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Measurements of the $^{234}\text{U}(\text{n},\text{f})$ and $^{236}\text{U}(\text{n},\text{f})$ cross-sections between 15 and 20 MeV with MicroMegas detectors

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Abstract The study of neutron-induced fission cross-sections of actinides is of great importance for the design of fast nuclear reactors and accelerator-driven systems. More specifically, ^{234}U and ^{236}U play an important role in the thorium cycle, therefore accurate fission cross-sections are required. In the energy range between 15 – 20 MeV, concerning $^{234}\text{U}(\text{n},\text{f})$ and $^{236}\text{U}(\text{n},\text{f})$, few available datasets exist in literature, measured with the time-of-flight technique, with errors up to 11% and discrepancies among them up to 8%. In this work a preliminary study is presented, for the measurement of the $^{234}\text{U}(\text{n},\text{f})$ and $^{236}\text{U}(\text{n},\text{f})$ total cross sections with monoenergetic neutron beams, produced via the $^3\text{H}(\text{d},\text{n})^4\text{He}$ reaction. In particular, a methodology proposed for the estimation of the parasitic neutrons is presented, along with its first experimental validation.

Keywords fission, quasi-monoenergetic neutrons, parasitic neutrons

INTRODUCTION

Neutron induced fission cross sections of actinides are of great importance for the design of fast nuclear reactors and accelerator-driven systems, as well as for the study of the fission process. The nuclei ^{234}U and ^{236}U play an important role in the thorium cycle, therefore fission cross-sections are required with an accuracy better than 5%. However, in the energy range between 15 and 20 MeV, few available cross-section datasets exist in literature. Concerning the $^{234}\text{U}(\text{n},\text{f})$ reaction, the existing cross-section data [1], [2], [3], all measured with the time-of-flight technique, show discrepancies up to 8% among them, with errors rising up to 11%. For the $^{236}\text{U}(\text{n},\text{f})$ reaction, the only available experimental dataset is by Tovenson et al. [1], measured also using the neutron-time-of-flight technique. In addition, the latest evaluated libraries ENDF/B-VII.1 [4], JEFF-3.2 [5], JENDL-4.0 [6], in the energy range between 15 and 20 MeV, show discrepancies up to 11%, for both reactions. In this scope, additional measurements with different techniques are required to improve the accuracy of the evaluations.

In this work, a preliminary study is presented, along with the first corresponding experimental results, in order to achieve a high-accuracy measurement of the above

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mentioned fission cross-sections, with quasi-monoenergetic neutron beams, produced via the $^3\text{H}(\text{d},\text{n})^4\text{He}$ reaction.

METHODOLOGY

The production of quasi-monoenergetic neutron beams in the energy range between 15 and 20 MeV is usually achieved via the $^3\text{H}(\text{d},\text{n})^4\text{He}$ reaction. The high Q-value (17.6 MeV) and the kinematics of the reaction allow for the production of monoenergetic neutron beams up to 20 MeV at forward angles. However, in experimental facilities with an absence of a supplementary neutron time-of-flight installation for the estimation of the neutron energies, the acute problem of the estimation of parasitic neutrons arises.

The parasitic neutrons are low energy neutrons, with respect to the main beam, and vary in energy from the thermal region up to a few MeVs, depending on the energy of the deuteron beam. Due to the low energy threshold in most fission cross-sections, pending on the reaction, these parasitic neutrons may yield a considerable contribution to the measured fission fragment yield, which needs to be carefully considered.

In this work, a methodology for the characterization of the beam is presented, using the multiple foil activation technique, as described in [7], [8]. The methodology is based on the understanding of the various sources of parasitic neutrons and the calculation of their intensity, through the activation of various, carefully selected foils. More specifically, the parasitic neutrons are produced 1) via reactions of deuterons with nuclei present in the tritiated target itself, 2) via reactions of deuterons with nuclei in the beam line (collimators, beam pipe, surrounding materials etc.) and 3) via reactions of neutrons with materials existing in the experimental hall area.

In particular, concerning the titanium tritiated target, ^{12}C nuclei are abundant from carbon built up, due to the presence of carbon in the beam line and vacuum pressure system and ^2H nuclei are implanted in the tritium target from previous irradiations. These nuclei, as shown in previous publications [9], produce parasitic neutrons via the reactions $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ and $^2\text{H}(\text{d},\text{n})^3\text{He}$ respectively. For deuteron energies greater than 3.7 MeV, the deuteron break-up reaction in the field of the ^3H nuclei is also possible. However, the tritiated target used in the experiments is not heavily used (less than 4 weeks of continuous irradiations in total), so the implanted deuterons in the target are not expected to produce a significant number of parasitic neutrons. So, while bombarding with deuteron energies lower than 3.7 MeV, parasitic neutrons are expected to be produced only via the $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ reaction, from the nuclei present in the tritium target.

On the other hand, the other two sources of parasitic neutrons, namely deuteron and neutron reactions in materials of the experimental area, such as deuteron break-up in the field of heavy nuclei and neutron scattering, do not result in distinct energy peaks in the flux spectrum, but rather appear as a low energy neutron tail.

Last but not least, parasitic neutrons can be produced via the $^{16}\text{O}(\text{d},\text{n})^{17}\text{F}$ reaction, since ^{16}O nuclei are always present in the beam line due to oxidization processes, and only marginally in the tritiated target. However, the threshold of this reaction is ~ 1.8 MeV, so as

long as the deuteron beam energy remains below this threshold, parasitic neutrons are not produced via the (d,n) reaction on ^{16}O .

To account for all the above-mentioned neutron groups, the analysis of at least three different reactions with the activation technique is necessary. Firstly, one reaction is required for the estimation of the flux of the main energy beam. The proposed reactions for this measurement are either the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$, or the $^{197}\text{Au}(n,2n)^{196}\text{Au}$ one. Both these reactions are used as standards for the fluence measurements of high energy neutrons, since their thresholds are relatively high, 3.2 MeV and 8.1 MeV respectively, and their cross-sections are well defined. Secondly, one additional reaction is required for the estimation of the parasitic neutrons produced via the $^{12}\text{C}(d,n)^{13}\text{N}$ reaction, with a threshold of 0.328 MeV. The energy of the parasitic neutrons produced, as results from the kinematics of the reaction, depends on the energy of the deuteron beam. At forward angles the neutrons emitted at 0° have energies ranging from 0.693 MeV to 3.65 MeV for deuteron beams with energies between 1 MeV and 4 MeV, respectively. The cross-section of the $^{115}\text{In}(n,n)^{115,\text{m}}\text{In}$ reaction is especially suitable for the measurement of these parasitic neutrons, since its cross-section has a value of approximately 0.3 b in the energy range between 2 and 10 MeV, while the cross-section below 0.4 MeV is negligible, as it is at least 3 orders of magnitude lower. At higher energies, the cross section is about 0.06 b, but the only contribution to the activation spectra at this high energy region is expected to originate from the high energy neutrons of the main flux, so a correction can be easily made. Finally, an additional foil is needed for the measurement of the low energy neutrons produced from neutron scattering in the experimental hall area and from the $^{16}\text{O}(d,n)^{17}\text{F}$ reaction, namely a Au foil, exploiting the well-studied $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction. These low energy neutrons are expected to have energies ranging from the thermal region due to neutron scattering, up to about 2 MeV due to the $^{16}\text{O}(d,n)^{17}\text{F}$ reaction, depending on the energy of the deuteron beam. The $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction has high cross-section values for low energy neutrons, while at high neutron energies the cross-section is negligible. So, a contribution from neutrons of the main beam is not expected, while the contribution from the neutrons produced from the $^{12}\text{C}(d,n)^{13}\text{N}$ reaction can easily be calculated.

The estimation of the low energy neutrons, via the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction, can only be achieved in a complimentary way, along with extensive MCNP simulations including a detailed description of the target and the detector setup geometry. More specifically, the neutron flux calculated by such MCNP simulations underestimates the induced low energy neutron tail. So, a correction factor is applied to the low energy flux in order to achieve a good agreement between the corrected MCNP calculations and the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ experimental values.

EXPERIMENTAL DETAILS

The validation of the methodology was achieved with an experimental setup consisting of a fission chamber and a holder containing the activation foils. The experiment was performed at the 5.5 MV Tandem van de Graaf accelerator laboratory National Centre for

Scientific Research “Demokritos”. The neutron beam was produced via the $^3\text{H}(d,n)^4\text{He}$ reaction, by bombarding a solid TiT target with 2.3 MeV deuterons, resulting in the production of 18.4 MeV neutrons.

The fission chamber was filled with a gas mixture of 90% Ar and 10% CO_2 at atmospheric pressure, containing both the fission targets and the micromegas detectors for the detection of the neutron induced fission events. The setup consisted of five actinide targets, as shown in Fig. 1. The ^{236}U and the ^{234}U target, as well as a ^{238}U and a ^{235}U target, were put back to back, to obtain the same flux in both target pairs. Yet, only the reference targets, namely ^{238}U and ^{235}U , were used for the validation of the methodology. A typical fission spectrum from ^{235}U is presented in Fig. 2. The estimation of the parasitic neutrons was achieved with the use of two foils, namely a Au foil and an In foil.

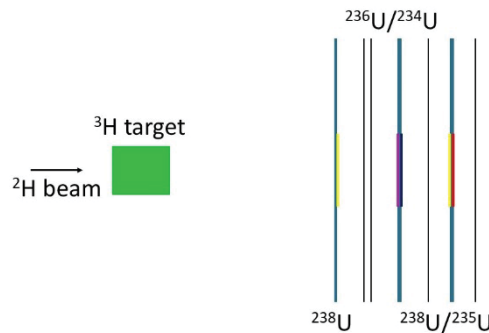


Fig. 1. Schematics of the target assembly in the MicroMegas chamber

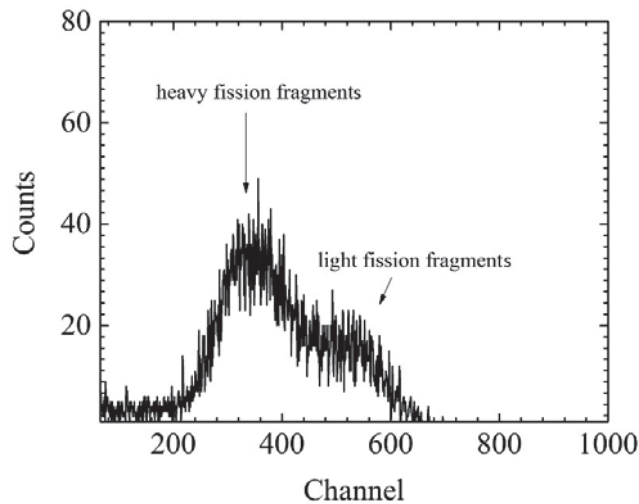


Fig. 2. Fission fragment spectrum from ^{235}U

The experimental fission counts were estimated from the analysis of the fission spectra. The experimental counts resulted from fission events both from the main flux and from parasitic neutrons of various energies. The intensity of the main flux was estimated via the $^{197}\text{Au}(n,2n)^{196}\text{Au}$ reaction, so by correcting for the different solid angle subtended by the Au foil and by each actinide target, the neutrons of the main flux incident on the targets were estimated. Since the fission cross-sections of the reference targets for incident neutrons at 18.4 MeV is well defined, the fission counts due to the main flux were calculated. Following, the same pattern, using the $^{115}\text{In}(n,n)^{115,\text{m}}\text{In}$ reaction, the intensity of the parasitic neutrons

originating from the $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ reaction was estimated. From the reaction kinematics, the energy of these parasitic neutrons equals 1.97 MeV at forward angles. At this energy the fission cross-sections of ^{235}U and ^{238}U are well defined. So, the fission counts, due to these parasitic neutrons can be calculated. Finally, the $^{197}\text{Au}(\text{n},\gamma)^{198}\text{Au}$ reaction was used for the estimation of a correction factor for various low energy neutrons to be applied in the MCNP flux calculations. The corrected flux for these low energy neutrons was used for the estimation of the fission counts in the ^{235}U target. Due to the high fission cross-sections at low energies, these low energy neutrons are responsible for a significant number of counts in the fission spectrum.

The comparison between the experimental fission counts and the calculated fission counts was in very good agreement. More specifically, the experimental and calculated fission counts differed by only 6% for the rear ^{238}U target, 4% for the ^{235}U target and 23% for the front ^{238}U . The latter discrepant result may be attributed to the fact that no correction has been made for the angular spread of the neutron beam, an effect which is more important for the targets closer to the TiT target.

RESULTS AND DISCUSSION

In this work, a methodology is proposed for measuring fission cross-sections with quasi-monoenergetic neutron beams in the energy range between 15 and 20 MeV, when complementary time-of-flight measurements are not possible for the estimation of the neutron energies. The methodology has been tested for a neutron beam of 18.4 MeV and yielded quite satisfactory results.

For the implementation of the fission experiments, additional measurements are proposed, for the estimation of the parasitic neutrons. More specifically, apart from the activation measurements already mentioned, a liquid scintillator will provide information for neutrons above 1 MeV. Additionally, measurements with the same setup but without the TiT target, will account for the parasitic neutrons produced from deuteron reactions in the beam line and surrounding materials.

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