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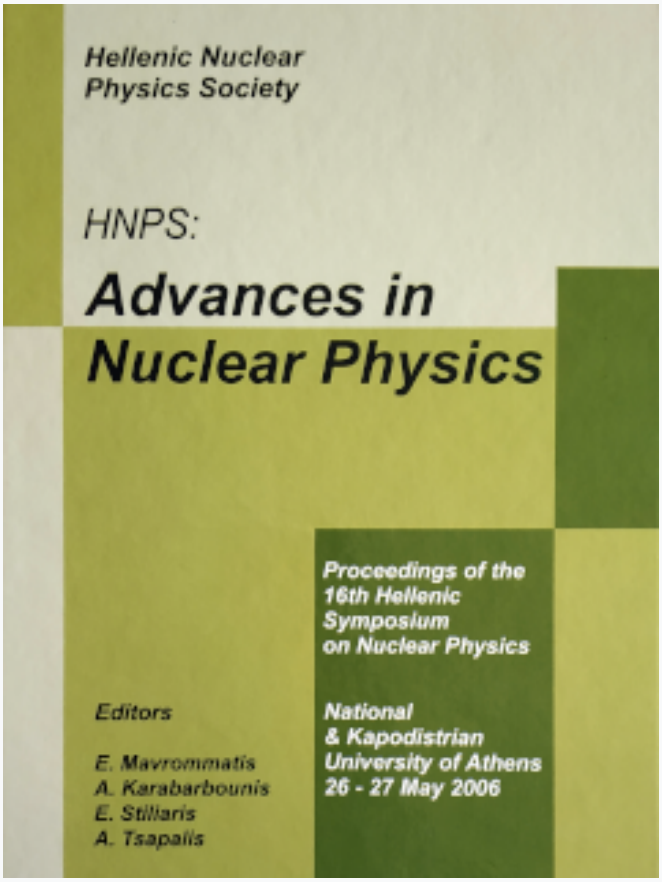


# Study of the (n,2n) reaction cross section on 174,176Hf isotopes

*M. Serris, et al.*

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# Study of the (n,2n) reaction cross section on $^{174,176}$Hf isotopes

M. Serris[a], S. Galanopoulos[a], C.A. Kalfas[b], M. Kokkoris[a], A. Lagoyannis[b], C.T. Papadopoulos[a], N. Patronis[a], G. Perdikakis[a,b] and R. Vlastou[a]

[a]National Technical University of Athens, Zografou Campus, 157.80 Athens, Greece

[b] Institute of Nuclear Physics, NCSR "Demokritos", 153.10 Aghia Paraskevi, Athens, Greece

**Abstract**

Cross section measurements of the $^{174}$Hf(n,2n)$^{173}$Hf and $^{176}$Hf(n,2n)$^{175}$Hf reactions have been performed at the VdG Tandem accelerator of NCSR "Demokritos" in Athens, in the neutron energy region from 8.8 to 11.5 MeV, using the activation technique. Statistical model calculations based on the Hauser - Feshbach theory have also been implemented by using the code EMPIRE-II with different sets of input parameters. The predictions were compared to the data of the present work as well as data from literature.

## 1. Introduction

Hf is widely used in industry due to its excellent mechanical and exceptional resistance properties. Especially the $HfO_2$ is a candidate for future generation insulators of high dielectric constant [1]. Furthermore, in assessing radioactive waste production in proposed fusion reactions, n-induced reactions on W and Ta in reactor materials, could lead to long lived isomeric states of Hf isotopes with rather harmful $\gamma$-ray production [2]. Besides the practical applications, neutron induced reactions play an important role in the investigation of the sensitivity of the input parameters inserting in the Hauser-Feshbach theory at the high A mass region. In the case of the $^{174}$Hf(n,2n)$^{173}$Hf and the $^{176}$Hf(n,2n)$^{175}$Hf reaction, very little information is available around 14 MeV with many discrepancies among the experimental data. In the present work, the cross section of the threshold reaction $^{174}$Hf(n,2n)$^{173}$Hf was measured at neutron beam energies 9.8 to 11.5 MeV where no other data exist in literature, while for the $^{176}$Hf(n,2n)$^{175}$Hf one, only an estimation of the cross section in the same energy range could be achieved with the available facility. Theoretical calculations implementing the code EMPIRE-II have also been performed and compared to the experimental data.

## 2. Experimental setup

Neutron activation measurements of the $^{174}$Hf(n,2n)$^{173}$Hf and $^{176}$Hf(n,2n)$^{175}$Hf reactions were performed in the 5.0 MV VdG Tandem accelerator of NCSR "Demokritos". The

neutron beam was produced via the $^{2}$H(d,n)$^{3}$He reaction, by bombarding a $D_2$ gas target [3,4] with deuteron beams at energies from 8.8 to 11.5 MeV. The quasi-monoenergetic neutron beams were produced at a neutron flux of $\sim 4 \times 10^6$ n/(cm$^2$ · s). The Hafnium targets were natural $HfO_2$ pellets of 1.3 cm diameter with masses varying from 0.667 g to 0.953 g. They were produced by compressing a mixture of high purity (99.9%) powder $HfO_2$ and C in concentrations 88% and 12%, respectively, necessary for the stability of the pellet. Thin matellic targets of high purity $HfO_2$ with masses $\sim$ 0.95 g of the same diameter with $HfO_2$ pellets, were also used for some of the measurements. In order to measure the neutron flux at the target position, reference metallic targets of high purity (Al, Nb and Au) - of the same diameter with the pellets - were placed on the front and the back of the pellets. The targets used in the irradiations were stacked together in an aluminium holder at 0 degrees with respect to the beam direction and at distances which varied from 7 to 12 cm from the end of the deuterium gas cell in order to subtend a small solid angle $\prec 5^o$ and therefore an almost monoenergetic neutron beam. The average effective flux on each sample was obtained by taking the mean of the calculated flux values for the front and back Al foils. The obtained values of the flux from all the reference foils were consistent, but the cross section of Al is given in literature with great accuracy (1.5%) [5] and it was thus chosen for the neutron flux determination. The decay data of the reference and Hf targets are presented in Table 1.

Table 1
Decay data of the $^{174}$Hf (n,2n)$^{173}$Hf and $^{174}$Hf (n,2n)$^{173}$Hf reactions along with those for the reference reactions.

| REACTION | Q (MeV) | $T_{1/2}$ | $E_\gamma$ (keV) | P (%) |
|---|---|---|---|---|
| $^{174}$Hf (n,2n)$^{173}$Hf | -8.51 | 23.6 h | 123.7 | 83 |
| $^{176}$Hf (n,2n)$^{175}$Hf | -8.16 | 70 d | 343.4 | 84 |
| $^{180}$Hf (n,n′ $\gamma$)$^{180}$Hf | | 5.5 h | 332.3 | 94 |
| $^{27}$Al(n,$\alpha$)$^{24}$Na | -3.13 | 14.96 h | 1368.6 | 100 |
| $^{93}$Nb(n,2n)$^{92m}$Nb | -8.83 | 10.15 d | 934 | 99 |
| $^{197}$Au(n,2n)$^{196}$Au | -8.07 | 6.2 d | 355.7 | 87 |

The activation measurements involved a continuous irradiation of the samples for about 48 h, at each neutron energy. For the monitoring of the neutron flux, a $BF_3$ detector was used, placed at a distance of 3 m from the gas cell and at 0 degrees with respect to the beam axis. Details of experimental setup used in this work are presented in ref [4]. After the irradiation, the samples were placed at 7 cm away from the window of HPGe detectors, with 80% or 85% relative efficiency. At this distance acceptable counting rate from the samples and reduced coincidence summing effects, could be achieved. The absolute efficiency of the detector was obtained using a calibrated $^{152}$Eu source placed at the same distance as the samples, while calibrated monoenergetic sources ($^{54}$Mn, $^{60}$Co, $^{56}$Co, $^{137}$Cs and $^{22}$Na) were used at distances of 28 and 7 cm away from the detector front in order to correct the $^{152}$Eu efficiency for summing effects, which did not exceed 6% in the least favourable case.

The determination of the (n,2n) cross section values, resulted by analyzing off line spectra, from the sample and from the reference targets. The yield of the $\gamma$-ray transitions was corrected for self absorption of the emitted $\gamma$-rays in the activated sample by using the code MCNP4C [6]. The corrections were found to be 35% in the case of $E_\gamma$=123 keV and 5% for the $E_\gamma$=343 keV. This result was consistent with the maximum absorption deduced for the $\gamma$-rays from the $^{152}$Eu source with and without the Hf pellet between the source and the HPGe detector.

Among all isotopes comprising natural Hf ($^{180,179,178,176,174}$Hf), only $^{180}$Hf, $^{176}$Hf and $^{174}$Hf(n,2n) reaction studies are in principle favourable using the activation technique, as shown in Table 1. Despite the fact that $^{174}$Hf is the lowest abundant isotope (0.162%), the $^{174}$Hf(n,2n)$^{173}$Hf reaction, is the most prominent one and due to its high energy threshold it cannot be influenced by the existence of low energy parasitic neutrons [4]. The residual nucleus $^{173}$Hf, which is produced via the $^{174}$Hf(n,2n) reaction, has spin and parity $1/2^+$ , half life of 23.6 h and decays by electron capture to the $^{173}$Lu. The characteristic $\gamma$-ray transition 123.7 keV from the de-excitation of $^{173}$Lu, is used for the determination of the cross section of $^{174}$Hf(n,2n)$^{173}$Hf reaction. A typical spectrum taken with the HPGe detector is given in Fig. 1, where the 123.7 keV line is clearly seen along with transitions coming from the reactions of other Hf isotopes as well as from background.

From the 343.4 keV $\gamma$-ray resulting from the $^{176}$Hf(n,2n)$^{175}$Hf reaction, no reliable values of the cross section could be deduced, due to the contamination from the $^{174}$Hf(n,$\gamma$)$^{175}$Hf reaction which is always present and open to low energy parasitic neutrons. In order to determine the contribution of the $^{174}$Hf(n,$\gamma$)$^{175}$Hf reaction in the cross section of $^{176}$Hf(n,2n)$^{175}$Hf, a set of reference foils (Al, Nb, Au) along with a $HfO_2$ target was put inside a Cd cell, which absorbs the thermal neutrons due to the high cross section of n+Cd reaction. The same set of targets was put in front of the Cd cell and the two target configurations were irradiated simultaneously with neutron beam at 11.0 MeV. The cross section value of the $^{176}$Hf(n,2n)$^{175}$Hf reaction obtained with the $HfO_2$ target inside the Cd cell was 15% lower than that without the Cd cell, indicating a significant amount of thermal neutrons in the neutron beam. Similarly, the $^{180}$Hf(n,n$'$ $\gamma$)$^{180}$Hf reaction is contaminated by the thermal neutron tail of the quasi monoenergetic neutron beam, since the threshold of this reaction is low enough to allow low energy parasitic neutrons to contribute to its cross section and induce a considerable uncertainty in the energy determination.

## 3. Data analysis

The cross section formula of a neutron induced reaction at the beam energy $E_n$, is given by the relation

$$\sigma = \frac{N_P}{N_T \Phi} \tag{1}$$

where $N_P$ is the number of the produced nuclei by the reaction during the irradiation, corrected for self absorption, counting geometry and coincidence summing effects, as well as for the decay of product nuclide during the irradiation and the counting collection time. $N_T$ is the number of target nuclei and $\Phi$ is the time integrated flux [4], deduced by

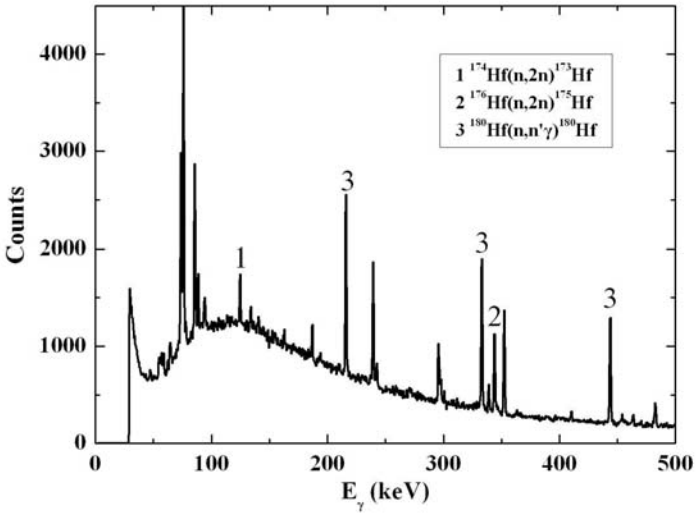


Figure 1. Typical off line spectrum of natural $HfO_2$ taken at neutron energy 11.1 MeV.

the activity of the reference foils and corrected for its fluctuations during the irradiation time.

The uncertainties involved in the cross section determination include errors in the efficiency of the HPGe detector, statistical errors of the photopeak areas, errors of the emission probabilities, errors in the determination of the target masses and errors of the cross section of reference reactions, leading to a total error of the order of 10%. The error of the neutron beam energy was determined by considering the kinematics of the d-d reaction, the actual size and material of the cell window, the pressure of the gas cell and the distance of the samples from the middle of the cell, and was found to be around 0.04 MeV.

## 4. Results and discussion

The experimental data are presented in Fig. 2 and Fig. 3 along with theoretical predictions for the $^{174}$Hf(n,2n)$^{173}$Hf and $^{176}$Hf(n,2n)$^{175}$Hf reactions using the statistical model code EMPIRE-II. For these calculations, the coupled channel code ECIS03 was used, within the Optical Model and Coupled Channels (CC) model, which provide a correct description of the strong population of collective discrete levels in the inelastic scattering, especially in the case of deformed nuclei. The pre-equilibrium mechanisms,

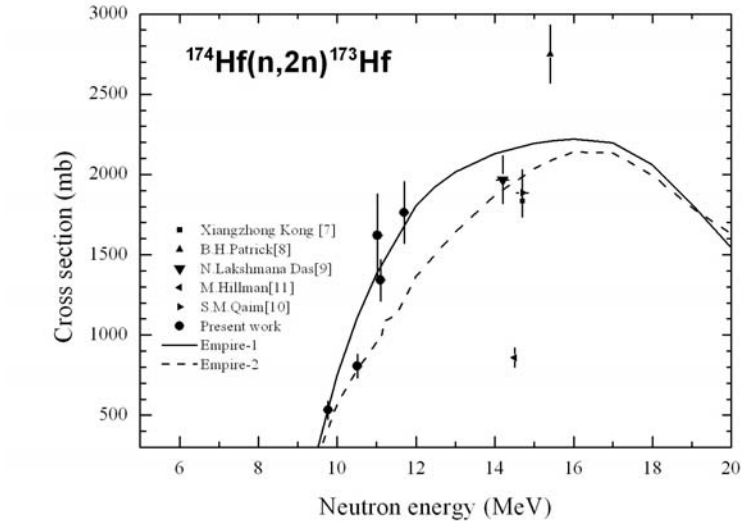


Figure 2. Comparison between measured cross sections and theoretical predictions for the $^{174}$Hf(n,2n)$^{173}$Hf reaction. The circles represent the values of the present work while the other experimental data have been taken from literature. The solid and dashed lines correspond to calculations performed using the code EMPIRE-II with different NLDs (see the text).

which are implemented via the Multi Step Compound (MSC), Multi Step Direct (MSD) [13] and exciton models, were used for the description of the particle and $\gamma$ - rays emission during the evaporation of the compound nucleus. Two different macroscopic Nuclear Level Densities were chosen for the calculations and the results were labelled as Empire 1,2. The first case, Empire-1 has been obtained using the BCS theory [14] along with the Fermi gas model taking into account deformation effects, and the second, Empire-2, corresponds to the NLD of Gilbert - Cameron [16] with the level density parameter $\alpha$ taken from the Ignatyuk's systematics [17].

The measurements of $^{174}$Hf(n,2n)$^{173}$Hf reaction are presented in Fig. 2 along with data from literature [7–11]. For this reaction a few experimental data were available around 14 MeV with many discrepancies among them, while the region near the threshold is covered only by the present data. In Fig. 2, the prediction of Empire-1 seems to better reproduce the data than that of Empire-2 in the region investigated in the present work, thus implying that the deformation of $^{174}$Hf ($\beta$=0.286) plays an important role in the description of neutron induced reactions in the A $\sim$ 180 mass region.

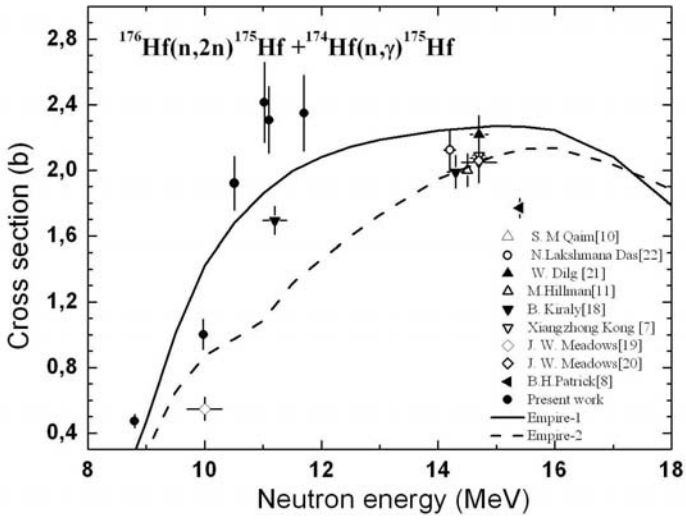


Figure 3. Comparison between measured cross sections and theoretical predictions for the $^{176}$Hf(n,2n)$^{175}$Hf reaction. The circles represent the values of the present work and include contribution from the $^{174}$Hf(n,$\gamma$)$^{175}$Hf reaction, while the other experimental data have been taken from literature. The solid and dashed lines correspond to calculations performed using the code EMPIRE-II with different NLDs (see the text).

In Fig. 3 the data of the present work, data from literature [7,8,10,18–22] along with theoretical calculations are presented. The data of present work are contaminated by the parasitic neutrons reacting with $^{174}$Hf through the (n,$\gamma$) reaction. Thus, our data are higher than the data of ref. [18,19] for the $^{176}$Hf(n,2n) alone. The theoretical calculations of the $^{176}$Hf(n,2n)$^{175}$Hf reaction were deduced using the same set of input parameters as in the case of $^{174}$Hf(n,2n)$^{173}$Hf reaction. The solid line which includes deformation effects ($\beta$=0.277) seem to represent the data of ref. [18,19] rather well, being lower than our data, as expected. The dashed line unterestimates the data as in the case of $^{174}$Hf(n,2n).

## 5. Summary

In the present work, the cross section of $^{174,176}$Hf(n,2n)$^{173,175}$Hf reactions have been measured at neutron beam energies in the range 8.8 to 11.5 MeV, for the first time using the activation technique. The measurements were carried out at the VdG tandem accelerator of NCSR "Demokritos" in Athens, by employing the $^{2}$H(d,n)$^{3}$He reaction. Theoretical

calculations using the code EMPIRE-II were in reasonable agreement with both experimental data measured in the present work and with data obtained from literature. Two different macroscopic models of NLDs were tried. The trend of the prediction which includes deformation effects seems to better reproduce the experimental points of both (n,2n) reactions of $^{174,176}$Hf. More experimental data, however, are needed, in the energy range En=12-17 MeV in order to further test the reliability of the theoretical calculations. In the case of $^{176}$Hf(n,2n)$^{175}$Hf reaction data, it is necessary to further investigate the influence of parasitic neutrons in the cross section values by using the target in Cd - out Cd cell method.

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