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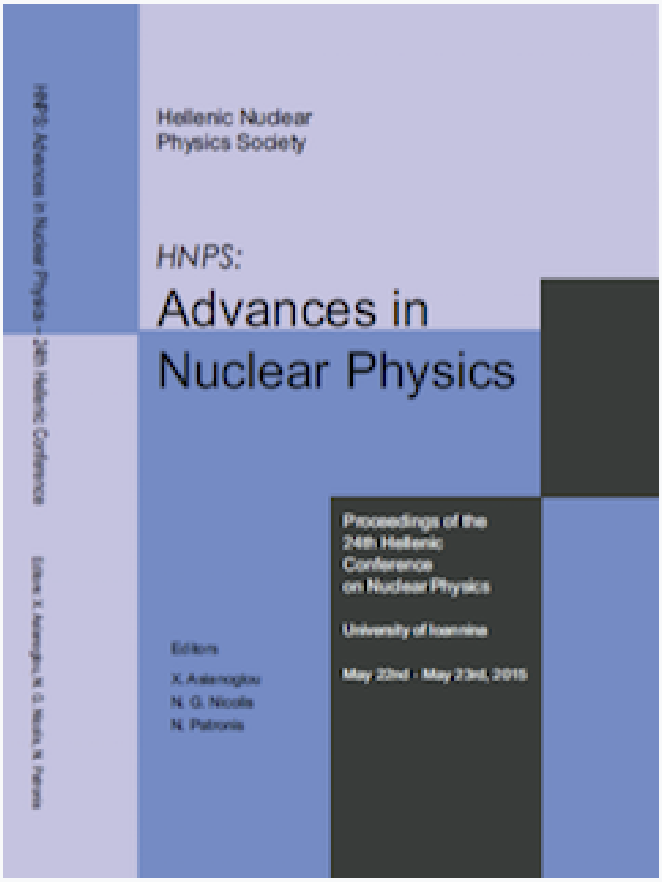

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Activation cross section of the (n,2n) reaction on Hf isotopes

A. Kalamara[1], M.Serris[2], A.Spiliotis[1], D. Sigalos[1], N.Patronis[3], M. Kokkoris[1], M. Diakaki[1,4], M.Axiotis[5], A. Lagoyannis[5] and R. Vlastou[1]

[1]Department of Physics, National Technical University of Athens, 15780 Athens, Greece.
[2] Hellenic Army Academy, 16673 Vari, Athens, Greece.
[3] Department of Physics, University of Ioannina, 45110 Ioannina, Greece
[4]CEA/ Sarlay-DSM, Gif-Sur-Yvette, France.
[4] Institute of Nuclear Physics, NCSR "Demokritos", 15310 Aghia Paraskevi, Greece.

Abstract Cross sections of the ^{174}Hf(n,2n)^{173}Hf and ^{176}Hf(n,2n)^{175}Hf reactions have been experimentally determined relative to the ^{27}Al(n,α)^{24}Na reference reaction at incident neutron energies of 15.3 and 17.1 MeV by means of the activation technique. The irradiations were carried out at the 5 MV tandem T11/25 Accelerator Laboratory of NCSR "Demokritos" with monoenergetic neutron beams provided via the ^{3}H(d,n)^{4}He reaction, using a new Ti-tritiated target of 373 GBq activity. In the determination of the ^{176}Hf(n,2n)^{175}Hf reaction cross section the contamination of the ^{174}Hf(n,γ)^{175}Hf and ^{177}Hf(n,3n)^{175}Hf reactions has been taken into account. Moreover, the neutron beam energy has been studied by means of Monte Carlo simulation codes and the neutron flux has been determined via the ^{27}Al(n,α)^{24}Na reference reaction.

Keywords cross section, activation, Hf isotopes

INTRODUCTION

The study of neutron threshold reactions is of considerable importance for testing and improving nuclear models. Hafnium is widely used in industry due to its excellent mechanical and exceptional resistance properties. For instance, it is used for reactor control rods in nuclear submarines, since apart from the fact that it is extremely corrosion resistant, it has good absorption cross section for thermal neutrons [1]. Furthermore, in assessing radioactive waste production, neutron induced reactions on W and Ta in reactor materials can lead to long-lived isomeric states of Hf isotopes with rather harmful γ radiation (^{178m2}Hf, $T_{1/2}$ = 31y) [2]. In the cases of ^{174}Hf(n,2n)^{173}Hf and ^{176}Hf(n,2n)^{175}Hf reactions, there are only few experimental data in literature with many discrepancies among them. Thus, the purpose of this work was the experimental determination of the cross section of the ^{174}Hf(n,2n)^{173}Hf and ^{176}Hf(n,2n)^{175}Hf reactions at incident neutron energies of 15.3 and 17.1 MeV.

THE EXPERIMENTAL PROCEDURE

The measurements were carried out at the 5.5MV tandem T11/25 Accelerator of NCSR "Demokritos". Quasi-monoenergetic neutron beams were produced via the ^{3}H(d,n)^{4}He reaction, implementing a new Ti-tritiated target of 373 GBq activity, which consist of a 2.1 mg/cm^2 Ti-T layer on a 1mm thick Cu backing for good heat conduction. The 2.0 and 2.5 MeV deuteron beams coming from the accelerator, enter through two 5 μm Mo foils in order to degrade their energy to 0.8 and 1.0 MeV respectively. In this energy region the deuteron beam is feasible to be produced with a current of 1-1.5 μA and the cross section of the $^{3}H(d,n)^{4}He$ reaction is high enough to produce neutron beams at 15.3 and 17.1 MeV at a flux of the order of ~10^5-10^6 n/s·cm^2. The irradiation at 15.3 MeV neutron energy lasted for ~26 h, while the duration of the other at 17.1 MeV was approximately 4 d and the experimental setup for both is presented in the Fig.1.

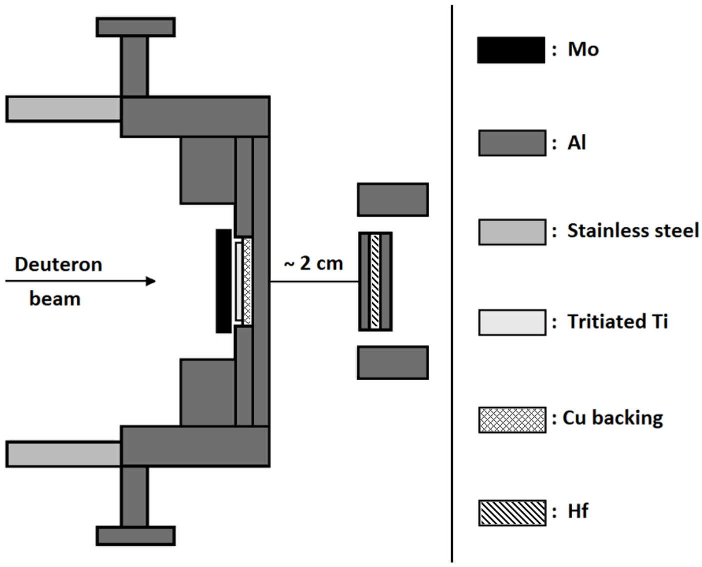


Fig. 1. Schematic representation of the irradiation setup geometry.

The variations of the neutron beams were monitored by a BF3 detector placed at a distance of 3 m from the neutron source. Data from the BF3 counter were stored at regular time intervals through a multichannel scaler, and were used to correct for the decay of the product nuclei during irradiation and to account for fluctuations in the beam flux in the subsequent off-line analysis.

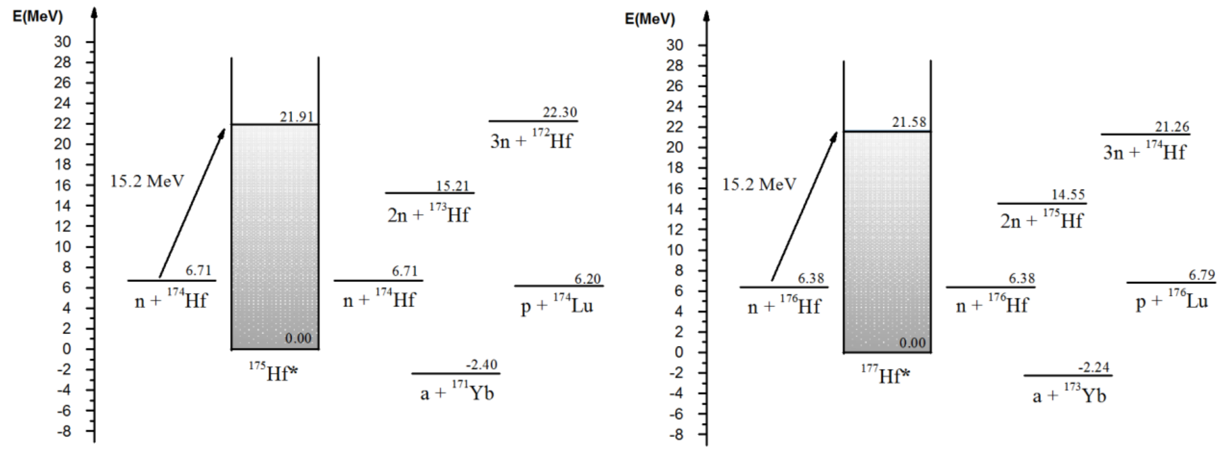
High purity Hf and Al reference foils were placed at a distance of ~2 cm from the neutron beam production (as shown in Fig.1) and the induced activity of product radionuclides from the irradiated foils was measured with two HPGe detectors of 56% and 100% relative efficiency, properly shielded with lead blocks to reduce the contribution of the natural radioactivity. Gamma-ray spectra were taken at a distance of approximately 10 cm from the detectors and their efficiencies were determined via a calibrated ^{152}Eu source. Moreover, corrections for self absorption were taken into account along with the decay of product nuclides over the whole time range and the fluctuation of the neutron beam flux over the irradiation time.

The study of neutron energy spectra generated by deuterons on the Ti-T target were carried out using the code NeuSDesc, developed at IRMM by Birgerssone and Lovestam [3]. The results for the neutron flux at a distance of ~2 cm from the Ti-T target were compared to the MCNP5 [4] simulation results for the neutron flux at the same distance and therefore, the mean neutron beam energies were estimated to be (15.3 ± 0.5) and (17.1 ± 0.3) MeV.

ANALYSIS AND RESULTS

The interactions n+^{174}Hf and n+^{176}Hf are compound nucleus reactions. When a neutron impinge on a ^{174}Hf or a ^{176}Hf nucleus, the compound nuclei ^{175}Hf and ^{177}Hf respectively, are produced in an excited state and the possible exit channels of the reactions are shown in

Fig.2a and 2b. In the present work the $^{174}Hf(n,2n)^{173}Hf$ and $^{176}Hf(n,2n)^{175}Hf$ channels have been investigated, in which the residual nuclei are produced in an excited state.



(a) (b)

Fig. 2. (a) Energy diagram for the n+^{174}Hf interaction. (b) Energy diagram for the n+^{176}Hf interaction.

For the $^{174}Hf(n,2n)^{173}Hf$ reaction the residual nucleus decays by eletron capture to ^{173}Lu and the two most intense characteristic γ-ray transitions of 123.7 keV (83%) and 297.0 keV (34%) were used for the determination of the reaction cross section. A typical spectrum taken with the HPGe detector is given in Fig.3. The yields of the γ-ray transitions were corrected for self-absorption of the emitted γ-rays in the activated samples using the code MCNP5 [4].

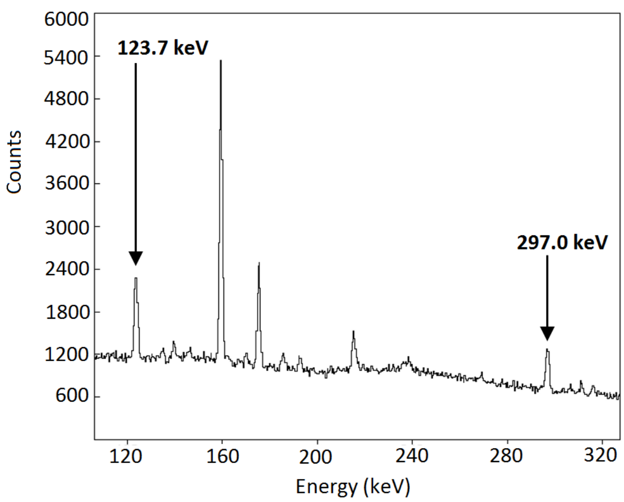


Fig. 3. Typical off line spectrum of natural Hf taken at neutron enery 15.3 MeV in which the γ-ray peaks of interest for the determination of the $^{174}Hf(n,2n)^{173}Hf$ reaction cross section are marked. The acquisition time of the measurement is 108000 sec.

For the $^{176}Hf(n,2n)^{175}Hf$ reaction the residual nucleus decays by β^+ reaction to ^{175}Lu and the characteristic γ-ray transition 343.4 keV cannot be used for the determination of the reaction cross section directly, because is contaminated. The nucleus ^{175}Hf is not only produced by the ^{176}Hf(n,2n) but also by the ^{174}Hf(n,γ) and the ^{177}Hf(n,3n) reactions, thus the intergral of the 343.4 keV γ-ray peak (shown in Fig. 4) must be corrected for the aforementioned contributions.

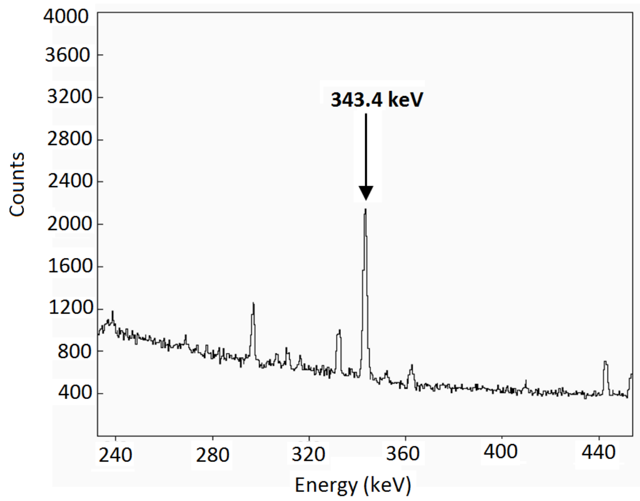


Fig. 4. Typical off line spectrum of natural Hf taken at neutron enery 15.3 MeV in which the γ-ray peak of interest for the determination of the $^{176}Hf(n,2n)^{175}Hf$ reaction cross section is marked. The acquisition time of the measurement is 108000 sec.

The $^{174}Hf(n,\gamma)$ reaction has zero energy threshold ($E_{threshold}$ = 0.0 MeV) which means that can be activated by the presence of low energy parasitic neutrons in the beam. In order to estimate the correction for this contribution a technique, which is described below, was developed.

The neutron beam just after the tritium target was defined with the use of the code NeuSDesc at a distance of 1 mm from the Ti-T target. This distribution was introduced as input source in the code MCNP5 which takes into account the whole geometry of the irradiation setup and therefore, the neutron energy distribution (shown in Fig. 5) at the targets was estimated.

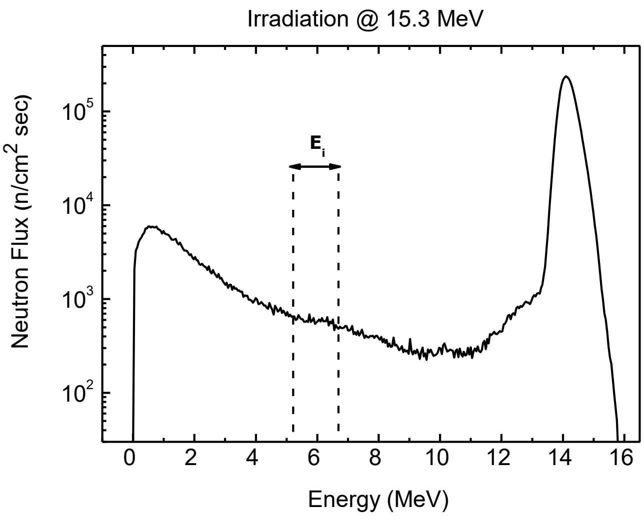


Fig. 5. Neutron flux calculated with NeuSDesc and MCNP codes at a 17 mm distance from the Ti-T target.

Given the neutron flux distribution from the code MCNP5, reaction rate induced at the irradiated foils can be determined according to the following expressions:

$$R.R = \int_{E_{th,i}}^{\infty} \sigma(E_i) \cdot \Phi(E_i)\, dE \quad \Rightarrow \quad R.R = \sum_{E_i} \sigma(E_i) \cdot \Phi(E_i) \qquad (1)$$

where the cross section values (σ) are taken from the ENDF database and the neutron flux ($\Phi(E_i)$) derived from MCNP5 is normalized according to the experimental neutron flux per second for each energy bin E_i (see Fig.5).
In addition, the number of the produced nuclei N_p is given by the equations:

$$N_p = \frac{N_\tau \cdot RR \cdot (1-e^{-\lambda t_B})}{\lambda} \qquad (2)$$

$$N_p = \frac{N_\gamma}{\varepsilon_\gamma \cdot F \cdot I \cdot D} \qquad (3)$$

Thus, according to equations (2) and (3), the number of the expected counts (N_γ) arising from the deexcitation of the residual nucleus can be determined via this expression:

$$N_\gamma = \frac{N_\tau \cdot RR \cdot (1-e^{-\lambda t_B}) \cdot \varepsilon_\gamma \cdot F \cdot I \cdot D}{\lambda} \qquad (4)$$

where ε_γ is the efficiency of the Ge detector, I is the intensity of the characteristic transition, F is the total correction factor to account for self-absorption of the sample and counting geometry, λ is the decay constant of the residual nucleus and

$$D = e^{-\lambda t_1} - e^{-\lambda t_2} \qquad (5)$$

is a correction factor for the counting collection, where t_1 and t_2 are time intervals from the end of the irradiation to the beginning and finish of the measurement with the Ge detector, respectively.

In order to validate this method, the expected counts N_γ for the energy peak at 411.8 keV produced by the residual nucleus of the ^{197}Au(n,γ)^{198}Au reaction were estimated and compared to the experimental ones and the results are presented in Table I.

E_n (MeV)	$N_{expected}$	$N_{experimental}$	Percentage Difference
15.3	22672	26700	15%
17.1	2256	3000	25%

Table. I. Results from the equation (4), $N_{expected}$, compared to experimental data and differeces between them in order to validate the method described above.

The experimental and expected counts are in fair agreement, thus validating the correction methodolody and the MCNP5 predictions.

The same methodology was implemented for the energy peak at 343.4 keV produced by the residual nucleus of the ^{174}Hf(n,γ)^{175}Hf reaction. The results showed that the contribution of the ^{174}Hf(n,γ)^{175}Hf was irrelevant, since the number of expected counts for the peak at 343.3 keV was almost zero. The correction in this case was found to be negligible due to the low (n,γ) cross section, combined with low flux of parasitic neutrons. Nevertheless in other cases the method can be used for this sort of corrections.

Regarding the $^{177}Hf(n,3n)^{175}Hf$ contribution, the correction was deduced by using the reaction cross section at the main incident neutron beam energies from ENDF database and by the following expression:

$$N_{\gamma} = \sigma \cdot N_{\tau} \cdot \Phi \cdot \varepsilon_{\gamma} \cdot I \cdot D \cdot f_c \cdot F \tag{5}$$

where f_c is a correction factor for the produced nuclei that decayed during irradiation and is expressed by:

$$f_c = \frac{\int_0^{t_B} e^{\lambda t} \cdot f(t) \cdot dt}{\int_0^{t_B} f(t) \cdot dt} \cdot e^{-\lambda t_B} \tag{6}$$

where t_B is the irradiation time.

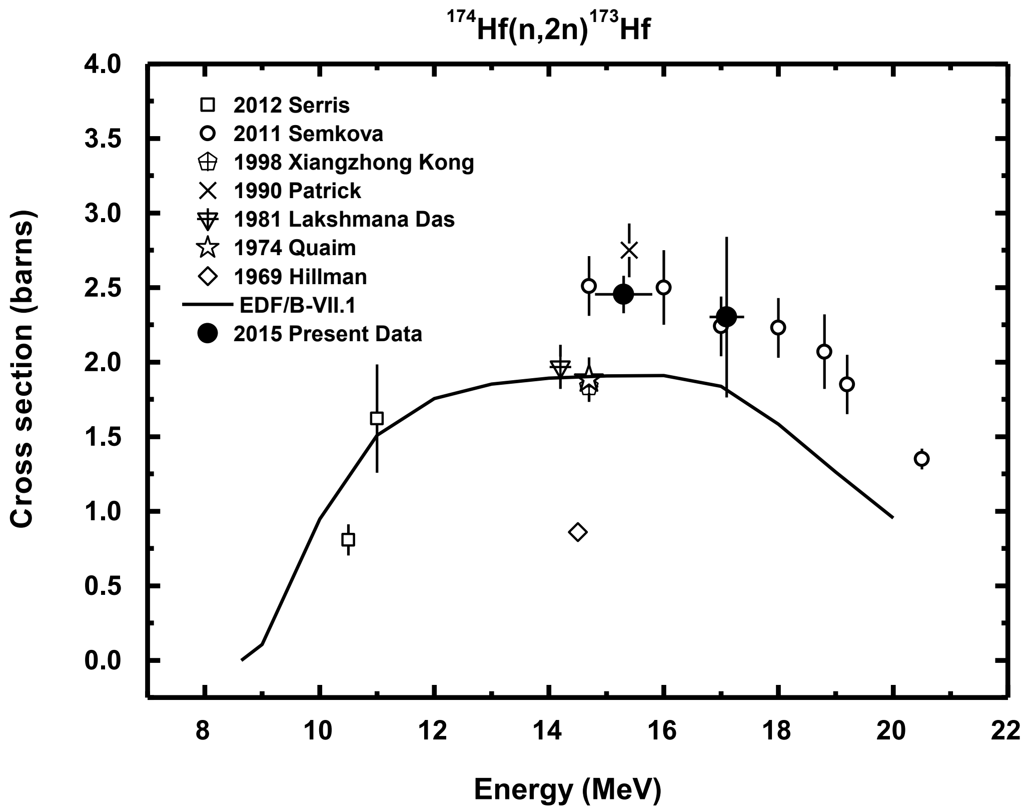


Fig. 6. Cross section data from the present work and EXFOR database of the $^{174}Hf(n,2n)^{173}Hf$ reaction

These corrections turned out to be the major ones. In more detail, at 15.3 MeV irradiation the correction was 311 out of 7500 counts for the 343.4 keV γ-ray peak, while for the 17.1 MeV irradiation was 2460 out of 5400 counts. As expected, at 15.3 MeV neutron beam energy in which the $^{177}Hf(n,3n)^{175}Hf$ reaction channel has just opened and the cross section is very low, the correction is only 4%. On the contrary, at 17.1 MeV in which the (n,3n) cross section is significantly higher the correction reaches up to 46%.

To conclude, the preliminary experimental results for the cross section of the ^{174}Hf(n,2n)^{173}Hf and ^{176}Hf(n,2n)^{175}Hf reactions, are presented in Fig. 6 and Fig. 7, respectively, along with EXFOR data from literature and ENDF/B-VII.1 library [5].

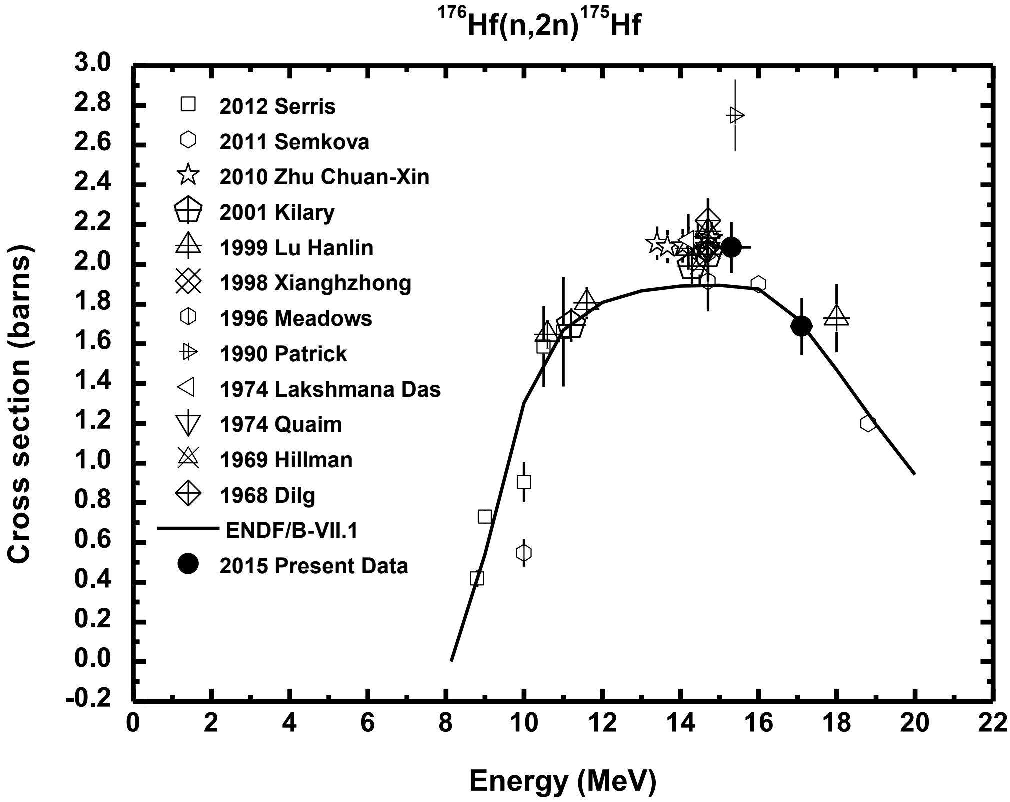


Fig. 7. Cross section data from the present work and EXFOR database of the ^{176}Hf(n,2n)^{175}Hf reaction.

SUMMARY

A new Ti-tritiated target of 373 GBq activity has been installed at the 5.5MV tandem T11/25 Accelerator of NCSR "Demokritos", to produce neutrons in the energy range ~ 15-20 MeV by using the ^{3}H(d,n)^{4}He reaction. The neutron beam energy has been studied by means of Monte Carlo simulation codes and the neutron flux has been determined via the ^{27}Al(n,a) reference reaction. The neutron beams at (15.3±0.5) and (17.1±0.3) MeV have been used for the cross section measurements of the (n,2n) reaction on ^{174}Hf and ^{176}Hf isotopes. In the determination of the ^{176}Hf(n,2n)^{175}Hf reaction cross section the contamination of the ^{174}Hf(n,γ)^{175}Hf and ^{177}Hf(n,3n)^{175}Hf reactions has been taken into account. In particular, a methodology has been developed for the estimation of the (n,γ) contribution and although it proved irrelevant for the ^{174}Hf, it can be useful in other cases for such corrections. Moreover, additional cross section measurements are planned in the 18-20 MeV region and statistical model calculations will be performed by using the EMPIRE code.

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