Annual Symposium of the Hellenic Nuclear Physics Society

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Βιβλιογραφική αναφορά:

Diakaki, M., Vlastou, R., Kokkoris, M., Papadopoulos, C. T., Tsinganis, A., Kalfas, C. A., Lagoyannis, A., & Provatas, G. (2019). Attempted study of the 237Np(n,2n)236Np reaction cross section at 9.5 MeV. *Annual Symposium of the Hellenic Nuclear Physics Society*, *18*, 101–106. https://doi.org/10.12681/hnps.2544

Attempted study of the ^{237}Np(n,2n)^{236}Np reaction cross section at 9.5 MeV

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Abstract

The cross section measurement of the ^{237}Np(n,2n)^{236}Np reaction has been attempted at an incident neutron energy of 9.5 MeV by means of the activation technique. The neutron beam was produced via the ^{2}H(d,n)^{3}He reaction at the VdG Tandem accelerator of NCSR "Demokritos". It is the second time that this measurement has been tried with a gamma spectroscopy method and the difficulties faced due to the high gamma ray background produced by the sample itself and the fission fragments produced by the irradiation, in combination with the very low intensity of the gamma ray of interest are being reported.

Introduction

The study of neutron threshold reactions on actinide isotopes is of considerable importance for practical applications, especially in reactor physics. The ^{237}Np isotope is a product of the fuel cycle of Uranium in the reactor and so the study of ^{237}Np(n,2n)^{236}Np reaction is of interest for the following reasons: Firstly, it is the major reaction channel in competition with fission that causes the emission of neutrons and, secondly, it leads to the production of the long-lived (80y) ^{232}U (see fig. 1), the decay of which contains a series of short-lived daughter nuclei and emission of 'hard' gamma rays. This explains why the study of this reaction is one of the priorities of the IAEA [1]. Nevertheless, it presents experimental difficulties and very few cross section data exist in literature (see fig. 1). More specifically, only one dataset exists for neutron energies up to 9.5 MeV [2] and a few around 14.5 MeV with large discrepancies among them [3-7]. These datasets are mainly deduced by measuring the α-particles of the ^{236}Pu decay after chemical separation of Pu from Np and using ^{239}Pu/ ^{242}Pu tracers.

The ^{236}Np isotope has two isomers, one short lived, with a half life of 22.5 h, and one long lived, with a half life of 153000 y, but no information exists on which is the ground state [9]. The short lived isomer decays with a probability of 50% to ^{236}U and with a probability of 50% to ^{236}Pu. The long lived isomer decays basically (with a probability of 86.3%) to excited states of ^{236}U (see fig. 2). In the present study, the measurement of the cross section of the production of the short lived isomer was attempted, with use of the activation method, i.e. by bombarding the Np target with the neutron beam and measuring the induced activity off-line with a HPGe detector. The detection of gamma rays from the decay of ^{236}U was chosen, although they have very low intensity: 642.4 and 687.6 keV, with (1.09 ± 0.08)% and (0.30 ± 0.02)%,

respectively [8]. This has already been tried in the past [5], and problems were reported due to the high gamma ray background from the target itself and the fission fragments produced, in combination with the low intensity gamma rays detected. The cross section data that occurred from the gamma ray spectroscopy were ~10% higher than the ones occurred from the measurement of the α-particles of the ^{236}Pu decay.
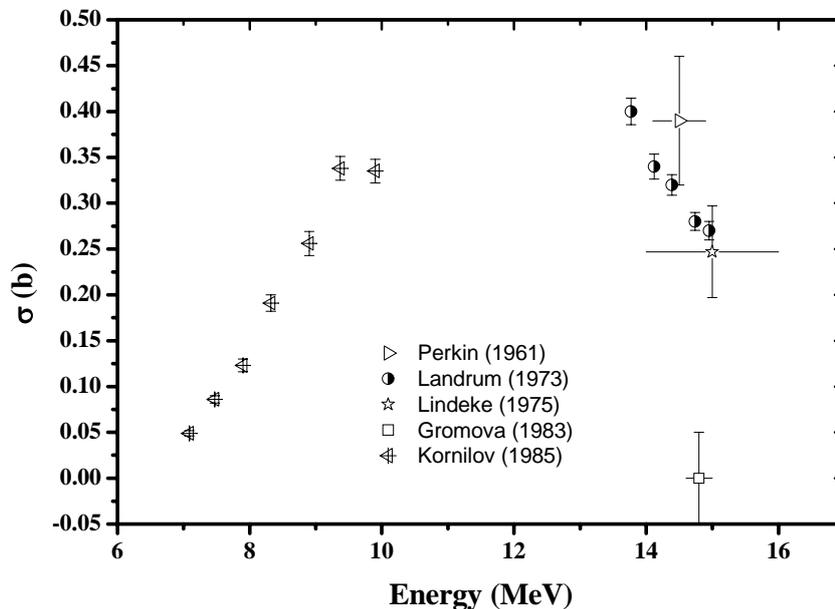

Fig. 1: Existing cross section data for the ^{237}Np(n,2n) reaction (for the production of the short lived isomer) [8].

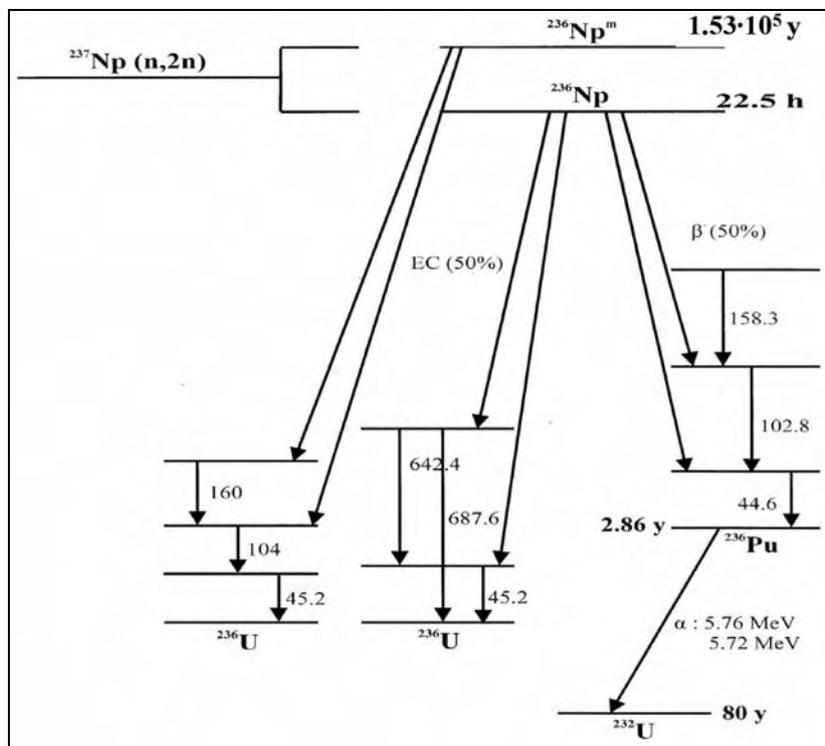Fig. 2: Decay scheme of the reaction ^{237}Np(n,2n). For the long lived isomer of ^{236}Np only the decay to ^{236}U (the most probable) has been included in the scheme.

Experimental Procedure

The measurement was performed at an incident neutron energy of 9.5 MeV. The quasi-monoenergetic neutron beam was produced via the ^{2}H(d,n)^{3}He reaction by bombarding a deuterium gas target with a deuteron beam at currents around 2-2.5 μA. The deuteron beam was provided by the 5.5 MV Tandem Van de Graaff accelerator of the N.C.S.R. "Demokritos". The gas target is fitted with a 5 μm molybdenum entrance foil and a 1 mm Pt beam stop and is constantly cooled with a cold air jet during irradiation to diminish the risk of damage to the Mo foil. The deuterium pressure was set to 1500 mbar. With this setup, the achieved neutron flux was of the order of ~10^6 n/(cm^2 sec). The absolute flux of the beam was obtained with respect to the ^{27}Al(n,α)^{24}Na reference reaction, while its variations during the irradiation were monitored by a BF$_3$ detector placed at a distance of 3 m from the neutron source.

The sample was 6.892 g of NpO$_2$ mixed with 40.152 g of S canned in 0.5mm Al and it was provided at the IRMM of Belgium. The nominal radioactivity of the sample was 155MBq. Shielding was considered necessary in order to reduce the high gamma ray background from the target itself and for radioprotection reasons. A mixed Pb-Cd shielding was chosen, thinner at the top and the bottom of the cylindrical sample (1mm Cd - 1mm Pb) and thicker at the side (3mm Cd - 5mm Pb). The shielded sample was placed at a distance of 3 cm from the deuterium gas cell, stacked between two 0.5 mm thick reference Al foils, and was irradiated for ~44 h i.e. for two half lives of the short lived ^{236}Np isomer. The integrated flux, obtained by the front Al foil was ~5·10^{10} n/cm^2 (±6%).

The induced activity of the sample and reference foils was measured off line with HPGe detectors of 80% and 56% efficiency respectively.
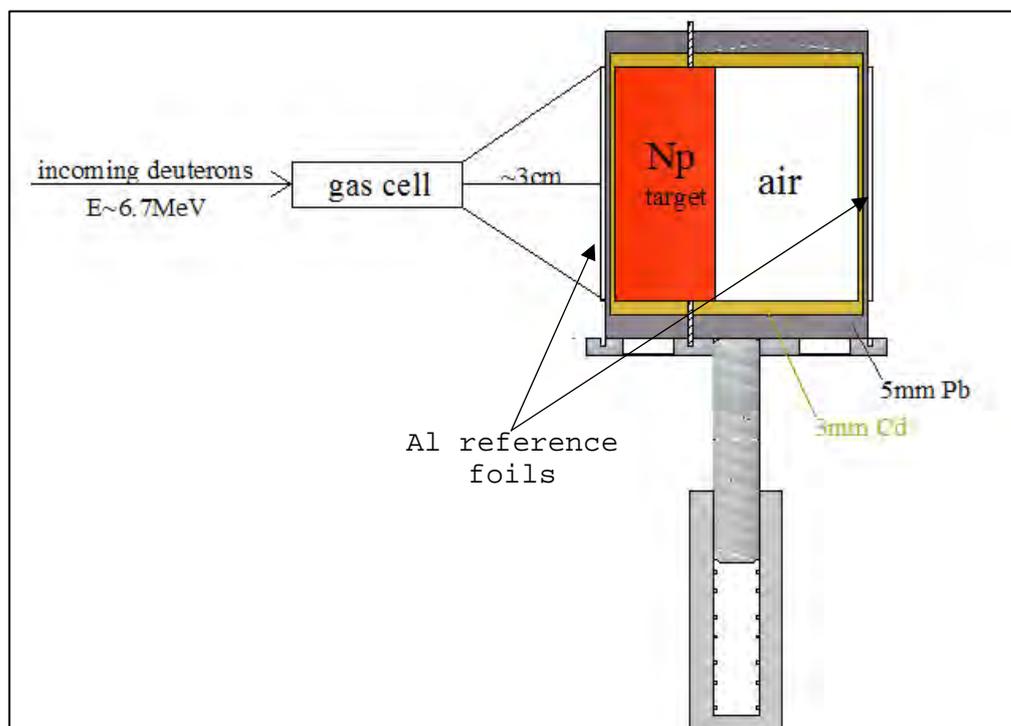

Fig. 3: The setup for the neutron irradiation of the Np target.

Problems faced during the activity measurements

The high radioactivity of the sample itself caused problems during the activity measurements. It was fabricated in 1994, and the reported isotopic purity of ^{237}Np (half -life of $2.14 \cdot 10^6$ years) initially was 99.99%, but at the time of the measurement (2010) the sample was contaminated by the short lived daughter nucleus ^{233}Pa (half - life of 27 d) in secular equilibrium with ^{237}Np, and this caused a severe increase at the already high gamma ray background, even before the irradiation. As a result, additional shielding of the HPGe detector was needed (3mm of Pb absorber was placed in front of the detector) and the sample-to-detector distance was increased to 43 cm. This detection setup caused a severe reduction of the efficiency by 1 order of magnitude for the gamma ray of interest ($\text{eff}_{642\text{keV}} \sim 0.0005$) and its attenuation by the shielding which reached ~40% (calculated using gamma ray attenuation data from XCOM [10] and validated with the measurement of the activity of a calibrated ^{152}Eu source with and without the Pb shielding of the detector).

A further problem was the production of additional gamma ray background by the fission fragments in the sample. In figure 4, a calibrated spectrum of the sample before the irradiation is presented, where it becomes clear that the dominant background comes from the ^{233}Pa present in the sample. The gamma ray peaks from the decay of ^{237}Np have energies below ~280 keV and were more efficiently absorbed by the shielding of the sample. In figure 5 a calibrated spectrum of the sample after the irradiation is presented in the energy region above ~450 keV (at lower energies the gamma ray activity of ^{233}Pa prevents the analysis of gamma ray peaks coming from isotopes produced by the irradiation). Most of the peaks are from fission fragments from the fission of ^{237}Np and ^{233}Pa [11,12]. The peak of 642 keV that would occur from ^{237}Np(n,2n)^{236}Np reaction should appear in the position circled on the spectrum. Nevertheless, after a careful analysis of the peak that appears in this position, taking into account the number of counts expected (based on the previous cross section measurements-see fig. 1), the FWHM of the peak in comparison with other neighbor peaks, the background etc., it turned out that it was contaminated.

Fig. 4: Calibrated spectrum of the sample before the irradiation.

An effort has been made to check which peak(s) contaminate 642 keV region. The candidate gamma rays mainly were from fission fragments from the actinides in the sample and their daughters as well as isotopes that would occur from other neutron induced reactions in the sample. The selection was made taking into account the decay curves and the intensities of the candidates, and their correlation to other peaks that appear in the spectrum. It turned out that the contamination most probably comes from gamma rays of ^{110}In (possible fission fragment of actinides) and/or from ^{233}Pa(n,γ) ($T_{1/2}$=6.7h) and/or ^{233}Pa(n,2n) ($T_{1/2}$=1.32d) . Other gamma rays from these residual nuclei are also seen in the spectrum. This unfortunate result precludes a conclusion on the accuracy of any possible analysis of the gamma ray peak of interest and thus the extraction of safe results for the cross section measurement has been impossible.
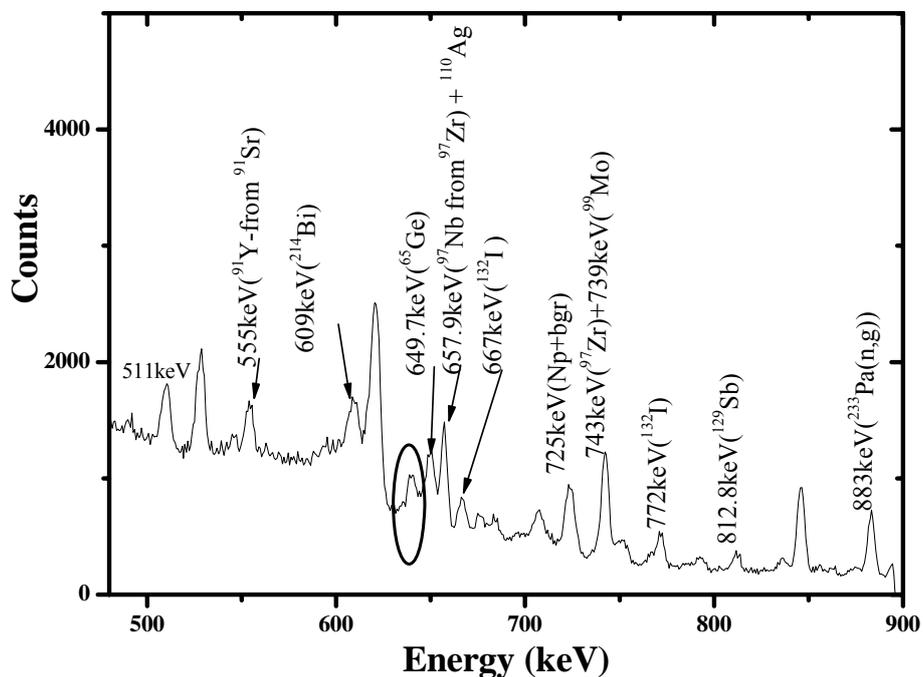

Fig. 5: Calibrated spectrum of the sample after the irradiation ($t_{measurement}$=10h), zoomed in the energy region of interest. The gamma ray energy of interest 642 keV is circled at the spectrum.

Conclusions

The cross section measurement of the ^{237}Np(n,2n)^{236}Np reaction has been attempted at an incident neutron energy of 9.5 MeV by means of the activation technique. The main difficulty faced was the high gamma ray background from the target itself. The main contribution to this background was the gamma rays coming from the short lived daughter nucleus ^{233}Pa which is in secular equilibrium with ^{237}Np. As a result, additional shielding of the detector and increase of the sample-to-detector distance became necessary, which reduced the efficiency of the detector by 1 order of magnitude. Furthermore, contamination of the gamma ray peaks coming from ^{237}Np occurred due to neighboring transitions from the ^{233}Pa content in the target. Thus, chemical separation of ^{233}Pa from ^{237}Np before the irradiation is considered essential

in order to measure cross sections of neutron induced reactions on Np with use of the activation method.

Acknowledgements
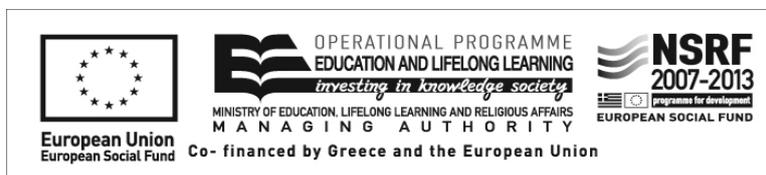
References

1. http://www-nds.iaea.org/manread/
2. N.V.Kornilov et al., At.Energ. 58 (1985) 131.
3. J.L.Perkin and R.F.Coleman, Reactor Science and Technology 14 (1961) 69.
4. J.H.Landrum et al., Phys.Rev.C 8 (1973) 1938.
5. K.Lindeke et al., Phys.Rev.C 12 (1975) 507.
6. E. A. Gromova et al., At.Energ. 54 (1983) 108.
7. E. A. Gromova et al., At.Energ. 60 (1986) 68.
8. www.nndc.bnl.gov/**exfor**/
9. K. Wisshak et al., Phys.Rev.C 73, 045807 (2006).
10. www.nist.gov/pml/data/xcom
11. M. N. Namboodiri, N. Ravindran, M. Rajagopalan, M. V. Ramaniah, J. inorg. nucl. Chem. Vol. 30 (1968) 2305-2310.
12. Fission Product Nuclear Data, Vol. I, IAEA-169, Proceedings of a panel on Fission Product Nuclear Data by IAEA, Bologna (1973).