

Cross Section Measurements and Theoretical Study of the $^{174,176}\text{Hf}(n,2n)^{173,175}\text{Hf}$ Reactions

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Abstract. Experimental cross section measurements for the $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ and $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ reactions were carried out, using the activation technique. The neutron beam energy in the range of 15.3–20.3 MeV was produced via the $^3\text{H}(d,n)^4\text{He}$ reaction at the 5.5 MeV Tandem Van de Graaf accelerator laboratory of NCSR "Demokritos". A thin metallic foil of natural Hf was used, while for the determination of the neutron flux at the target position, reference foils of Al were placed at the front and back of the Hf target. The irradiations were continuous for ~24–48 hours, leading to a total neutron fluence of 10^{10} – 10^{11} n/cm² and a BF₃ detector was used for monitoring the neutron flux during the irradiations. After the end of each irradiation, the activity of the Hf target and the Al reference foils were measured off-line by two HPGe detectors. The $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ reaction has been corrected for the contribution of the $^{177}\text{Hf}(n,3n)^{175}\text{Hf}$ and $^{174}\text{Hf}(n,\gamma)^{175}\text{Hf}$ reactions. Statistical model calculations based on the Hauser-Feshbach theory have also been performed using the EMPIRE 3.2.3 code. The predictions have been compared with the data of the present work as well as with data from literature.

1 Introduction

The importance of neutron-induced reactions is widely recognized, mostly due to their fundamental role in the research of Nuclear Physics and Astrophysics as well as in their numerous practical applications [1–3]. In the case of Hafnium, it is worth mentioning that is one of the most expensive elements in the world, with important applications in the areas of nuclear technology, medicine and industry. Among its main properties, Hafnium's high absorption cross section for thermal neutrons is exploited in the manufacturing of reactor control rods for nuclear submarines. In addition, neutron induced reactions on W and Ta in reactor materials could lead to long lived isomeric states of Hf isotopes with rather harmful γ -ray production. Thus, the knowledge of neutron cross-sections on Hf isotopes is of great importance both for applications concerning the interaction of neutrons with matter as well as for testing nuclear models.

Regarding $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ and $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ reactions, the experimental cross section data in literature are scarce and discrepant. In 2012, these two (n, 2n) reaction cross sections were studied by our group at five neutron beam energies between 8.8 and 11.0 MeV [4] and the present work constitutes a continuation of the previous one in order to determine the cross section at higher neutron energies in the region between 15.3 and 20.3 MeV. Furthermore, in this work the $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ reaction has been corrected for the contribution of the $^{177}\text{Hf}(n,3n)^{175}\text{Hf}$

and $^{174}\text{Hf}(n,\gamma)^{175}\text{Hf}$ reactions, as will be described in detail in section 3. Theoretical calculations of the $^{174}\text{Hf}(n,2n)$ and $^{176}\text{Hf}(n,2n)$ cross sections have also been performed using the code EMPIRE 3.2.3 [5] and will be presented in section 4 along with the experimental data.

2 Experimental procedure

Experimental cross-section measurements for the $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ and $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ threshold reactions, using the activation technique, were performed at the 5.5-MV Van de Graaf Tandem accelerator of the National Centre for Scientific Research (NCSR) "Demokritos". The neutron beams were produced via the $^3\text{H}(d,n)^4\text{He}$ reaction, implementing a Ti-tritiated target of 400GBq activity on a 1 mm thick Cu backing for good heat conduction. The flange with the Ti-T target assembly was air-cooled during the irradiations. The Hf target was a thin metallic foil of natural Hf of 14 mm diameter and 0.51 mm thickness. In order to measure the neutron flux at the target position, reference foils of high purity Al, Au and Nb (in some of the irradiations), of the same diameter as the Hf foil, were placed in the front and the back of the Hf target. The cross section values for the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$, $^{197}\text{Au}(n,2n)^{196}\text{Au}$ and $^{93}\text{Nb}(n,2n)^{92m}\text{Nb}$ reference reactions were adopted from the IRDFF 1.05 library [6]. The values of the neutron flux obtained from all the reference foils were consistent. The target and reference foils were placed at a distance of ~2 cm

from the flange with the Ti-T target, thus limiting the angular acceptance to $\pm 19^\circ$, where the neutron beam can practically be considered as monoenergetic.

Extensive Monte Carlo simulations by means of the NeuSDesc [7] and MCNP5 codes [8] were also used for the neutron flux determination, taking into account the whole geometry of the irradiation setup. As an example, for the irradiation at 18.9 MeV, apart from the Hf foil, additional foils of natural Tl, ^{74}Ge and ^{72}Ge (isotopically enriched targets) were studied in parallel, using one Au and three Al foils as reference. The detailed setup is illustrated in Fig. 1. Results from the measurements of Tl and Ge isotopes are also presented in the proceedings of the ND2022 conference.

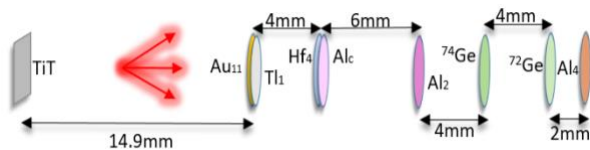


Fig.1. Schematic representation of the target foil assembly for the irradiation at 18.9 MeV, as described in the text.

The detailed geometry of this foil setup along with the target holder and the flange with the TiT target, was introduced in the MCNP5 input file and the resulting simulated neutron fluences at the positions of the Hf target and reference foils are presented in Fig.2 as purple squares. The experimental neutron fluence deduced from the reference foils (one Au and three Al foils) are also shown in Fig.2 as black circles. The fair agreement between the simulated and experimental fluences, within their statistical uncertainties, verifies the validity of the simulations. Thus, the simulated neutron fluence has been used for the determination of the cross section for the Hf isotopes as well as for the other targets of the assembly (natural Tl and $^{72,74}\text{Ge}$) under investigation.

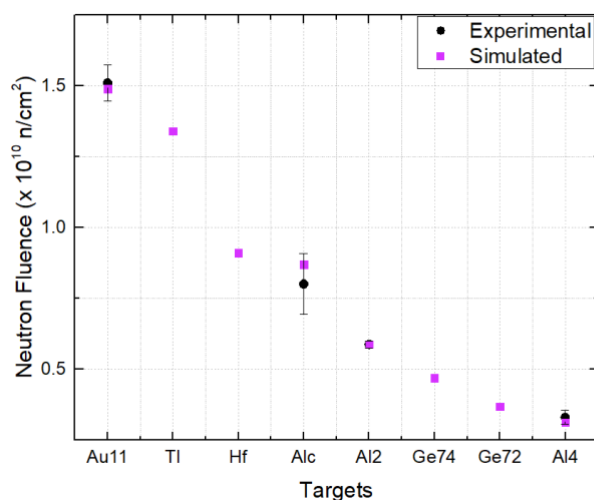


Fig. 2. Experimental fluences in the reference and target foils (shown in Fig.1) along with the simulated ones.

Similar methodology has been successfully used in the rest of the irradiations of this work.

The activation measurements involved continuous irradiation of the samples for about 24-48 h at different neutron energies. For the monitoring of the neutron flux, a BF_3 detector was placed at a distance of 3 m from the target assembly and its spectra were saved at regular time intervals ($\sim 300\text{s}$) in a separate ADC during the irradiation. The beam fluctuations were taken into account in the offline analysis to correct for the decay of the product nuclei during the irradiation.

After the termination of the irradiation phase, the activity of the sample and reference targets was measured off-line by three properly shielded HPGe detectors of 80%, 56% and 16% efficiency, respectively. The absolute efficiency of each detector was obtained separately, by using a calibrated ^{152}Eu point source, placed at the same distance as the samples. The activity measurements of all samples were carried out at a distance of 10 cm from the detector window, thus there was no need for significant pile-up or true coincidence summing-effect corrections. The yield of the γ -ray transitions was corrected for self-absorption and counting geometry of the emitted γ -rays in the activated sample using Monte Carlo simulations by means of the MCNP5 code.

3 Data analysis and corrections

Natural Hf consists of five stable isotopes ^{176}Hf , ^{177}Hf , ^{178}Hf , ^{179}Hf , ^{180}Hf (with $\sim 5\%$, 19% , 27% , 14% and 35% abundances, respectively) and one radioactive ^{174}Hf (abundance: $<0.2\%$) with estimated half-life $t_{1/2} = 2 \times 10^{15} \text{y}$. Of all these isotopes, two neutron induced reactions, the $^{176,174}\text{Hf}(n,2n)$ ones, produce reasonably long lived residual nuclei and will be studied in the present work by using the neutron activation technique.

3.1 The residual nuclei

^{174}Hf is the lowest abundant isotope (0.162%), but the $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ reaction has a high cross-sectional value and could thus be measured in the present work. The residual nucleus ^{173}Hf has spin and parity $1/2^-$ and half-life of 23.6 h, and it decays by electron capture to ^{173}Lu . The characteristic γ -ray transitions 123.7 keV (with 83.3% intensity) and 297 keV (with 33.9% intensity) from the de-excitation of ^{173}Lu , were both used for the determination of the cross section of the $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ reaction. The weighted average was obtained from the two γ -rays and two cross-section values, respectively.

Concerning the $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ reaction, the residual nucleus ^{175}Hf has spin and parity $5/2^-$ and a half-life of 70 days, and decays by β^+ reaction to ^{175}Lu , whose de-excitation to its ground state involves the characteristic 343.4 keV transition of 84% intensity. These characteristic γ -rays from the two reactions

measured at 15.3 MeV are illustrated in the spectra presented in Fig.3.

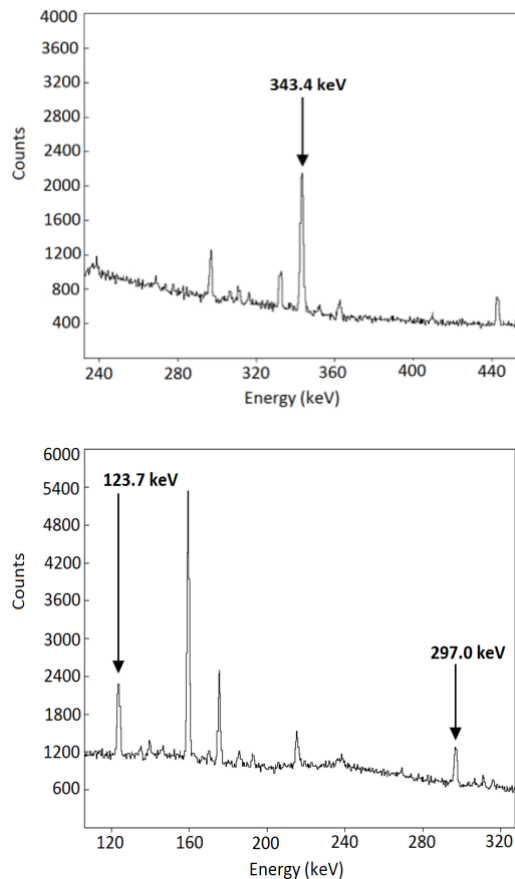


Fig. 3. Off-beam γ - ray energy spectra obtained after the neutron irradiation of natural Hf at 15.3 MeV.

3.2 Corrections for the $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ reaction

The $^{176}\text{Hf}(n,2n)$ reaction leads to the formation of ^{175}Hf isotope which is also produced from the $^{177}\text{Hf}(n,3n)^{175}\text{Hf}$ reaction as well as from the $^{174}\text{Hf}(n,\gamma)^{175}\text{Hf}$ one. Therefore, these contaminations have been taken into consideration and the corresponding corrections have been implemented in the counts of the 343.4 keV characteristic γ -ray.

For the contribution of the $^{174}\text{Hf}(n,\gamma)^{175}\text{Hf}$ reaction, the methodology described in detail in [9] has been used. The resulted correction was irrelevant due to the low abundance of ^{174}Hf , the low (n,γ) cross section at high energies, combined with low flux of parasitic neutrons present in the beam. It should be mentioned that the parasitic neutrons may come from deuteron break-up reactions, from the TiT target itself, from reactions with materials of the beam-line and from scattering in the materials of the room [10].

The contribution of the $^{177}\text{Hf}(n,3n)^{175}\text{Hf}$ reaction was deduced by using the reaction cross section at the main incident neutron beam energies from the ENDF evaluations [6]. The two data points by Semkova et al. [11] for the $^{177}\text{Hf}(n,3n)^{175}\text{Hf}$ reaction are in agreement with the ENDF/B-VIII.0 values, thus ensuring the reliability of these evaluations and confirm the validity of the correction. The corresponding contribution of the $^{177}\text{Hf}(n,3n)^{175}\text{Hf}$ reaction was quite important,

varying from 4% at 15.3 MeV to 70% at 18.9 MeV, due to the high $(n,3n)$ cross section at this energy, introducing significant uncertainty in the determination of the $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ cross section.

3.3 Cross section results

The preliminary results for the experimental cross section of the $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ and $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ reactions, are presented in Fig. 4 as black points, along with data from literature, ENDF evaluations [6] and theoretical predictions, described in the next section. It is interesting to notice that the general trend of the evaluations underestimates the data at high energies, revealing the importance of new experimental data in this energy region.

The experimental data of both reactions are in agreement with previous measurements by Semkova et al. [11] that have been deduced implementing isotopically enriched targets. These data are the only ones available in the high energy region, above 15 MeV [12], while for ^{176}Hf there is also one more data point at 18 MeV by Lu Hanlin et al. [13], in fair agreement with the rest of the existing data, within their experimental uncertainties.

For the estimation of the experimental errors, shown in Fig. several sources of uncertainties were taken into account. More specifically, the determination of the efficiency of the HPGe detectors and target masses, statistical errors of the photopeak areas, errors in the emission probabilities and also the errors in the reference reactions' cross sections. In addition, the corrected cross sections of the $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ reaction include an uncertainty of up to 8% (for higher energies) to account for the inherent assumptions of the correction method.

4 Theoretical calculations

Theoretical cross-section calculations of the $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ and $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ reactions were performed in the 8-20 MeV energy range, according to the Hauser-Feshbach compound nucleus theory using the EMPIRE code (3.2.3 version). Several combinations of nuclear level density models (NLD) and optical model potentials (OMP) 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 $^{174,176}\text{Hf}$ isotopes were achieved by using the optical model potentials for outgoing neutrons by F.D.Becchetti, Jr. and G.W.Greenlees [14] and the Enhanced Generalized Superfluid Model [15] for nuclear level densities. These results are presented in Fig. 4 as red lines, along with all existing experimental data including the present data.

Theoretical calculation results satisfactorily reproduce the experimental data for $(n,2n)$ reactions on both $^{174,176}\text{Hf}$ isotopes in the low energy region, however, this is not the case at higher neutron energies.

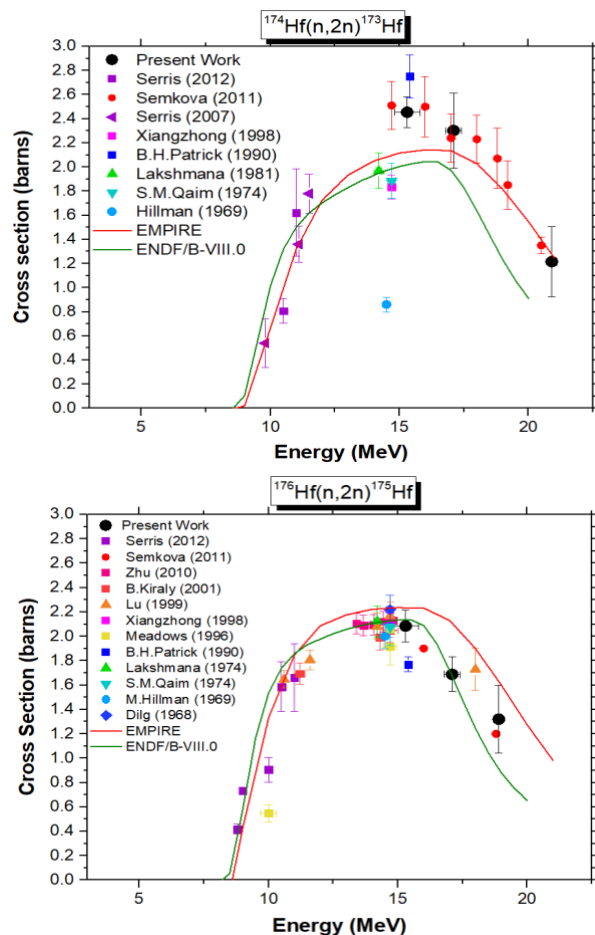


Fig. 4. Cross section data from the present work and EXFOR database of the $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ and $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ reactions. Theoretical calculations with the EMPIRE 3.2.3 code are also shown as red lines while the green lines represent the ENDF/B-VIII.0 evaluation.

More specifically, concerning the $^{174}\text{Hf}(n,2n)$ reaction (Fig. 4, top subfigure), the EMPIRE results significantly underestimate the cross section for neutron energies above 13 MeV. As far as the $^{176}\text{Hf}(n,2n)$ theoretical cross section results are concerned (Fig. 4, bottom subfigure), there is a quite good agreement with the experimental data at the plateau region, however, above 15 MeV the theoretical calculations seem to overestimate the cross section (opposite trend compared to the EMPIRE results for the $^{174}\text{Hf}(n,2n)$ reaction cross section). It is clear that further theoretical investigation is needed in order to achieve better agreement between experimental data and theoretical calculations for both reactions.

5 Summary & conclusions

In this work, preliminary results of cross section measurements are presented for the $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ reaction at 15.3, 17.1 and 18.9 MeV and for the $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ reaction at 15.3, 17.1 and 20.3 MeV. The activation technique has been used, with neutron beams produced via the $^3\text{H}(d,n)^4\text{He}$ reaction at the 5.5-MV Tandem accelerator laboratory of the National Centre for Scientific Research (NCSR) "Demokritos". Additional measurements are planned in the near future

at energies between 15 and 20 MeV to cover this important energy region and to provide more experimental information which will help to improve evaluations and theoretical calculations.

In addition, theoretical calculations based on the compound nucleus theory of Hauser-Feshbach, have been performed in the ~ 8 -20 MeV energy range, using the EMPIRE 3.2.3 code. Although the theoretical calculation results satisfactorily reproduced the experimental data of both studied reactions in the low energy region, they significantly underestimated and overestimated the data at higher neutron energies for the $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ and $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ reactions, respectively. Thus, further theoretical investigation is needed in order to draw firm conclusions for the simultaneous reproduction of both reactions along with other exit channels which will be included in the calculations. The pre-equilibrium emission mechanism, which is very important at energies above 15 MeV, will also be thoroughly investigated, in an attempt to meliorate the theoretical results in this energy region.

Acknowledgments

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