EPJ Web of Conferences **146**, 11054 (2017) DOI: 10.1051/epjconf/201714611054
ND2016

The ^{236}U neutron capture cross-section measured at the n_TOF CERN facility

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Abstract. The ^{236}U isotope plays an important role in nuclear systems, both for future and currently operating ones. The actual knowledge of the capture reaction of this isotope is satisfactory in the thermal region, but it is considered insufficient for Fast Reactor and ADS applications. For this reason the ^{236}U(n, γ) reaction cross-section has been measured for the first time in the whole energy region from thermal energy up to 1 MeV at the n_TOF facility with two different detection systems: an array of C_6D_6 detectors, employing the total energy deposited method, and a 4π total absorption calorimeter (TAC), made of 40 BaF_2 crystals. The two n_TOF data sets agree with each other within the statistical uncertainty in the Resolved Resonance Region up to 800 eV, while sizable differences (up to $\simeq$ 20%) are found relative to the current evaluated data libraries. Moreover two new resonances have been found in the n_TOF data. In the Unresolved Resonance Region up to 200 keV, the n_TOF results show a reasonable agreement with previous measurements and evaluated data.

1. Introduction

^{236}U plays an important role in nuclear systems, both innovative or already in use. In the current reactors based on U/Pu it is important in the neutron balance in the core and in the equilibrium fuel composition. Regarding the future reactors based on the Th/U cycle, ^{236}U plays the same role as ^{242}Pu in the traditional fuel cycle, therefore its contribution to the fraction of absorbed neutrons is relevant. The actual knowledge of the capture cross-section of ^{236}U is considered satisfactory in the thermal region, but insufficient for Fast Reactors and ADS applications; in these cases the target accuracy is 10% [1], which is therefore the aim of the present measurement.

Major data libraries are based on a few experimental measurements characterized by poor accuracy and resolution and covering incomplete energy ranges. Apart for the low-energy 1/v region, where the major evaluations like JENDL-4.0, ENDF/B-VII.1 and JEFF-3.2 show a smooth trend and are in agreement between each other within a few percent, discrepancies are observed between evaluated data and experimental results, in particular in the Resolved Resonance Region (RRR), which are well above the required accuracy. Moreover, the RRR is limited to 1.5 keV in most of the libraries, while it extends up to 4.0 keV in JENDL-4.0. In the Unresolved Resonance Region (URR), evaluated cross-section below 200 keV are consistent to each other, although experimental data show some discrepancies, while above 200 keV both evaluations and experimental data show large variations.

The present situation is therefore unacceptable for future nuclear applications. This situation motivated a new measurement of the capture cross-section of ^{236}U at the CERN n_TOF facility [2]. To reduce systematic uncertainties, the measurement was carried out with two independent detection techniques based on a Total Absorption Calorimeter [3] and a pair of C_6D_6 detectors [4].

2. The experimental setup

The C_6D_6 detectors, characterized by a low neutron sensitivity and γ-ray effciency, are mounted at 135° with respect to the sample; they rely on the use of the Pulse Height Weigting Technique [5] which requires that at most one γ-ray per capture event is detected. The second detection system (TAC), made of 40 BaF_2 crystals and characterized by high geometric and intrinsic effciciency, allows detection and reconstruction of the full γ-ray cascade of the capture event. However, due to its large sensitivity to neutrons and to the γ-flash, it was not possible to collect data above a few tens of keV.

The uranium sample consists of a high purity pressed disk pellet 99.85% enriched in $^{236}U_3O_8$ with a total mass of 399 mg. Known contaminants are 0.05% of ^{235}U and 0.1% of ^{238}U. The pressed pellet is placed inside a disk shaped capsule made of high purity and normal aluminum. A capsule without sample inside, called "dummy", was built with the same dimensions of the aluminum capsule. The sample was inserted between two thin kapton foils and this assembly was glued on a carbon fiber frame mounted on the remotely-controlled sample exchanger together with other samples used for the determination of the background: an "empty" sample, made of just the kapton foils, the dummy sample, a ^{nat}Pb sample for in-beam γ-rays and a natural gold sample used for normalization purposes. All samples were made with the same diameter as the U_3O_8 pellet.

3. Results

3.1. The resolved resonance region

The background-subtracted capture yield measured with the C_6D_6 setup is shown in Fig. 1. The resolved resonance region was analyzed by means of the R-matrix code SAMMY. The yield has been parametrized via the Reich-Moore approximation; experimental effects as Doppler broadening, sample scattering, isotopic correction for contaminants, self-shielding and n_TOF resolution function are properly taken into account within the SAMMY code.

In the SAMMY fit a residual constant background was considered that was probably due to an underestimated contribution of the aluminum capsule. However, in the Resolved Resonanance Region, this component is only $\simeq$ 0.04% in the extracted capture yield.

In the fitting procedure for the Resonance Shape Analysis (RSA), the resonance energy (E_R) and both partial widths (Γ_γ and Γ_n) were allowed to vary. While the resulting ^{236}U resonance parameters are not very accurate, this procedure provides an accurate value for the capture kernel, $g\Gamma_\gamma\Gamma_n/\Gamma$. As a starting point for the SAMMY fits, the resonance parameters from the JENDL-4.0 data library

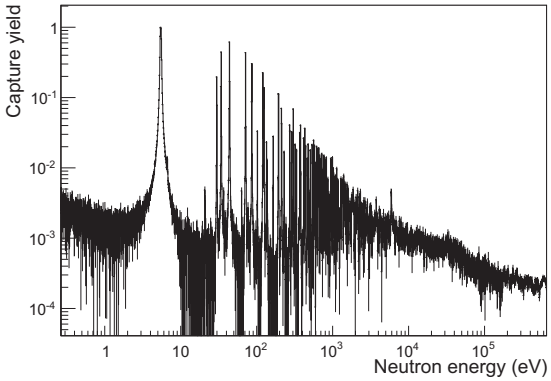

Figure 1. Capture yield obtained after normalization and background subtraction. The n_TOF data shown here are collected with C_6D_6 detection system.

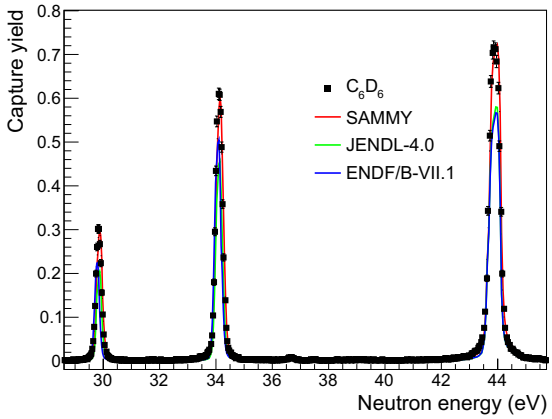

Figure 2. Comparison between C_6D_6 data fitted by SAMMY and major data libraries.

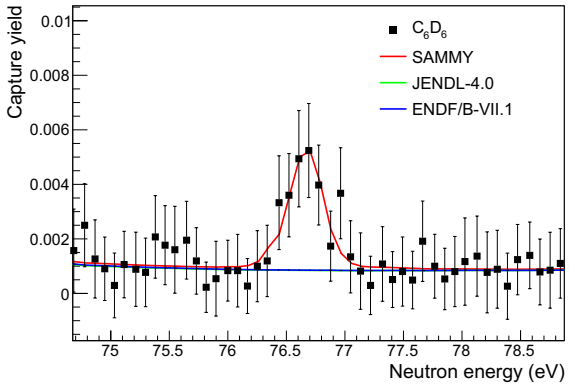

Figure 3. Structure at 76.7 eV in the C_6D_6 data compared with major data libraries. The resulting SAMMY fit is also shown.

were taken for the C_6D_6 and from ENDF/B-VII.1 for the TAC analysis.

Although structures are present, the lower statistics of C_6D_6 comparated to the TAC data does not allow to analyze resonance above 800 eV with a statistical uncertainty on the capture kernel below 40%. An example of the fitted yield of C_6D_6 data is shown in Fig. 2.

Two structures at 76.7 and 362.9 eV not present in major data libraries have been found in the C_6D_6 data set, as shown in Figs. 3 and 4, and were confirmed by the TAC measurement. A possible candidate is a ^{237}Np contamination in the sample as a result of the capture

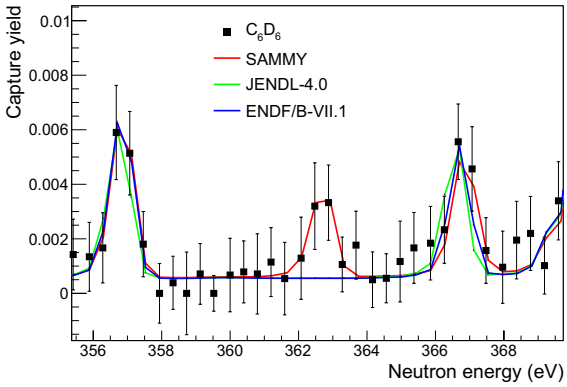

Figure 4. Structure at 362.9 eV of C_6D_6 data compared with major data libraries. The resulting SAMMY fit is also shown.

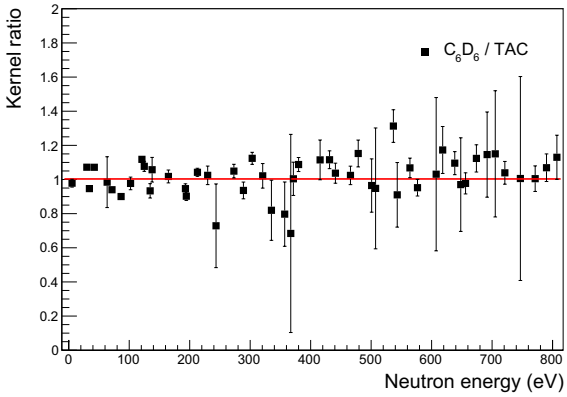

Figure 5. Comparison between the capture kernels determined from the C_6D_6 and TAC data.

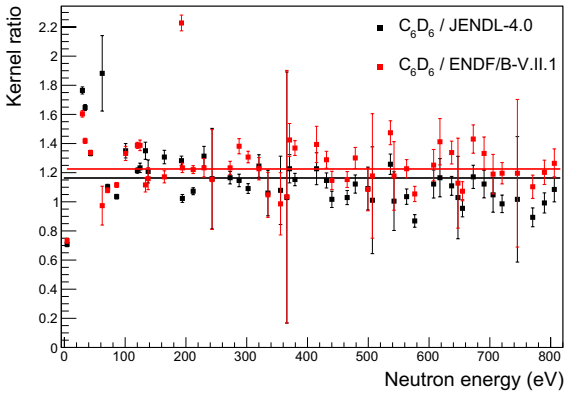

Figure 6. Comparison between capture kernels obtained from the C_6D_6 measurement with the corresponding values from data libraries.

reaction on ^{236}U(n, γ)^{237}U followed by β-decay. However, since there is no evidence of other, stronger resonances of ^{237}Np in the analyzed energy range, these resonances could be attributed to ^{236}U.

The capture kernel is sensitive to systematic effects related to the experimental setup, in particular to the neutron sensitivity. In the present analysis, these effects have been correctly taken into account, as demonstrated by Fig. 5, where the ratio between the capture kernels calculated from the resonance parameters obtained from C_6D_6 and from the TAC data is shown. The good agreement between the two datasets and the resulting average difference of less than 1% confirms that the

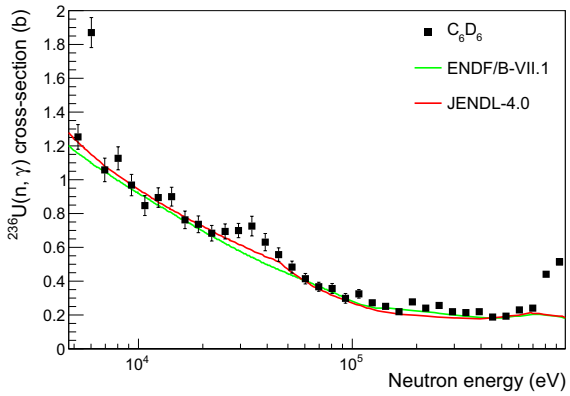

Figure 7. Comparison between C_6D_6 data and major data libraries in the URR.

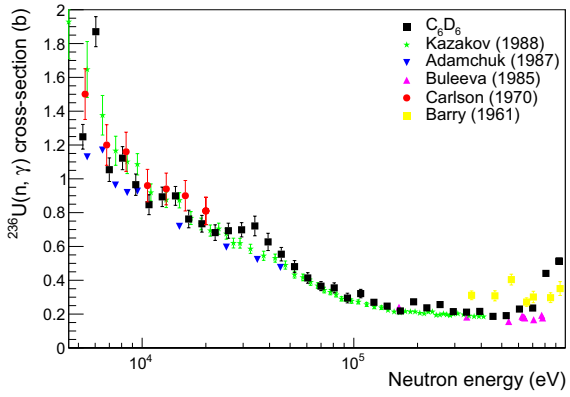

Figure 8. Comparison between C_6D_6 data and previous results in the URR.

capture yield in both independent analyses was consistent and reliable.

A comparison of the capture kernels measured at n_TOF with the ones calculated from data libraries (see Fig. 6) shows a deviation of 16% relative to JENDL-4.0 and of 23% relative to ENDF/B-V.II.1. Between 800 eV and 1.5 keV, the resonance analysis has been performed only on the TAC data. Structures above 1.5 keV are present in both data sets, confirming the structures in the evaluated cross section of JENDL-4.0. However, in this case the low statistics makes it difficult to perform a resonance analysis above 1.5 keV neutron energy.

3.2. The unresolved resonance region

Regarding the unresolved resonance region, a comparison between the C_6D_6 average cross-section data and major data libraries shown in Fig. 7 reveals that in this energy range the present results agree to better than 5% with major evaluations up to 700-800 keV. Due to the prompt γ-flash and inelastic reactions, data above this energy range are not very reliable.

The two structures in the data at 5.9 and 34.7 keV are not due to the ^{236}U(n, γ) reaction, but are consistent with the ^{27}Al(n, γ) reactions in the capsule, which are affected by a sizable uncertainty. This result and the constant background found in the RRR, indicate that the contribution of the aluminum capsule to the total capture yield is underestimated.

A comparison with previous results shown in Fig. 8, indicates that the present data, apart from the discrepancies due to the aluminum structures, agree better than 7% with most of the previous measurements up to 700–800 keV.

As a final remark, it should be considered that the present data, affected by a systematic uncertainty around 7%, are certainly the most accurate obtained so far in this energy region that is particularly important for the development of fast reactors.

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