Study of the (n,2n) cross section of the ^{174}Hf isotope

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Abstract

Cross section measurements of the $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ reaction have been carried out at the VdG Tandem accelerator of NCSR “Demokritos” in Athens, Greece in the neutron energy range 9.8–11.5 MeV, using the activation technique. Statistical model calculations based on the Hauser–Feshbach theory have also been performed by using the code EMPIRE-II with different sets of input parameters. The predictions were compared with the data of the present work as well as with data from literature.

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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. Moreover, the element has a good absorption cross section for thermal neutrons which is almost 600 times that of Zr, has excellent mechanical properties and is used for reactor control rods as for example in nuclear submarines [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 γ -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 $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ 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 $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ was measured at neutron beam energies 9.8, 11.1 and 11.5 MeV where no other data exist in literature. Theoretical calculations implementing the code EMPIRE-II are also presented.

2. Experimental setup

Neutron activation measurements on the $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ reaction were performed in the 5.0 MV VdG Tandem accelerator of NCSR “Demokritos”, Athens, Greece, using neutrons at the energy range from 9.8 to 11.5 MeV. The neutron beam was produced via the $^2\text{H}(d,n)^3\text{He}$ reaction, by bombarding the D_2 gas target [3,4] with deuteron beam currents, typically kept between 2–6 μA . A 5 μm molybdenum foil served as the entrance window and a Pt foil as the beam stop of the gas cell. During the irradiations the gas target was cooled through a cold air jet, in order to minimize the effect of heating in the deuterium gas pressure, which was continuously controlled through a micrometric valve. Using this setup, quasi-monoenergetic neutron beams at a flux of the order of $4 \times 10^6 \text{ n}/(\text{cm}^2 \text{ s})$ were achieved. The targets used in

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the irradiations were stacked together in an aluminium holder at 0° 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 $\pm 5^\circ$ and therefore an almost monoenergetic neutron beam. 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. 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 average effective flux on each sample was obtained by taking the mean of the calculated flux values for the front and back Al foils, since the obtained values of the flux from all the reference foils were consistent while the cross section of Al is given in literature with great accuracy (1.5%) [5]. The decay data of the reference and Hf targets are presented in Table 1.

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° with respect to the beam axis. The data from the BF_3 detector were registered in a separate ADC every 100 s, during the whole period of irradiation. This neutron flux history file was used in the analysis, for off-line correction of the fraction of ^{173}Hf nuclei, which had already decayed during activation. After the irradiation, the samples were placed at 7 cm away from the window centre of a high volume HPGe detector, with 80% relative efficiency. The choice of this distance was the result of the compromise between assuring acceptable counting rate from the samples and reducing coincidence summing effects. The absolute efficiency of the detector was obtained using a calibrated ^{152}Eu source placed at the same distance as the samples. The same procedure was repeated by using calibrated monoenergetic sources (^{54}Mn , ^{60}Co , ^{56}Co , ^{137}Cs and ^{22}Na) at distances of 28 and 7 cm away from the detector front in order to correct the ^{152}Eu efficiency for summing effects. Thus, the overall error in the efficiency measurements 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 γ -ray transitions was corrected for self absorption of the emitted γ -rays in the activated sample by using the code MCNP4C [6] and the geometry of the real measurement. The correction was found to be 35% due to the low energy (123 keV) of the γ -ray emitted inside the activated Hf sample. This result was consistent with the maximum absorption deduced for the γ -rays from the ^{152}Eu source with and without the Hf pellet between the source and the HPGe detector.

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

Reaction	Q (MeV)	$T_{1/2}$	E_γ (keV)	p (%)
$^{174}\text{Hf}(n,2n)^{173}\text{Hf}$	−8.64	23.6 h	123.7	83
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	−3.13	14.96 h	1368.6	100
$^{93}\text{Nb}(n,2n)^{92m}\text{Nb}$	−8.82	10.15 d	934	99
$^{197}\text{Au}(n,2n)^{196}\text{Au}$	−8.07	6.2 d	355.7	87

Fig. 1. Representation of the excitation and evaporation of the ^{175}Hf nucleus.

3. The reaction $^{174}\text{Hf}(n, 2n)^{173}\text{Hf}$

Among all isotopes comprising natural Hf (^{180} , ^{179} , ^{178} , ^{176} , ^{174}Hf), only ^{180}Hf , ^{176}Hf and $^{174}\text{Hf}(n, 2n)^{173}\text{Hf}$ reaction studies are in principle favourable using the activation technique. Despite the fact that ^{174}Hf is the lowest abundant isotope (0.162%), the $^{174}\text{Hf}(n, 2n)^{173}\text{Hf}$ reaction, is the most prominent one, mainly due to its high energy threshold. Thus its study cannot be influenced by the existence of low energy parasitic neutrons, originating from the occurring reactions of the deuteron beam with the gas cell entrance window or beam stop, from deuteron break-up or from low-energy neutrons scattered by the surrounding materials [4]. The residual nucleus ^{173}Hf , which is produced via the $^{174}\text{Hf}(n, 2n)$ reaction, has spin and parity $1/2^+$, half life of 23.6 h and decays by electron capture to the ^{173}Lu , as shown in Fig. 1. The characteristic γ -ray transition 123.67 keV from the de-excitation of ^{173}Lu , is used for the determination of the cross section of $^{174}\text{Hf}(n, 2n)^{173}\text{Hf}$ reaction. A typical spectrum taken with the HPGe detector is given in Fig. 2, where the 123.67 keV line is clearly seen along with transitions coming from the reactions of other Hf isotopes as well as from background. From the γ -rays resulting from the $^{176}\text{Hf}(n, 2n)^{175}\text{Hf}$ reaction, no reliable values of the cross section could be deduced, due to the contamination from the $^{174}\text{Hf}(n, \gamma)^{175}\text{Hf}$ reaction which is always present. Similarly, the $^{180}\text{Hf}(n, n'\gamma)^{180}\text{Hf}$ reaction is contaminated by the thermal neutron tail of the quasi monoenergetic neutron beam.

4. Data analysis

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

$$\sigma = \frac{N_p}{N_T \Phi}, \quad (1)$$

Fig. 2. Typical off line spectrum of natural HfO_2 taken at neutron energy 11.1 MeV.

where N_p is the number of the produced nuclei by the reaction during the irradiation, N_T is the number of the target nuclei and is the integrated over the irradiation time t_B neutron flux $f(t)$ which is provided by the expression

$$\Phi = \int_0^{t_B} f(t) dt \quad (2)$$

N_p can be extracted from the number of gamma ray events N_γ registered in the Ge detector, by the expression

$$N_p(t_B) = \frac{N_\gamma}{\varepsilon F I_\gamma D f_C}, \quad (3)$$

where ε is the peak efficiency of the measured γ -ray and I_γ its transition probability. F is the total correction factor of the activity to account for self absorption of the sample, coincidence summing effects of cascading gamma rays and counting geometry. $D = [\exp(-\lambda t_1) - \exp(-\lambda t_2)]$, where t_1 and t_2 are time intervals from the end of the irradiation and the start and finish of the measurement, correspondingly and λ is the decay constant of the residual nucleus. f_C is the fraction of ^{173}Hf nuclei which are decaying during the irradiation and is expressed by the formula

$$f_C = \frac{e^{-\lambda t_B} \int_0^{t_B} e^{\lambda t} f(t) dt}{\int_0^{t_B} f(t) dt}$$

The cross section measurements include the following errors, the error in the efficiency ($\sim 4\%$) of the HPGe detector, the statistical error of the photopeak area, the error of the emission probabilities, the errors in the determination of the target masses ($\sim 2\%$) and the error of the cross section of reference reactions ($\sim 1.5\%$) 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.

5. Results and discussion

The measurements are presented in Fig. 3 along with data from literature [7–11] which are scattered around 14 MeV. Theoretical calculations using the statistical model code EMPIRE-II [12] were performed taking into consideration appropriate pre-equilibrium reaction mechanisms. More specifically, the coupled channel code ECIS03 was used for calculations 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, 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 γ -rays emission during the evaporation of the compound nucleus. The dominant input

Fig. 3. Comparison between measured cross sections and theoretical predictions for the $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ 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 text).

parameters entering in the statistical model calculations at energies up to some tens of MeV are the optical model potential (OMP), and nuclear level density (NLD). In the present work, the sensitivity of the calculations was tested using two different macroscopic NLDs, and the corresponding calculations, labelled as Empire-IIa,IIb, are shown in Fig. 3. In this figure, the solid line, Empire-IIa, has been obtained using the BCS theory [14] along with the Fermi gas model taking into account deformation effects. In this approach the low energy part of the level densities is calculated in terms of the superfluid BCS model [15]. The dashed line, Empire-IIb, corresponds to the NLD of Gilbert–Cameron [16] with the level density parameter α taken from the Ignatyuk’s systematics [17].

The prediction of Empire-IIa seems to reproduce better the data than that of Empire-IIb 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. Further tests, however, are needed before a firm conclusion is reached. As we note from Fig. 3, in the energy region from 9 to 16 MeV, both predictions from Empire-IIa and Empire-IIb exhibit a similar behaviour with energy, reaching the maximum plateau around 15 MeV. They reveal, however, a quantitative difference of about 25%, while at higher energies (>16 MeV) they tend to agree. Due to the lack of experimental data at energies up to 20 MeV and to the discrepant data at 14 MeV, in order to check the reliability of the input parameters entered in calculations, it is necessary to perform additional cross section measurements of this reaction at higher energies.

6. Summary

In the present work, the cross section of $^{174}\text{Hf}(n,2n)^{173}\text{Hf}$ reaction has been measured at neutron beam energies 9.8, 11.1 and 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, Greece by employing the $^2\text{H}(d,n)^3\text{He}$ reaction. Theoretical calculations using the code EMPIRE-II were in good agreement with both experimental data measured in the present work and with data obtained from literature, with the exception of only two data points centered around 14 MeV. Two different macroscopic models of NLDs were tried. The trend of the prediction which includes deformation effects seems to better reproduce the experimental points. More experimental data, however, are needed, in the energy range $E_n = 12$ –17 MeV in order to further test the reliability of the theoretical calculations.

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