EPJ Web of Conferences **284**, 01027 (2023) https://doi.org/10.1051/epjconf/202328401027
ND2022

^{233}U(n,γ) measurements at LANSCE

(LA-UR-22-29638)

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Abstract. Uranium-233 plays an important role in the Th-U fuel cycle, which has been proposed as an alternative to the U-Pu fuel cycle due to its reduced amount of transuranium elements. The available experimental ^{233}U(n,γ) cross section data in the literature are scarce, [1–3]. In 2008, the ^{233}U(n,γ) cross section was investigated at LANL using the DANCE detector combined with a PPAC, however the statistics in the keV regime were inadequate for a reliable extraction of the cross section at 100 keV. An accurate measurement of the ^{233}U(n,γ) cross section is required by the NCSP to complete the neutron-induced cross section data; a new evaluation reported the need of ^{233}U capture data.
The challenge in this measurement lies in the difficulty of measuring capture cross section data due to the competing capture and fission channels. Fission reactions are around one order of magnitude more likely than capture for ^{233}U. The accuracy in the capture cross section measurement relies on the discrimination between the γ-rays produced in capture and fission reactions, for which an experimental setup combining capture and fission detectors is needed.
Following this requirement, a new measurement has been performed at LANSCE combining the γ-ray array DANCE with the neutron detector NEUANCE to identify fission and neutron-capture events.
This measurement will provide results of the ^{233}U capture-to-fission ratio in the Resolved and Unresolved Resonance regions.

1 Introduction

In the Th-U fuel cycle, ^{232}Th transmutes into the fissile isotope ^{233}U, that when fissioning produces a large number of neutrons enough to maintain the chain reaction. The Th-U fuel cycle is of interest for thermal breeder reactors and fast reactors. In order to understand this cycle, accurate experimental data of the neutron-induced capture and fission reactions in ^{233}U are needed. The experimental ^{233}U(n,γ) data available in the literature are scarce and were measured some time ago, therefore new measurements are required to complete the neutron-induced reactions database.

Neutron-induced capture experiments are being carried out at the Los Alamos Neutron Science Center (LANSCE), at Los Alamos National Laboratory (LANL), using the DANCE γ-calorimeter. Neutrons are produced at LANSCE via spallation reactions caused by an 800 MeV proton beam with a nominal FWHM per pulse of 145 ns, hitting a tungsten target. Details of the LANSCE spallation source are found in [4]. DANCE is located at the flight path 14 of the Manuel Lujan Jr. Neutron Scattering Center, at 20 m from the spallation target, where water is used to moderate the fast neutrons before entering the flight path, producing a white neutron spectrum from thermal to 500 keV.

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The energy of the incident neutrons is measured using the time-of-flight technique. It consists in measuring the time that a neutron takes to travel from the spallation source to the reaction sample knowing the distance between both. The neutron energy is calculated using the relativistic equation, which, for neutron energies below 1 MeV, can be simplified to the non-relativistic equation.

2 The DANCE and NEUANCE detectors and targets

The **Detector for Advanced Neutron Capture Experiments (DANCE)** [5] is an array composed of 160 BaF_2 crystals in a 4π geometry. The crystals have a length of 15 cm and four different shapes, all of them covering the same solid angle. The sample under study is placed inside the 17 cm DANCE inner cavity, in the interior of the beam pipe.

The **NEUtron detector array at dANCE (NEUANCE)** [6] consists of 21 stilbene crystals arranged in a cylindrical geometry around the beam pipe, placed inside the 17 cm radius DANCE inner cavity, see figure 1. A ^{nat}Li-loaded polyethylene shell has been placed inside the DANCE inner cavity, between NEUANCE and DANCE, in order to reduce the scattered background from the neutrons hitting the sample backing. NEUANCE detects neutrons with energies above 500 keV, which corresponds to the energies of the fission neutrons, therefore low energy scattered neutrons with energies below this threshold are discriminated.

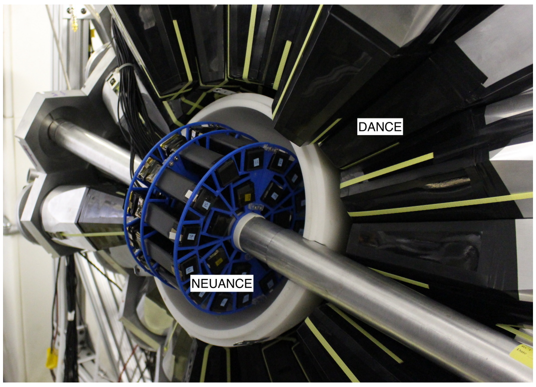

Figure 1. NEUANCE detector placed inside the DANCE detector inner cavity.

The DANCE and NEUANCE detectors are used in combination to perform this measurement in order to distinguish the fission and the capture γ-rays. One of the advantages of measuring fission reactions by detecting the fission neutrons instead of the fission fragments is the possibility of using a thick target as neutrons have no charge, reducing the required beam time.

Two ^{233}**U samples** of 10 mg and 20 mg were built at LANL. The 30 mg of ^{233}U material, with a 99.98% purity, used to prepare the samples were provided by Oak Ridge National Laboratory (ORNL) through the National Isotope Development Center (NIDC).

3 Data Analysis

Pulse Shape Discrimination methods have been used to separate γ-rays from α-particles with DANCE, and γ-rays from neutrons with NEUANCE, using the information of the fast and slow, and short and long components of the waveforms respectively.

The DANCE crystals have been calibrated using γ-sources, and the intrinsic radioactivity of the BaF_2. More specifically using the α-decay chain of the ^{226}Ra present on it, these are the ^{226}Ra (4.8 MeV), ^{222}Rn (5.5 MeV), ^{218}Po, and ^{214}Po peaks in the spectrum. NEUANCE has been calibrated using the γ-peaks: ^{22}Na (511 keV and 1274.537 keV) ^{137}Cs (661.657 keV), and ^{88}Y (898.047 keV and 1836.090 keV).

The first step on the data analysis is the search for coincidences between the γ-rays detected with DANCE and the neutrons detected with NEUANCE. The DANCE γ-rays found in coincidence are tagged as fission γ-rays, and the remaining events are left untagged. In figure 2, the cluster multiplicity (defined as a group of neighbour crystals) is plotted as a function of the total energy of the γ-rays; one can observe that the events in the diagonal of the figures correspond to fission reactions, and the events at the bottom of the untagged spectrum correspond to capture reactions. As NEUANCE tagging is not 100%, the remaining fission background left untagged had to be identified and subtracted. In order to do so, a window containing only fission events in the spectrum has been selected (see figure 2). The tagged spectrum has been then normalized to the untagged spectrum inside this window and subtracted.

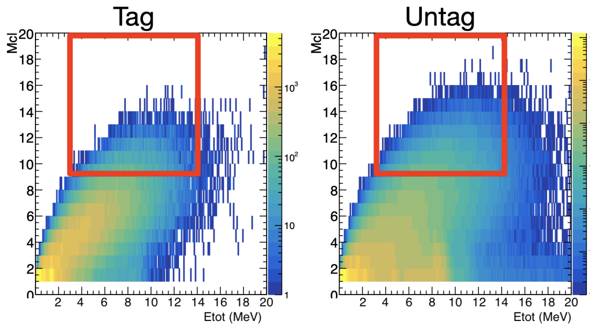

Figure 2. Cluster multiplicity as a function of the total energy of the γ-rays showing the tagged events (left figure) and the untagged events (right figure). The normalization window is indicated by the red rectangle.

The background varies with the incident neutron energy, therefore it has been identified and subtracted per neutron energy bin. Apart from fission, there are some other components of the background as it is the component due to scattering reactions on the sample backing and surrounding materials, and the delayed fission γ-rays. These components are shown in the total energy of the γ-rays plot, see figure 3. As the scattering background dominates at cluster multiplicities below 3, and the fission background does at multiplicities above 6, the analysis has been performed selecting the events with cluster multiplicities 4 and 5. The scattering events have been normalized in the figure to the scattering peak around 9 MeV, and the delayed fission γ-rays have been studied by taking data prior to the beam and it has been normalized to the binwidth of each energy bin. Once that the background has been subtracted, a cut around the Q-value peak, found at 6.845 MeV for capture reactions in ^{233}U, has been used to select the capture events.

4 Results

Providing the capture-to-fission ratio instead of a direct measurement of the capture cross section reduces the systematic uncertainties coming from the use of a different measurement to provide the neutron flux. The capture-to-fission ratio in ^{233}U has been calculated and normalized to the ENDF/B-VIII.0 evaluation in the energy region proposed by the evaluators between 8.1-14.7 eV [7]. The experimental data have been provided using different number of bins per decade depending on the statistics, therefore 1000 bins per decade have been used in the neutron energy region from 0.3-3 keV, and 125 bins per decade have been used from 3-300 keV, see figure 4.

A good agreement is observed between the experimental data and the ENDF/B-VIII.0 evaluation in the Resolved

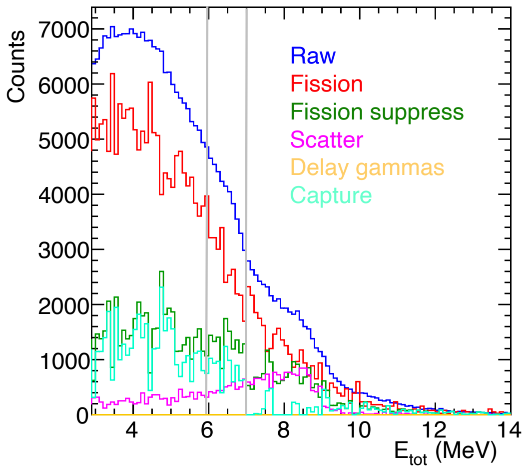

Figure 3. Total energy of the γ-rays showing the background components around 10 eV neutron energies. A cut has been performed around the Q-value for capture reactions in ^{233}U to select the capture events.

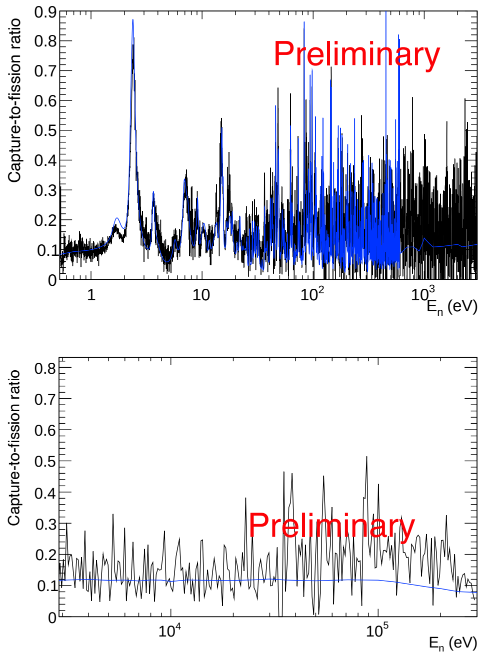

Figure 4. Prelimimary result (black line) of the capture-to-fission ratio in ^{233}U normalized to the ENDF/B-VIII.0 evaluation (blue line) in the energy region from 8.1-14.7 eV. Data with 1000 bins per decade are provided in the RRR (top figure), and 125 bins per decade in the URR (bottom figure).

Resonance Region (RRR). This measurement will provide more information to the existing experimental data available in the EXFOR database, especially in the Unresolved Resonance Region (URR) up to 300 keV for future evaluations.

5 Conclusions

A new measurement of the capture-to-fission cross section ratio in ^{233}U has been performed at LANSCE (LANL), combining the capture and fission detectors DANCE and NEUANCE at the end of 2020 and 2021. The ^{233}U material was provided by ORNL, and two samples of 10 mg and 20 mg were built at LANL. The preliminary results of the capture to fission ratio for incident neutron energies between 0.3-300 keV are here presented. The data have been normalized to the ENDF/B-VIII.0 evaluation in the energy region proposed by the evaluators between 8.1-14.7 eV. The measurement of the capture-to-fission ratio will reduce the systematic uncertainties that would come from the independent measurements required to provide a direct measurement of the capture cross section.

6 Acknowledgements

This work was supported by the Nuclear Criticality Safety Program (NCSP), funded and managed by the National Nuclear Security Administration for the Department of Energy. This work benefited from the use of the Los Alamos Neutron Science Center and was supported by the U.S. Department of Energy through the Los Alamos National Laboratory. Los Alamos National Laboratory is operated by Triad National Security, LLC, for the National Nuclear Security Administration of U.S. Department of Energy (Contract No. 89233218CNA000001). The isotope(s) used in this research was supplied by the U.S. Department of Energy Isotope Program, managed by the Office of Isotope R&D and Production.

References

[1] E. Berthoumieux, U. Abbondanno, G. Aerts, H. AÍvarez, F. AÍvarez-Velarde, S. Andriamonje, J. Andrzejewski, P. Assimakopoulos, L. Audouin, G. Badurek, et al., Pro- ceedings of the International Conference on Nuclear Data for Science and Technology 22-27, 571 (2007), URL https://doi.org/10.1051/ndata:07665.

[2] L. W. Weston, R. Gwin, G. deSaussure, R. R. Fullwood, and R. W. Hockenbury, Nuclear Science and Engineering 34, 1 (1968), URL https://doi.org/10.13182/NSE68-A19361.

[3] J. C. Hopkins and B. C. Diven, Nuclear Science and Engineering **12**, 169, (1962), URL https://doi.org/10.13182/NSE62-A26055.

[4] P. W. Lisowski, C. D. Bowman, G. J. Russell, and S. A. Wender, Nuclear Science and Engineering 106, 208 (1990), URL https://doi.org/10.13182/NSE90-A27471.

[5] M. Heil, R. Reifarth, M. Fowler, R. Haight, F. Kaȅppeler, R. Rundberg, E. Seabury, J. Ullmann, J. Wilhelmy, and K. Wisshak, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 459, 229 (2001), ISSN 0168-9002.

[6] M. Jandel, B. Baramsai, T. Bredeweg, A. Couture, A. Favalli, A. Hayes, K. Ianakiev, M. Iliev, T. Kawano, S. Mosby, et al., Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 882, 105 (2018), ISSN 0168-9002.

[7] L. Leal and M. Pigni (private communication).