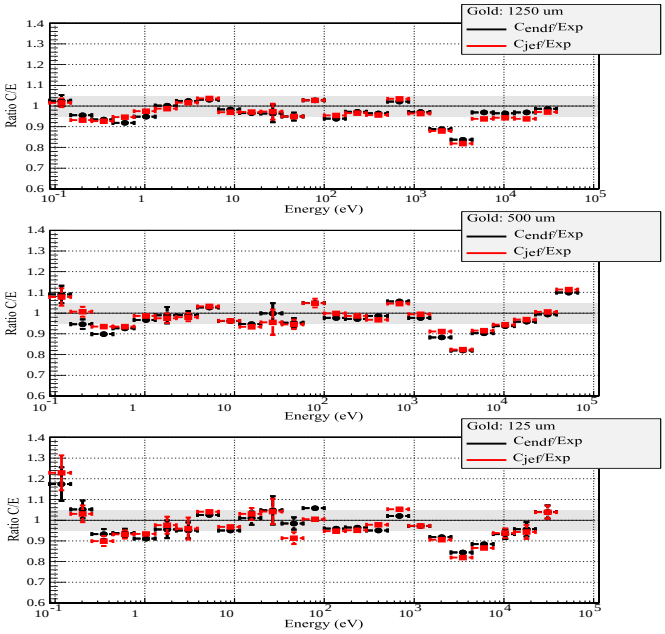


Figure 9: ENDF/BVI and JEF2.2 simulation to experiment ratio for 1250 µm, 500 µm and 125 µm Gold samples in the energy range from 0.1 eV to 40 keV. The large grey box around C/E = 1 corresponds to an uncertainty of 5%.

Tantalum:

In the case of 2000 µm, 200 µm and 100 µm Tantalum targets, the resolved resonance zone from 1eV to 200eV is correctly described. However, a deficit is observed for neutron energies lower than 1 eV for the thickest targets. A good agreement between simulation and experimental results is found with JENDL3.2 data base for $300eV<E_n<1keV$. In this energy range, ENDF/BVI and JEF2.2 data bases lead to a capture rates which are higher than the experimental one. This overestimation increases with the target thickness (see figure 10). For $2keV<E_n<20keV$ a good agreement between experiment and simulation is found for ENDF/BVI and JEF2.2 data bases. In this energy range, the use of JENDL data base produces simulated capture rates which are too low when compared to the experimental one.

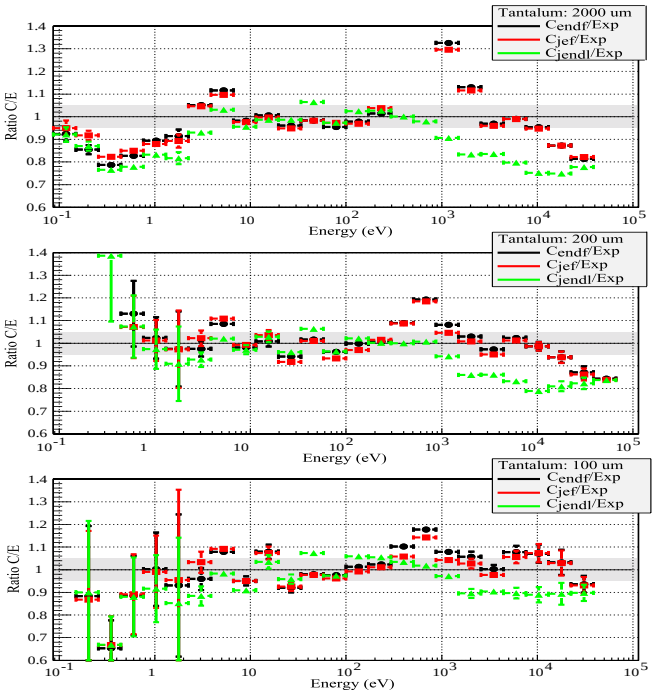


Figure 10: ENDF/BVI, JEF2.2 and JENDL3.2 simulation to experiment ratio for 2000 µm, 200 µm and 100 µm Tantalum samples in the energy range

from 0.1 eV to 40 keV

Indium:

In the case of 2000 µm, 500 µm and 300 µm Indium targets, ENDF/BVI, JEF2.2 and JENDL3.2 simulation to experimental ratio give a good agreement in the range from 0,1eV to 1keV (see figure 11). For 1keV<E_n<40keV, the use of ENDF/BVI and JENDL2.2 data bases leads to a capture rate which is higher than the experimental one for 2000 µm and 500 µm thickness targets. However, the use of JEF2.2 data base leads to a capture rate, which is lower than the experimental one for each target thickness.

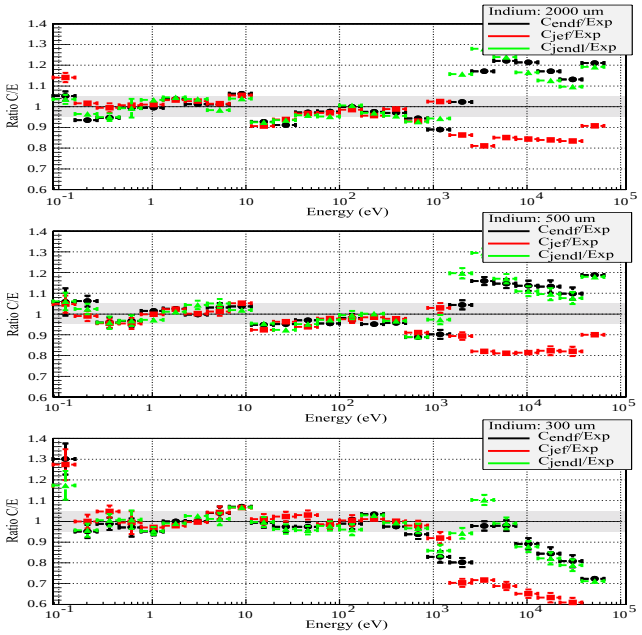
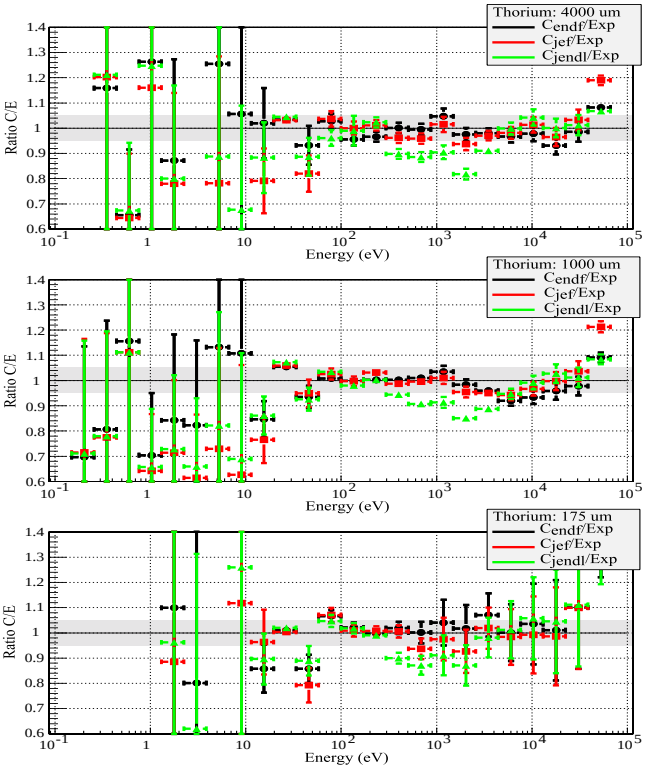


Figure 11: ENDF/BVI, JEF2.2 and JENDL3.2 simulation to experiment ratio for 2000 µm, 500 µm and 300 µm Indium samples in the energy range from 0.1 eV to 40 keV

Thorium:

In the case of 4000 µm, 1000 µm and 175 µm Thorium targets ENDF/BVI and JEF2.2 simulation to experimental ratio give a good agreement (better than 5%) in the range from 10 eV to 40 keV (see figure 12). For E_n<10eV, due to the low Thorium capture cross section and the target radioactivity, the experimental rate has high uncertainties. From 300 eV to 3keV, the use of JENDL3.2 data base leads to a capture rate which is lower than the experimental one for each target thickness. If we compare the results of ENDF/BVI or JEF2.2 simulation to the experimental ratio, in the unresolved resonances zone (upper than 100 eV), it shows the self-shielding effects in target good description by MCNP/4B code, well described for thickness up to 4000 µm which is of main interest to take into account accurately these effects in reactor fuel



rods.

Figure 12: ENDF/BVI, JEF2.2 and JENDL3.2 simulation to experiment ratio for 4000 μm, 1000 μm and 175 μm Thorium samples in the energy range from 0.1 eV to 40 keV

V Conclusion

The neutron capture cross section profiles of various targets (Gold, Tantalum, Indium and Thorium) have been measured with a slowing-down lead spectrometer in the neutron energy range from 0.1eV to 40 keV with a precision of 5%. The experimental results are compared to Monte Carlo simulations with the MCNP/4B code using ENDF/BVI, JEF2.2 and JENDL3.2 data bases. Measurements on the well-know Gold nucleus are well reproduced by simulation. The agreement for different target thicknesses validates our method, and shows that the self-shielding effect is well taken into account by the MCNP code. For Tantalum and Indium targets, a discrepancy between experiment and simulation is observed for neutron energy greater than 300 eV, in the region of the unresolved resonances. For Thorium targets, the JENDL3.2 cross section seems under evaluated by 10% in the energy range from 300 eV to 3 keV.

In conclusion, the lead spectrometer appears to be a very useful tool, allowing quick cross section validation and transmutation rates evaluation.

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