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Measurement of neutron capture and fission cross sections of ^{233}U in the resonance region

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Abstract. In the framework of studies concerning new fuel cycles and nuclear wastes incineration experimental data of the α ratio between capture and fission cross sections of ^{233}U reactions play an important role in the Th/U cycle. The safety evaluation and the detailed performance assessment for the generation IV nuclear-energy system based on ^{232}Th cycle strongly depend on this ratio. Since the current data are scarce and sometimes contradictory, new experimental studies are required. The measurement will take place at the neutron time-of-flight facility GELINA at Geel, designed to perform neutron cross section measurements with high incident neutron-energy resolution. A dedicated high efficiency fission ionization chamber (IC) as fission fragment detector and six C_6D_6 liquid scintilators sensitive to γ-rays and neutrons will be used. The method, based on the IC energy response study, allowing to distinguish between gammas originating from fission and capture, in the resonance region, will be presented.

1 Introduction

Energy production is the subject of the sustainable development debate. The production and consumption of energy are at the heart of economic development and social progress. One of the most frequently asked question is related to the treatment and storage of the waste generated by the production of nuclear energy. Scientific projects put forward several solutions, especially those of less polluting nuclear power reactors.

The need for proliferation-resistance longer fuel cycles that would result in higher burn-up efficiency and lower production of high-radiotoxicity nuclear waste has led to renewed interest in the thorium based fuel cycle. The ^{232}Th/^{233}U cycle has a significant advantage, as compared to the ^{238}U/^{239}Pu cycle used in current reactors. Thorium natural abundance exceeds that of uranium and the use of the fuel cycle based on ^{232}Th could substantially reduce the build-up of highly radioactive transuranium nuclides, especially americium and curium. The Th cycle, with its lower atomic and mass numbers, produces a reduced amount of these minor actinides.

Little used industrially, the thorium cycle has also been less explored. Therefore, for the feasibility

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study and design of innovative systems based on the use of ^{232}Th cycle, accurate and consistent neutron cross section data are required. Recent nuclear data sensitivity analysis [1,2] of the breeding of the thorium cycle, have shown that the fissile regeneration is dominated by uncertainties in the α ratio between the capture and fission cross section of ^{233}U. The available experimental data are at the present insufficient [3]-[7], and inconsistencies still exist among nuclear data libraries [8]. Therefore, new and accurate measurements for fission and neutron capture reactions of ^{233}U are required to improve the reliability of cross section data.
In this respect, the ACEN group of CENBG decided to focus its interest on these reactions. The experiments will be made at GELINA (Geel Linear Accelerator) neutron time-of-flight (TOF) facility at IRMM [9], a powerful pulsed white spectrum neutron source designed to perform neutron cross section measurements with high resolution for the incident neutron energy.

2 Present data status

So far, nuclear reactors are mainly based on the uranium cycle. ^{235}U is the only natural fissile isotope nucleus and represents 0.7% of natural uranium consisting mostly of ^{238}U (99.3%). ^{238}U, which is not fissile produces the fissile element ^{239}Pu, after neutron capture and subsequent β^- decays.
Unlike natural uranium, natural thorium does not contain any fissile material and is made up of the fertile ^{232}Th isotope. ^{232}Th is likely to produce a fissile nucleus in the same way that ^{238}U produces ^{239}Pu. After a neutron capture and two β^- decays, the fissile element ^{233}U is produced. An important property of this nucleus for the chain reaction is reflected by the breeding gain (BG), one of the most important parameter for the evaluation of the feasibility of the reactors based on ^{232}Th cycle. The BG is given by the following approximation:

$$BG = \nu - 2 * (1 + \alpha) \tag{1}$$

where ν is the fission neutron yield and α is fission to capture ratio defined by the relation:

$$\alpha = \frac{\sigma_{capture}}{\sigma_{fission}} \tag{2}$$

where $\sigma_{capture}$ is the capture cross section and $\sigma_{fission}$ is the fission cross section.
A positive BG means that the nuclear reactor generates more fissile material in fuel than it consumes. The design and realization of the nuclear power stations based on Th cycle require accurate knowledge of α ratio. In one significant respect, the ^{232}Th/^{233}U is more flexible than ^{238}U/^{233}Pa, because of the positive BG for the whole neutron spectrum, from thermal to high energy range. In addition to the interest from advanced technologies, neutron induced cross sections at low energy can provide significant knowledge on nuclear properties, which is a fundamental input in nuclear structure and reaction models, as for example is the level density at neutron binding energy, that can be directly obtained from high resolution neutron resonance spectroscopy.
The measurement of the neutron capture cross section of ^{233}U is complicated by the low energy neutron-induced fission that will also give γ-rays. However, the most critical region is the resolved resonances in connection with the epithermal spectrum proposed for some Molten Salt Reactors [10], CANDU and PWR [11] based on ^{232}Th cycle, typically from 1 eV to 100 eV neutron-energy range. We can observe variations of several orders of magnitude for the ^{233}U(n,f) and ^{233}U(n,γ) cross sections. For example Fig. 1 illustrates this phenomenon. Fig. 1 represents the capture and the fission cross sections of ^{233}U taken from the ENDF/BVII evaluated data library from thermal to fast neutron energy.
Until now, only two measurements are available in the resonance region, the first one performed by Brooks et al. [4] up to 11 eV with poor energy resolution, and the second performed by Weston et al. [6] up to 2 keV using the fission tagging technique. Most of the previous measurements of the neutron-induced capture and the fission cross section of the ^{233}U in the thermal and resonance region were done before 1975. The available data for this ratio present a dispersion of more than 25% due to the high uncertainties of the capture cross section [1,2]. For these experimental data little information

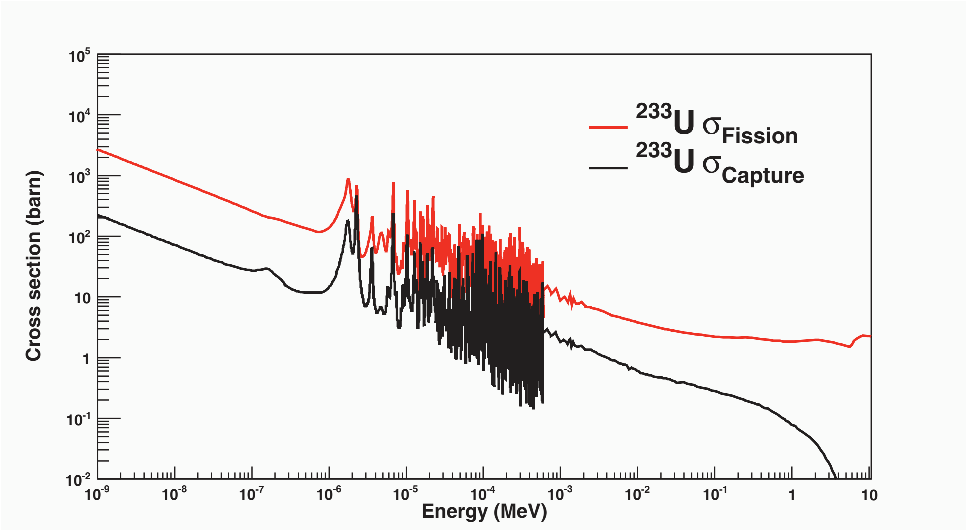

Fig. 1. Evaluated neutron-induced capture and the fission cross section of ^{233}U from the ENDF/BVII evaluated data library, for the incident neutron energy range from thermal to 10 MeV.

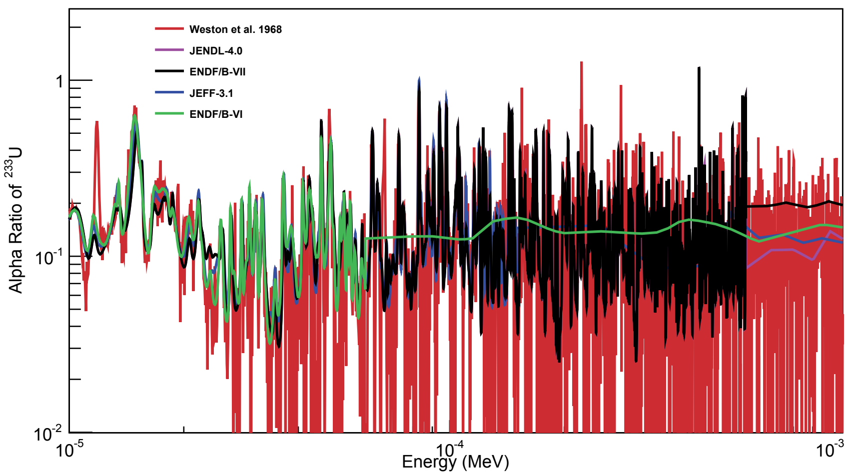

Fig. 2. α ratio of ^{233}U, comparison of experimental results and evaluated libraries in the resolved resonances region.

is available on the measurement methods, types of detectors used, the neutron source, uncertainties as well as the energy ranges [3]-[7]. Fig. 2 shows the wide disparities between the different evaluated data and the only experimental data for the α ratio of the ^{233}U obtained in the resonance region. This confirms that the α ratio of the ^{233}U is poorly known and requires further experiments to validate the existing data, constrain the models and especially to reduce the uncertainties. In this context, recent measurements of capture and fission cross sections of ^{233}U have been proposed.

3 Method and Experimental Set-up

The neutron induced reaction cross section measurements such as capture σ_γ and fission σ_f present difficult experimental problems compared to the total cross section measurements that are also the most accurate type. Capture and fission cross sections require specific geometrical configuration, measurements at an energy for which the reaction yield (the count rate) is well known and additional flux measurements. When a neutron is absorbed in the sample of ^{233}U, it is either captured or induces fission. The α ratio is given by the following equation:

$$\alpha = \frac{\sigma_{capture}}{\sigma_{fission}} = \frac{\frac{N_\gamma}{\varepsilon_{C_6D_6} * \Phi(E)}}{\frac{N_f}{\varepsilon_{IC} * \Phi(E)}} \tag{3}$$

where N_γ is the number of capture events detected, N_f the number of fission events detected, $\varepsilon_{C_6D_6}$ the efficiency of the C_6D_6 detectors (for capture events detection), ε_{IC} the efficiency of the IC (for Fissin Fragments (FF) events detection), and Φ(E) is the neutron flux.

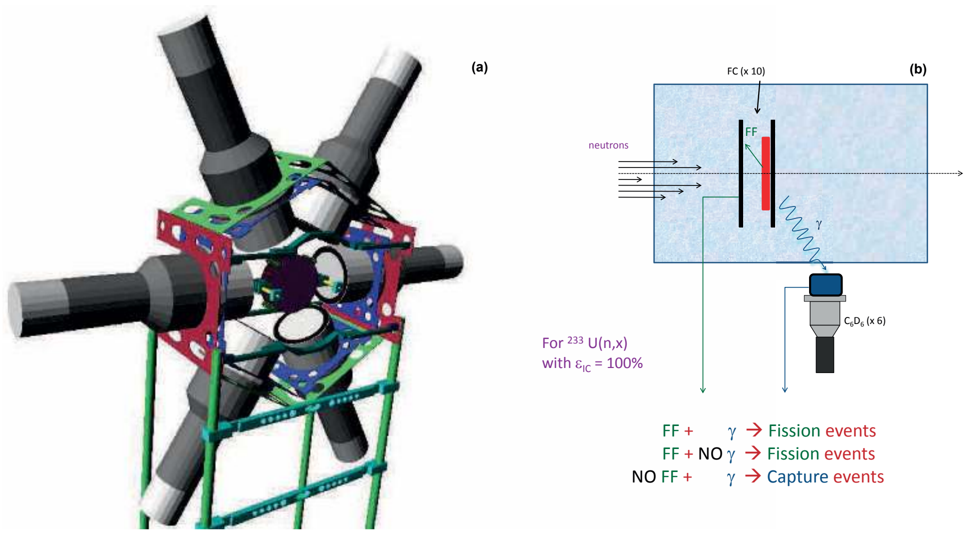

Fig. 3. (a) Experimental set-up: six C_6D_6 detectors around the IC containing ten ^{233}U targets inside. (b) Capture to fission discrimination diagram.

Since neither neutrons nor γ-rays are charged particles their detection efficiency is low and it is difficult to discriminate between them especially when they are both present. Neutron induced capture cross section measurements in the resonance region are mostly done using γ-ray detectors with good timing characteristics and with low sensitivity to neutrons such as C_6D_6 liquid scintillators [12, 13]. Since the pulse widths for neutrons are wider in time than for γ-rays thus doing that the neutrons emitted after fission to be discriminated using the difference in detected pulse shape. The energy resolution of this type of detectors does not allow the selection of the individual primary γ-rays because they interact with the detector via the Compton scattering process. The purpose of this detector is to accurately determine the number of (n,γ) events produced in a target by counting the γ-ray cascades using the weighting function method [14]. For the detection of the FF produced in ^{233}U(n,f) reaction a multianode-multicathode IC filled with argon methane gas is used, Fig. 3(a). The FF loses energy due to Coulomb interactions with the atoms and molecules of the gas, and the number of the electron-ion pairs created is proportional to the energy of the fragment. This IC has the advantage to be used both as detector and target. As the fission process is followed by the γ-rays emission (about 2 times more per event than the capture process) and is 5 to 10 times more likely, it is essential to identify these events precisely for discriminating them from the γ-rays from the capture process. The discrimination between these γ-rays is the most challenging issue in this experiment. The γ-rays detected in coincidence with the FF correspond to a fission event. The capture event is identified by putting a VETO on FF. A representative diagram of the detection events is shown in Fig. 3(b). To achieve this performance the efficiency of the IC has to be known very precisely. If a fission event is not detected in IC, one of the six C_6D_6 scintillators could detect a prompt γ-ray and the fission event will be interpreted as a capture event. It is then necessary to have a very high efficiency events detection system for this type of measurements. A high efficiency is also difficult to be obtained because of the high α activity of ^{233}U which produces pile-up of the events in the IC.

Another obstacle in this measurement is the neutron sensitivity. Indeed, a neutron can be scattered by the experimental set-up and be captured by the C_6D_6 detector material. This event is interpreted as a capture event but in fact is related to the background. To reduce the interactions of the neutron beam with the detector system and to minimize the production of background by the IC itself, the components of the experimental set-up were made of as less as possible aluminium material.

The experimental set-up for this experiment has been designed and built at CENBG and is presented in Fig. 3(a). The detection set-up for the measurements of the neutron-induced fission cross section is based on the IC, an assembly of 10 parallel-plates fission chambers with 5 mm spacing between electrodes and operating with 90% Ag +10% CH_4 at 1.0 atm. The ^{233}U samples required for this experiment are prepared at IRMM by electro-deposition technique on 50 μm thick high purity Al foil with a diameter of 10 cm stretched on aluminium ring. Obviously the material used for the backing should be as thin as reasonable possible and made out of material with low neutron interaction probability. To maximize the amount of the ^{233}U material without increasing the thickness of the layer, the diameter of the uranium deposit is 6 cm. The plates are electrically isolated, having a separate signal connector for each of them, and assembled along the direction of the neutron beam. The anodes of the IC are biased to 100 V high voltage with a power supply system. The cathode signals are connected to the input of fast charge amplifiers.

Several parameters determine the behaviour of the IC, as the gas pressure or the high voltage between the electrodes. The impact of these parameters has been studied to find the ideal operating point: a good energy separation between α/ FF and a good timing resolution.

4 First experiment

Preliminary measurements were performed at the CENBG. As mentioned above, it is imperative to know precisely the efficiency of the IC. We therefore made different tests using a ^{252}Cf source which is a spontaneously fissioning isotope. This source is also necessary to set the electronic modules that allow the pulse shape discrimination between the γ-rays and neutrons in the C_6D_6 detectors.

Fig. 4(a) shows the spectrum of fission events obtained with the IC. We can observe a very good separation in energy between the α particles and FF obtained with the ^{252}Cf source. This measurement has to be done also with the ^{233}U source because the IC response is related to the activity, thickness, shape and homogeneity of the deposit. The detection efficiency for FF was measured using the ^{252}Cf source and was found to be (97 ± 0.5)%. This result was found by coincidence between the neutrons detected in the C_6D_6 and FF detected in the IC. The efficiency of IC is the ratio between the number of the coincidence-neutrons and the number of the single-neutrons. To distinguish between γ-rays and neutrons detected in the C_6D_6 scintillator we used the pulse shape discrimination (PSD) method. Fig. 4(b) shows the selection of single-neutrons in one of the C_6D_6 detector. To validate this result additional measurements need to be done with the ^{233}U source in a thermal neutron flux.

The first test of the experimental set-up was done at the TOF facility pulsed neutron source GELINA at IRMM. The GELINA neutron source is based on a linear electron accelerator [9]. A typical beam operation mode uses 100 MeV average energy, 10 ns pulse length, and 800 Hz repetition rate. The accelerated electrons produce Bremsstrahlung in a uranium target which produces neutrons by photonuclear reactions. The neutron-energy distribution emitted by the target ranges from subthermal to about 20 MeV, with a peak at 1-2 MeV. To have a significant number of neutrons at energy below 100

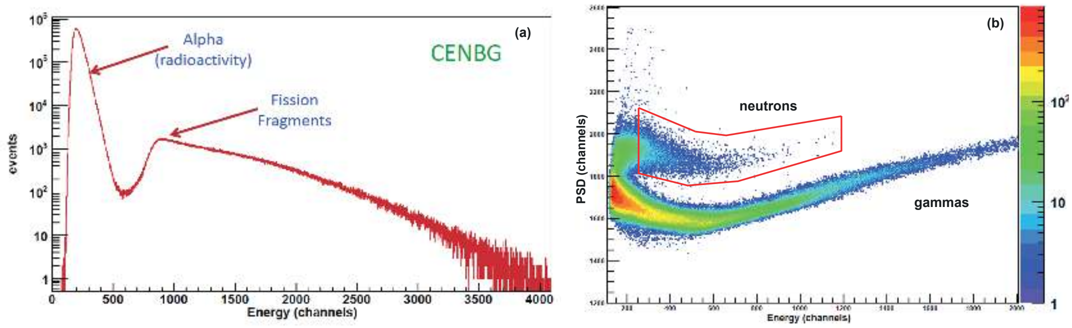

Fig. 4. (a) α and FF energy discrimination obtained with the ^{252}Cf source in the IC at CENBG. (b) neutrons cut selection in one of the C_6D_6 detectors.

keV, a hydrogen-rich moderator is added. Neutron beams are selected for the experimental neutron flight paths, located at different distances starting from 10 m up to 400 m. The determination of the neutron energy is made by the TOF method which consists in measuring the time interval between the production of neutrons burst and detection of an event. The measurement was done in a reduced geometry of two C_6D_6 detectors and two ^{233}U targets at the 10 m flight path. This flight lenght is needed for the required high quality data in the resonance region. Fig. 5 shows the TOF accordeon spectrum

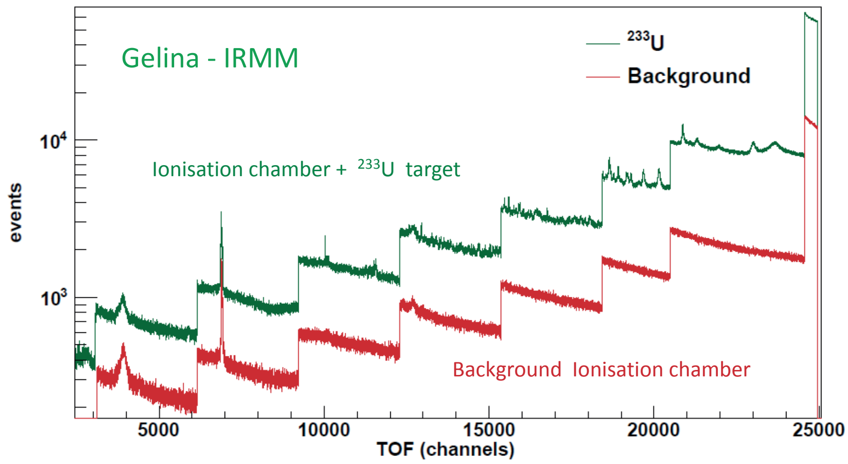

Fig. 5. TOF spectra of the number of fission events detected by the multi-plate IC with and without ^{233}U target obtained at Gelina facility.

obtained with the empty IC which is the background spectra, and with ^{233}U target. The multi-plate IC showed a very high stability with respect to the gamma flash. The good performance of IC is reflected by the fission spectrum as a function of neutron energy. One can see some of well known resonances of ^{233}U and the characteristic background noise of the device without uranium (no parasite resonances). In this experiment we also made measurements in coincidence with the two C_6D_6, observing the same fission resonances of ^{233}U.

In the near future we will perform the measurement in a complete configuration with ten ^{233}U targets and six C_6D_6 detectors. At the same time, we will determine the efficiency of the IC in real conditions in a dedicated experiment.

5 Conclusions

An experimental set-up (CENBG) has been developed for the determination of the α ratio of ^{233}U in the resonance region. Preliminary studies without the neutron beam for testing the measurement conditions have been performed. Measurements showed a good response of the IC and we have obtained good results for the discrimination between α particles and FF using a ^{252}Cf. The first test of the experimental set-up was made at GELINA-IRMM in a reduced geometry and the validation of the material, geometry and reduced background has been done.

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