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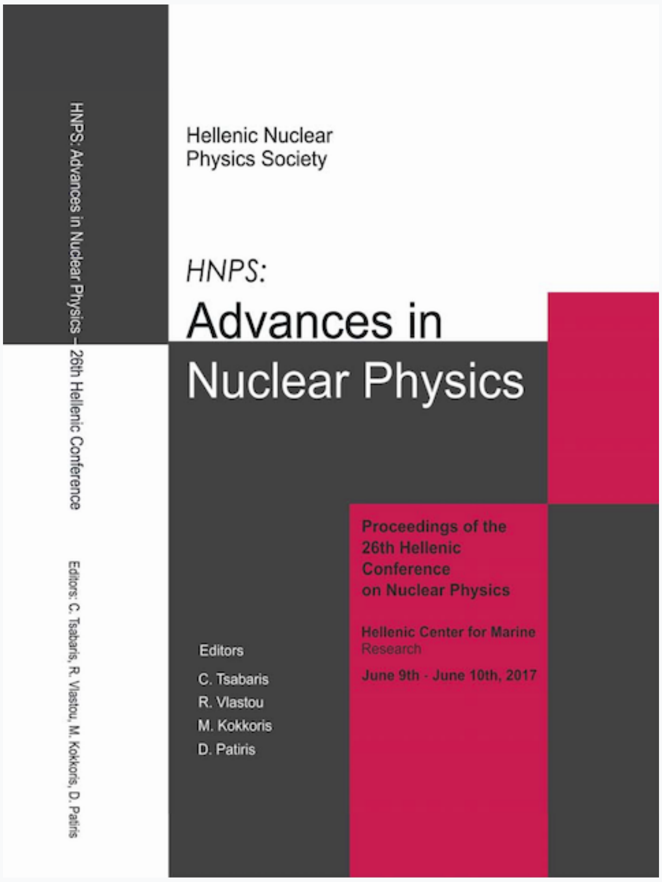

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Experimental and theoretical study of the (n,2n) reaction on ^{174,176}Hf isotopes

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Abstract In the present study we performed experimental cross-section measurements for the ^{174}Hf(n,2n)^{173}Hf and ^{176}Hf(n,2n)^{175}Hf threshold reactions at the 5.5-MV Van de Graaf Tandem accelerator of NCSR "Demokritos", using the activation technique. The neutron energy beam of 18.9 MeV was produced via the ^{3}H(d,n)^{4}He reaction. The estimation of the neutron flux, at the order of 10^{10} n·cm^{-2}, was implemented by NeuSDesc and MCNP Monte Carlo simulations along with the multiple foil activation technique at 18.9 MeV, while a BF3 detector was monitoring the flux variation during irradiation. The ^{176}Hf(n,2n)^{175}Hf yield has been corrected from the contribution of ^{177}Hf(n,3n)^{175}Hf and ^{174}Hf(n,γ)^{175}Hf reactions. Theoretical calculations of the ^{174}Hf(n,2n)^{173}Hf and ^{176}Hf(n,2n)^{175}Hf cross-sections were performed using the nuclear statistical code "EMPIRE 3.2.2" in direct comparisons with experimental data. The impact of the different Optical Model Potentials and Nuclear Level Densities - available as input options in the code - was investigated in detail, as well as the importance of pre-equilibrium emission and relative contributions obtained with the different pre-equilibrium models.

Keywords Nuclear reactions, (n,2n) reaction cross section, neutron activation, Hafnium, "EMPIRE" nuclear cross section estimation

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INTRODUCTION

The significance of neutron-induced reactions is widely recognized, mostly due to their fundamental role in Nuclear Physics and Astrophysics research, as well as to their numerous practical applications. In the case of Hafnium, it is worth noticing that is one of the most expensive elements in the world, with important applications in areas such as nuclear technology, medicine and industry. Among its key properties, Hafnium's high absorption cross section for thermal neutrons is exploited in the manufacturing of reactor control rods for nuclear submarines. Regarding ^{174}Hf(n,2n)^{173}Hf and ^{176}Hf(n,2n)^{175}Hf reactions, the experimental cross section data for the energy region 18-20 MeV are scarce and discrepant [1-10]. In the current work, we present the cross-section measurements of ^{174}Hf(n,2n)^{173}Hf and ^{176}Hf(n,2n)^{175}Hf reactions, performed at the 5.5 MV Van de Graaff Tandem T11/25 accelerator for the neutron beam energy of 18.9 MeV. We implemented the activation technique relative to the ^{27}Al(n,α)^{24}Na and ^{93}Nb(n,2n)^{92m}Nb reference reaction cross sections.

Furthermore, theoretical statistical model calculations were performed using the code

“EMPIRE 3.2.2” in comparison with all available data.

EXPERIMENTAL DETAILS

Experimental cross-section measurements for the ^{174}Hf(n,2n)^{173}Hf and ^{176}Hf(n,2n)^{175}Hf threshold reactions, using the activation technique, were performed in the 5.5-MV Van de Graaf Tandem accelerator of the National Centre for Scientific Research (NCSR) "Demokritos". The neutron energy beam of 18.9 MeV was produced via the ^{2}H(d,n)^{3}He reaction, implementing a Ti-tritiated target of 400GBq activity (mass: 2305 μg cm^{-2}), consisted of 1.53 mg/cm^2 Ti-T layer, on a 1 mm thick Cu backing for good heat conduction. Two collimators of 5 and 6 mm in diameter were used. The deuteron beam current was measured both at the collimators and the target and was kept at ~ 550-600 nA.The Hf target was a thin metallic foil of natural Hf (diameter: 14.08±0.01mm, thickness: 0.51±0.05mm, mass: 0.691±0.001g). In order to measure the neutron flux at the target position, reference foils of high purity Al (diameter: 14.13±0.11mm, thickness: 0.58±0.05mm, mass: 0.206±0.001g) and Nb (diameter: 13.32±0.02mm, thickness: 0.30±0.05mm, mass: 0.300±0.001g) were placed in the front and back of the target. The sample and the reference foils, were placed at a distance of ~1.5 cm from the flange with the tritium target assembly in the following order Nb→Hf→Al.

The irradiation was continuous for about 24 hours, while a BF_3 detector at a distance of ~3 m from the neutron production was monitoring the neutron flux. These spectra were saved at regular time intervals (~60s) in a separate ADC during irradiation. After the termination of the irradiation phase, the activity of the sample and reference targets was measured off- line by two properly shielded HPGe detectors of 100% and 16% efficiency respectively. The absolute efficiency of each detector was obtained separately, by using a calibrated ^{152}Eu source, placed at the same distance as the samples. The efficiency value at the 121.79keV gamma-ray of ^{152}Eu, was properly corrected for the contamination due to ^{154}Eu in the source.

Characterization of the (n,2n) cross-sections was achieved by analyzing off-line spectra from both the sample and the reference targets. The ^{176}Hf(n,2n)^{175}Hf yield has been corrected for the contribution of ^{177}Hf(n,3n)^{175}Hf, ^{174}Hf(n,γ)^{175}Hf reactions. In a similar manner, the ^{174}Hf(n,2n)^{173}Hf yield has been corrected from the ^{180}Hf(n,2n)^{179m2}Hf reaction. Additionally, contaminations corresponding to the self-absorption of the sample, counting geometry, coincidence summing effects, of cascading γ- rays along with the product nuclides decay as well as the neutron beam flux fluctuation upon the whole irradiation time have also been taken into consideration.

RESULTS AND DISCUSSION

Natural Hf consists of five stable isotopes ^{176}Hf, ^{177}Hf, ^{178}Hf, ^{179}Hf, ^{180}Hf (respective abundances: 5%, 19%, 27%, 13% and 35%) and one radioactive ^{174}Hf (abundance: 0.2%) with estimated half life $t_{1/2}$ = 2x10^{15}y. Consequently, the reactions under study ^{176}Hf(n,2n)^{175}Hf and ^{174}Hf(n,2n)^{173}Hf are accordingly contaminated by the reactions

^{177}Hf(n,3n)^{175}Hf, ^{174}Hf(n,γ)^{175}Hf and ^{180}Hf(n,2n)^{179m2}Hf. These contaminations have been taken into account at the determination of (n,2n) reaction cross-section for both ^{174,176}Hf isotopes, as follows.

The ^{174}Hf(n,2n)^{173}Hf reaction

The residual nuclei ^{173}Hf with a half-life of 23.6h decays to the stable ^{173}Lu isotope. In this work, the calculation of the experimental cross-section measurement of the ^{174}Hf(n,2n)^{173}Hf reaction was accomplished using the characteristic γ-ray transitions of ^{173}Lu at 123.67 and 296.97 keV. The experimental cross-section along with the available data and theoretical estimations is shown in Fig.1a.

The ^{176}Hf(n,2n)^{175}Hf reaction

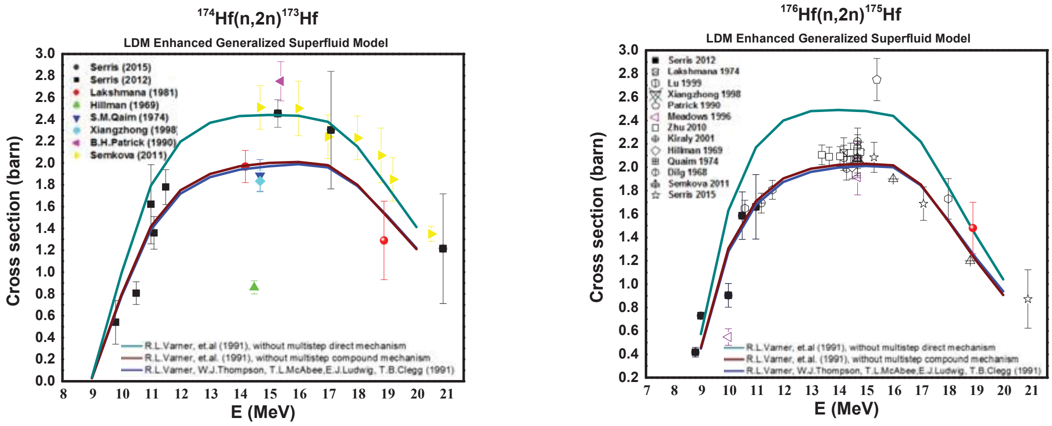
The ^{176}Hf(n,2n) reaction leads to the formation of ^{175}Hf isotope with a half life of 70d, which decays to the ^{175}Lu. The characteristic 343.4keV transition of ^{175}Lu was used for the determination of the ^{176}Hf(n,2n)^{175}Hf reaction cross section. However, the ^{175}Hf isotope was also produced from the ^{177}Hf(n,3n) reaction, having a high cross-section in the energy region of 18-20 MeV. Therefore, this contamination was taken into consideration and a corresponding correction has been implemented following three steps. First, the well estimated cross-section of the ^{177}Hf(n,3n)^{175}Hf reaction at 18.9 MeV neutron energy was retrieved from existing data in the literature (σ^r = 1.017 barn). Secondly, the actual number $N_T{}^r$ of the initial ^{177}Hf nuclei in the target was calculated and thirdly, using the neutron flux on the target (Φ^r), the number of the produced ^{175}Hf nuclei during irradiation was extracted by using the formula $\Phi^r = \frac{N_P^r}{N_T^r \cdot \sigma^r}$. Thus, the yield from the ^{176}Hf(n,2n)^{175}Hf reaction in the total yield of ^{175}Hf, was corrected by subtracting the ^{177}Hf(n,3n)^{175}Hf yield from the spectrum.

The same treatment, to account for the correction due to the ^{174}Hf(n,γ)^{175}Hf reaction, yielded irrelevant contribution, due to the very low (n,γ) cross section in the high neutron energy of 18.9 MeV.

Figure 1b presents the experimental cross-section of the ^{176}Hf(n,2n)^{175}Hf reaction with the existing experimental data and theoretical calculations.

Theoretical calculations

Theoretical cross-section calculations of ^{176}Hf(n,2n)^{175}Hf and ^{174}Hf(n,2n)^{173}Hf reactions were performed according to the Hauser-Feshbach compound nucleus theory using the nuclear statistical code "EMPIRE" (3.2.2 version). Several combinations of nuclear level density (NLD), optical model proton potential (OMP) and optical model potential (OMP) for outgoing neutrons were tested. In all of them, pre-equilibrium effects were taken into account through the multi-step-direct (MSD) and multi-step-compound (MSC) formulations of the code. The most promising results for both ^{174,176}Hf isotopes revealed upon the combination of Varner [11] et. al. OMP for neutrons, Konig [12] et. al. OMP for protons, while the nuclear level density was determined via the Generalized Superfluid Model. These results are presented in Fig.1a,b along with all existing experimental data including the data from this study. It is clear that the theory is not able to accurately reproduce the experimental data –not even within errors- for both ^{174,176}Hf isotopes. More thorough theoretical investigation is needed in order to achieve better agreement between experimental data and theoretical calculations.



(a) **(b)**

Fig. 1. Theoretical results of **(a)** ^{174}Hf(n,2n)^{175}Hf and **(b)** ^{176}Hf(n,2n)^{177}Hf (E_n=18.9 MeV) cross section calculation along with all existing data. The present experimental work is shown by the red point.

CONCLUSIONS

In the current work we aimed to provide a complete study of the (n,2n) reaction of ^{174,176}Hf isotopes, by a combination of experimental and theoretical cross-section measurements at 18.9 MeV neutron energy. It can be seen that following the aforementioned corrections, the experimental results are in agreement with the existing data in literature, despite previous measurements' potential inadequacies and discrepancies. The theoretical calculations led to the conclusion that none of the combinations of OMP for neutrons and protons and NLD model (available in RIPL-3 for neutron energy region that includes 18.9 MeV) can give satisfactory results compared to the experimental data for both ^{174,176}Hf isotopes. Thus, further theoretical investigation is needed in order to better reproduce the existing experimental data.

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