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Measurement of the ^{238}U Radiative Capture Cross Section with C_6D_6 at the CERN n_TOF Facility

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We present the preliminary results of the ^{238}U(n,γ) reaction cross section measurement, performed on April 2012 at the CERN n_TOF facility using C_6D_6 scintillation detectors over an energy range from thermal to 1 MeV. The goal of this measurement, which is part of a larger proposal, is to reach an uncertainty of 2% in the cross section. The experimental set-up and the methods used to obtain this result are described.

I. INTRODUCTION

The need to reduce energy sources based on fossil fuels led to a significant effort towards improved reactor technologies. The new reactors concepts require accuracy and precision that still challenge the present knowledge of nuclear data [1, 2]. In this context, the measurement of the ^{238}U radiative capture cross section is of high priority and is part of the NEA High Priority Request LIST [3], a compilation of the most relevant nuclear data requirements. In fact, though many measurements are available for the ^{238}U(n,γ) reaction cross section (consult the EXFOR database [4]), inconsistencies are still present both in the low energy regime and in the unresolved resonance region. This uncertainty concerns fast as well as thermal reactor concepts and contributes to the uncertainty of the Pu inventory in spent fuel elements. Therefore a proposal of joint measurements at the n_TOF facility at CERN (Switzerland) and at the EC-JRC-IRMM facility GELINA (Belgium) [5] was launched to reach an uncertainty in the cross section below 2% throughout an energy range from thermal to hundreds of keV [6].

II. EXPERIMENTAL METHOD

The measurement was performed at the neutron time of flight facility n_TOF at CERN, which takes advantage of the very intense 20 GeV/c proton bunches from the CERN Proton Synchrotron (PS) impinging on a cylindrical lead target. Thanks to the spallation mechanism the n_TOF experiment can count on an extremely high instantaneous neutron flux at the experimental area. The very long flight path of about 185 m provides an excellent energy resolution, which permits to resolve closely spaced neutron resonances. These characteristics, together with a low repetition rate, allow to perform very accurate (n,γ) cross section measurements in an energy range from thermal to about 1 MeV. During the entire measurement the flux at the sample position is kept under control with a monitor based on the standard reaction ^{6}Li(n, α)^{3}H [7].

The measurement was carried out using two C_6D_6 scintillators, placed one head on the other at 90° with respect to the beam, 9 mm away from the sample: one commercial BICRON and one custom made (*Forschungszentrums Karlsruhe*−FZK) [8]. Both the detectors and the geometry are optimized to have a very low sensitivity to γ-rays induced by scattered neutrons. The effective neutron flight path of 184.21 ± 0.01 m was determined by means of the well-known s-wave resonances of Au [9].

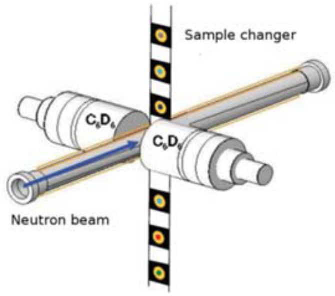

FIG. 1. Experimental set-up used.

All samples were rectangles 53×30 mm, similar to the ^{238}U sample, a metal plate 0.235 mm in thickness with an isotopic enrichment of 99.99 %. This sample was protected by a cover of 20 μm aluminum and a 25 μm Kapton foil. Additional samples of Pb and C were used for measuring the background due sample-scattered neutrons. These background runs and the determination of the contributions from the natural radioactivity and the radioactivity of the sample were complemented by a series of runs with black neutron filters to determine the time-dependence of the background. The careful evaluation of al background contributions is crucial to achieve the desired 2% accuracy in the final results.

Furthermore, two ^{197}Au samples, 50 and 300 μm in thickness, and a 300 μm thick Ag sample, were used for measuring the normalization factors discussed below.

III. DATA REDUCTION

Capture events are recorded by means of the total energy detection technique, which requires proportionality between the γ-ray efficiency and the total radiative energy released in a capture event. This proportionality is achieved with the Pulse Height Weighting Technique (PHWT) [11]. To calculate the weighting functions, the precise knowledge of the detector response as a function of energy is required, which is obtained from very detailed Monte Carlo simulations using Geant4 [13] with a complete description of the experimental setup. For the 300 μm thick Au sample, as well as for the ^{238}U and Ag samples, we distinguished two cases in the calculation of the weighting functions. The γ-ray emission within the sample was simulated in one case with a uniform distribution of the γ-ray initial position, while in the other other

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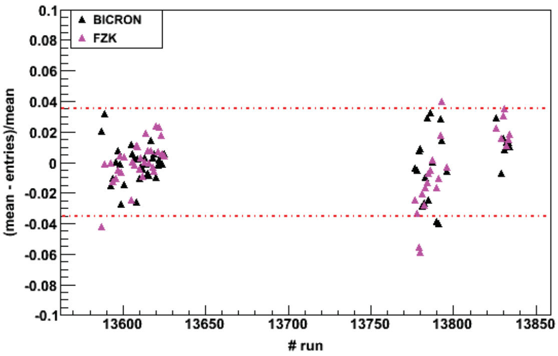

FIG. 2. U sample: percentage deviation of each run entries (normalized at the number of protons) from the mean value for both detectors. Selecting only those runs for which this deviation is below the 3.5% we reject two runs with the BICRON and four with the FZK detector.

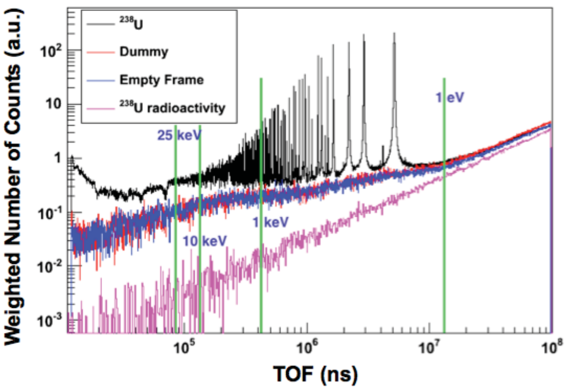

FIG. 3. Weighted counts versus neutron time-of-flight for ^{238}U (black), background due both to experimental hall and ^{238}U cover foils (red), background due only to experimental hall (blue) and ^{238}U natural radioactivity (magenta).

case an exponential γ-attenuation was assumed. The first case is used for the resonance shape analysis (RSA) and the analysis of the unresolved resonance region, while the second one is applied for the extraction of the normalization factor, since it is only at the saturated resonance energies that the exponential attenuation of the neutron beam due to the self absorption is relevant.

In order to be as precise as possible, an accurate study of the calibrations between the *flash*-ADC channels and the deposited energy was performed. Six calibration lines were extracted from six measurement of three different calibration sources: ^{137}Cs, ^{88}Y and Am-Be.

The stability of the apparatus was also tested, both for the flux and the gain of the scintillators. Concerning the neutron flux stability, we investigated the ratios between the hits registered by the silicon monitors and the number of protons derived from the pick-up signal from the CERN-PS. We selected only runs where ratios deviated by no more than 3.5%. Moreover, the stability of the two C_6D_6 detectors was investigated by a gate on the time-of-flight (TOF)-region corresponding to the first saturated resonances in ^{238}U and ^{197}Au (measured with the 300 μm sample). For the gold sample, where the number of events per run deviates from the mean by only 0.6% for the FZK and by 1% for the BICRON detector, all runs were included in the analysis.

For the ^{238}U sample we found instead that the number of entries deviates by about 6% from the mean value, a percentage too big to be acceptable for our analysis. We then put a gate on the number of entries registered by the detectors, choosing only those runs which percentage deviation is less than 3.5% (Fig. 2). After this cut the distribution of the percentage deviation is studied, and it results that more than the 90% of the runs deviates from the mean less than 2%. By these cuts we reject in total 1.5% of the data from the BICRON and 5% for the FZK detector.

In Fig. 3 the time of flight spectra are shown for the ^{238}U sample and the main backgrounds. As can be seen from the figure the uranium radioactivity dominates for long flight times, i.e. at low neutron energies. Concerning the background, Fig. 3 shows that the contribution arising from the cover foils of the ^{238}U sample is negligible with respect to the background coming from the experimental hall.

All the results shown hereafter are background and radioactivity subtracted.

IV. PRELIMINARY RESULTS

The experimental yield determined in a capture measurement, which is by definition tied-up with the cross section, could be expressed as function of the incident neutron energy as

$$Y_c(E_n) = N \, \frac{C_w(E_n)}{\varphi_n(E_n)}. \tag{1}$$

Here, N indicates the normalization factor which includes both the solid angle of the detection system and the effective area of the neutron fluence φ_n intercepted by the sample. C_w denotes the counts detected by the scintillation detectors after application of the weighting functions described above. The yields are calculated for both detectors separately and are afterwards combined for the determination of the final cross section.

TABLE I. Normalization factors calculated separately for the BICRON and FZK detectors.

Sample	N_{BICRON}	N_{FZK}
$^{\text{nat}}$Ag	0.915 ± 0.003	1.090 ± 0.003
^{197}Au (50 μm)	0.913 ± 0.003	1.110 ± 0.003
^{197}Au (300 μm)	0.911 ± 0.002	1.113 ± 0.002
^{238}U	0.862 ± 0.001	1.027 ± 0.001

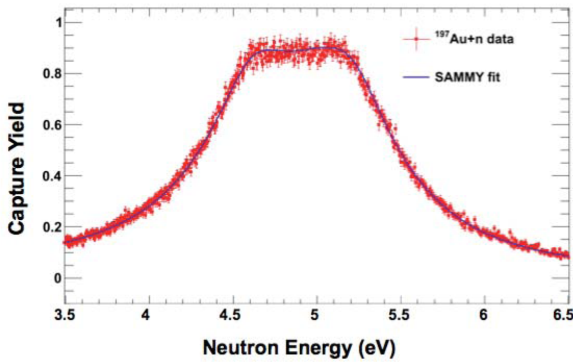

FIG. 4. First s-wave resonance in gold measured with the 300 μm thick sample. The fit with the R-matrix code SAMMY is shown in blue.

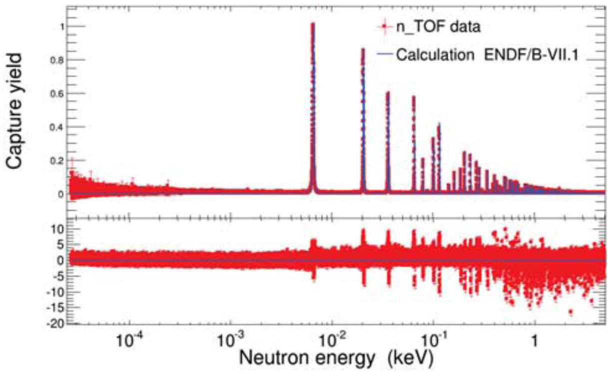

FIG. 5. Experimental yield for the ^{238}U(n,γ) reaction compared with the calculation based on ENDF/B-VII.1 [15] resonance parameters (blue line). In the lower panel residuals are shown.

As pointed out before, an accurate study of the normalization factor N was performed, using the the silver sample, the two gold samples, and the uranium sample itself, which, thanks to the very well known first saturated resonance, can be self-normalized. The fits were done with the R-matrix code SAMMY [12] as shown in Fig. 4 for the first gold resonance at 4.9 eV. As a further validation, the normalization factor was also determined by fits of other well-known gold resonances between 10 and 50 eV. All these values agree within 2%.

The comparison of the normalization factors in Table I shows that the values obtained with the Ag and Au samples are consistent within 2%, while the normalization factors for ^{238}U deviate by $6-7$ % with respect the other values. As mentioned before, the ^{238}U can be self-normalized, so this discrepancy does not affect the precision of the measurement. We studied also the impact of the dead time on the normalization factors and found negligibly small.

The neutron flux φ_n at the sample position as a function of neutrons energy E_n is determined by a combination of several flux monitors based on the three standard reactions ^{6}Li(n, α)^{3}H, ^{10}B(n, α)^{7}Li and ^{235}U(n, f) as described in Ref.[14].

In Fig. 5 the experimental yield for the ^{238}U(n,γ) reaction is shown together with the calculation based on resonance parameters of the evaluated library ENDF/B-VII, in an energy range from thermal to 5 keV.

V. CONCLUSIONS

The innovative features of the neutron time-of-flight facility n_TOF at CERN have been used for an accurate measurement of the ^{238}U(n,γ) cross section. Great care has been devoted for characterizing all critical aspects of the measurement, in particular with respect to the determination of the relevant background components and of the flux normalization factors. The result of this measurement will be combined with n_TOF data obtained with a 4π BaF$_2$ total absorption calorimeter and with the outcome of an independent experiment at the EC-JRC-IRMM facility GELINA.

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