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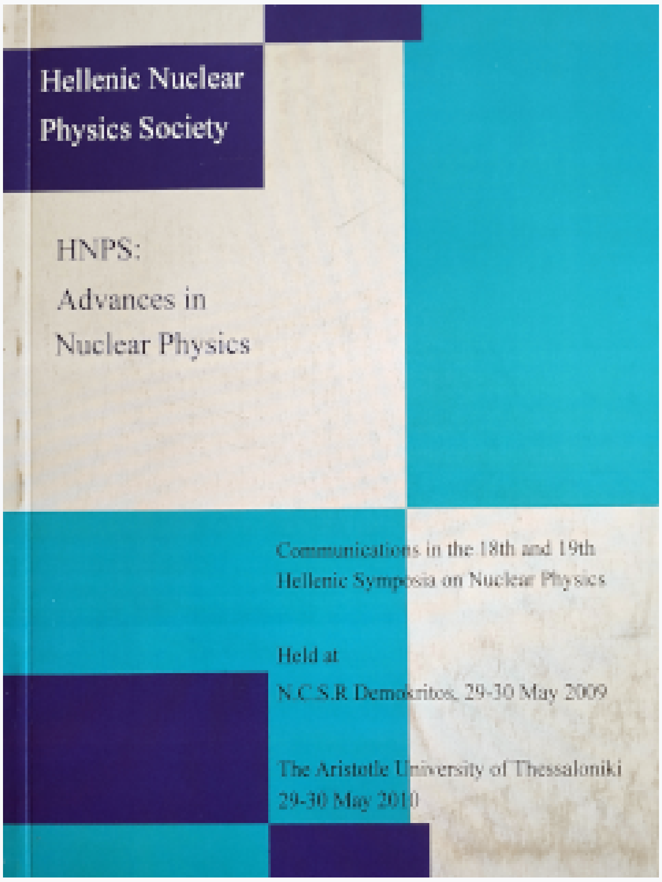

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Influence of parasitic neutrons to the ^{176}Hf(n,2n)^{175}Hf reaction cross section

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Abstract

Cross sections for ^{176}Hf(n,2n)^{175}Hf reaction have been measured at the VdG Tandem accelerator of NCSR \Demokritos", in the neutron energy region from 8.80 to 11.02 MeV, using the activation technique. In order to account for the contamination of the ^{176}Hf(n,2n)^{175}Hf by the ^{174}Hf(n,γ)^{175}Hf reaction activated by the presence of parasitic neutrons, an experimental method has been developed based on the influence of the parasitic neutrons in the case of the (n,2n) and (n,γ) reactions on ^{197}Au. The results were found to be consistent with the energy spectrum of the neutron beam which has been studied by means of the multiple foil activation analysis technique as well as by a liquid scintillator BC501A detector and subsequent deconvolution of its recoil energy spectra.

Introduction

Studies of neutron induced reactions are of considerable significance, both for their importance to fundamental research in Nuclear Physics and Astrophysics and for practical applications in nuclear technology, medicine and industry. In the energy region up to 20 MeV, many reaction channels, which may proceed via different reaction mechanisms, are open and therefore can be simultaneously studied both experimentally and theoretically. In particular, neutron induced reactions on Hf are important since Hf is used for reactor control rods in nuclear submarines due to its high thermal neutron absorption cross section. 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. In the case of the ^{176}Hf(n,2n)^{175}Hf reaction, experimental data are available only at ~14MeV with many discrepancies among them.

Measurements of neutrons on natural Hf for the determination of the ^{176}Hf(n,2n)^{175}Hf cross section, are influenced by the presence of parasitic neutrons which accompany the main neutron beam and activate the ^{174}Hf(n,γ)^{175}Hf reaction. In order to account for this contamination of the ^{175}Hf production, a method has been developed, based on the investigation of the parasitic neutrons in the case of the (n,2n) and (n,γ) reactions on ^{197}Au. These two reactions produce different daughter nuclei ^{196}Au and ^{198}Au, respectively, thus allowing the comparison between the useful neutrons which activate the threshold (n,2n) reaction and the parasitic ones which activate the (n,γ) reaction.

Experimental Procedure

Cross section measurements of (n,2n) threshold reactions on high purity natural Hf have been performed in the energy range 8.80 -11.02 MeV, at the 5 MV Tandem T11/25 accelerator laboratory of NCSR "Demokritos", by using the activation method. Quasi-monoenergetic neutron beams were produced via the $^{2}H(d,n)^{3}He$ reaction at a flux of the order of ~10^6 n/(cm^2 sec). Thin metallic targets of high purity natural Hf with masses ~0.95 g were used, stacked between two Al foils and placed at 0 degrees with respect to the beam direction and at distances which varied from 7 to 12cm from the end of the deuterium gas cell. The absolute flux of the beam was obtained with respect to the $^{27}Al(n,\alpha)^{24}Na$ reference reaction, while its variation was monitored by a BF_3 detector placed at a distance of 3 m from the neutron source. The average effective flux on each sample was obtained by taking the mean values for the front and back Al foils. The induced activity of product radionuclides and reference foils was measured with HPGe detectors of 80% and 56% efficiency. The characteristic γ-rays were corrected for self absorption in the target, summing effects of cascading transitions and counting geometry.

The neutron beam however, is not purely monoenergetic due to parasitic neutrons mainly originating from the deuteron break up reactions. In order to study the influence of the parasitic neutrons on the main neutron beam, an investigation of the energy dependence of the neutron flux has been carried out [1] by using two experimental methods: deconvolution of recoil energy spectra taken with a liquid scintillator BC501A detector, with the DIFBAS code [2], as well as via the multiple foil activation technique in combination with the SULSA [3] unfolding code. The results from the two methods seem to be in fair agreement, indicating that around nominal neutron energies E_n = 7 MeV, parasitic neutrons start to appear and above E_n = 9 MeV, they contribute considerably to the neutron flux. The tail of parasitic neutrons is important in the region 2-3 MeV, mainly due to the deuteron break-up process above E_d = 4.5 MeV [4].

Natural Hf consists of 6 isotopes $^{174, 176, 177, 178, 179, 180}$Hf and three of them produce long lived residual nuclei which can thus be studied by using the neutron activation technique. The $^{174}Hf(n,2n)^{173}Hf$, $^{176}Hf(n,2n)^{175}Hf$ and $^{174}Hf(n,\gamma)^{175}Hf$ are the three reactions which can be studied with the activation mehod. The $^{174}Hf(n,2n)^{173}Hf$ reaction cross section has already been investigated [5], while the data for the $^{176}Hf(n,2n)^{175}Hf$ reaction is contaminated by the $^{174}Hf(n,\gamma)^{175}Hf$ reaction and needs more consideration. The cross section of the $^{176}Hf(n,2n)^{175}Hf$ reaction can be determined via the 343.4 keV characteristic γ-ray from the deexcitation of the ^{175}Hf. It is a threshold reaction (Ethr=8.2MeV) and opens with neutrons of energy above ~9MeV, while the contaminant reaction $^{174}Hf(n,\gamma)^{175}Hf$ has no threshold and opens with parasitic low energy neutrons. In order to account for this contamination of the ^{175}Hf production, the effect of parasitic neutrons has been studied in the case of the (n,2n) and (n,γ) reactions on ^{197}Au. These two reactions produce different daughter nuclei ^{196}Au and ^{198}Au, respectively, thus allowing the comparison between the useful neutrons which activate the threshold (n,2n) reaction and the parasitic ones which activate the (n,γ) reaction. Furthermore, the cross sections of the $^{174}Hf(n,\gamma)^{175}HF$ and $^{197}Au(n,\gamma)^{198}Au$ reactions, have similar behavior with respect to neutron energy. The technique implemented to perform these corrections is described below.

The correction technique

At the end of the irradiation of the ^{197}Au target, the number of the produced nuclei, is given by the relations:

$$N^{196} = \sigma_{2n} N^{197} \Phi_{2n} f(t)_{2n} \tag{1}$$

$$N^{198} = \sigma_{\gamma} N^{197} \Phi_{\gamma} f(t)_{\gamma} \tag{2}$$

where N^{196} and N^{198} are the numbers of the produced nuclei of ^{196}Au and ^{198}Au respectively, N^{197} is the number of the target nuclei, σ_{2n} and σ_{γ} are the cross sections of the ^{197}Au(n, 2n)^{196}Au and ^{197}Au(n, γ)^{198}Au reactions, respectively, Φ is the neutron flux and f(t) is the correction factor which represents the fluctuations of the neutron beam.

The fraction of the produced nuclei which are decaying during the irradiation is expressed by the formula :

$$f_c(t) = \frac{\int_0^{t_B} f(\tau) e^{\lambda\tau} d\tau}{\int_0^{t_B} f(\tau)} e^{-\lambda t_B} \tag{3}$$

where t_B is the irradiation time and λ is the decay probability for the reaction c. Considering that f(t) remains constant with time we are lead to the relation :

$$f(t) = \frac{\lambda(1-e^{-\lambda t_B})}{t_B} \tag{4}$$

Finally from the relations (1), (2) and (4) we end up to the expression :

$$\Lambda = \frac{\overline{\sigma_\gamma \Phi_\gamma}}{\Phi_{2n}} = \frac{\sigma_{2n} N^{198} \lambda^{196}(1-e^{-\lambda^{196} t_B})}{N^{196} \lambda^{198}(1-e^{-\lambda^{198} t_B})} \tag{5}$$

with the mean values representing the average cross section and neutron flux with respect to the whole neutron energy spectrum which activates the (n,γ) reaction on ^{197}Au. The experimental ratio Λ has been deduced for all the measurements carried out from 8 to 10.5 MeV and is presented in Figure 1.

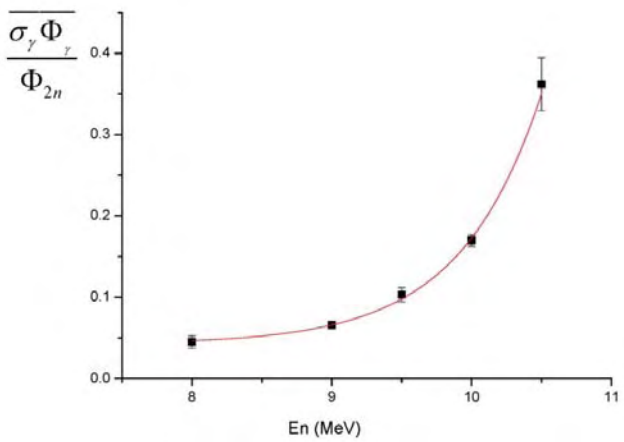

Fig.1. The ratio Λ with respect to the nominal neutron beam energy.

From this figure correction factors, have been extracted for each energy with respect to the 8 MeV value of Λ, energy which corresponds to the threshold where the $^{176}Hf(n,2n)^{175}Hf$ reaction is closed and the production of the 343.4 keV γ-ray derives only from the contaminant (n,γ) reaction. Assuming that these correction factors representing the effect of the contamination of (n,2n) by the (n,γ) reaction, are the same for both Hf and Au, they have been used to extract the corrected values of $^{176}Hf(n,2n)^{175}Hf$ cross sections, as described below.

For energy E_l=8MeV lower than the threshold energy of the $^{176}Hf(n,2n)^{175}Hf$ reaction and for energy E_h higher than the threshold energy of the $^{176}Hf(n,2n)^{175}Hf$ reaction, the number of the parasitic events from the $^{174}Hf(n,\gamma)^{175}Hf$ reaction is given respectively by the relations:

$$C_l^{\gamma} = \overline{\sigma_{Hf}^{\gamma l}} N_{\tau}^{l} \overline{\Phi_l^{\gamma}} f_c^{l} \varepsilon_{\gamma}^{l} I_{\gamma} K_{\gamma} e^{-\lambda t_w^{l}} \left(1 - e^{-\lambda t_m^{l}}\right) \tag{6}$$

and

$$C_h^{\gamma} = \overline{\sigma_{Hf}^{\gamma h}} N_{\tau}^{h} \overline{\Phi_h^{\gamma}} f_c^{h} \varepsilon_{\gamma}^{h} I_{\gamma} K_{\gamma} e^{-\lambda t_w^{h}} \left(1 - e^{-\lambda t_m^{h}}\right) \tag{7}$$

where N_τ is the number of the target nuclei, ε_γ is the peak efficiency of the measured γ-ray and I_γ its transition probability, K_γ is the correction factor of the activity to account for self absorption of the sample, t_w is the waiting time between the end of the irradiation and the start of the measurement and t_m is the measurement time.

Dividing relations (6) and (7) by parts and multiplying and dividing the numerator and the denominator with the flux Φ, we end up to the expression:

$$C_h^{\gamma} = C_l^{a} \frac{\overline{\dfrac{\sigma_{Hf}^{\gamma h} \Phi_h^{\gamma}}{\Phi_{2n}^{h}}} \Phi_{2n}^{h} N_{\tau}^{h} f_c^{h} \varepsilon_{\gamma}^{h} I_{\gamma} K_{\gamma} e^{-\lambda t_w^{h}} \left(1 - e^{-\lambda t_m^{h}}\right)}{\overline{\dfrac{\sigma_{Hf}^{\gamma l} \Phi_l^{\gamma}}{\Phi_{2n}^{l}}} \Phi_{2n}^{l} N_{\tau}^{l} f_c^{l} \varepsilon_{\gamma}^{l} I_{\gamma} K_{\gamma} e^{-\lambda t_w^{l}} \left(1 - e^{-\lambda t_m^{l}}\right)} \tag{8}$$

The values of the fractions $\dfrac{\overline{\sigma_{Hf}^{\gamma} \Phi_h^{\gamma}}}{\Phi_{2n}}$ in the previous expression, for each energy, are taken from Figure 3, assuming that is the same for the (n,2n) and (n,γ) reactions on both Hf and Au.

Results

The measured cross sections of the $^{176}Hf(n,2n)^{175}Hf$ reaction at the neutron energies of 8.8, 9, 9.975, 10.51 and 11.023 MeV are presented in Table 1 and in Figure 2. The black squares represent the values of the $^{176}Hf(n,2n)^{175}Hf$ cross section and include the contribution of the $^{174}Hf(n,\gamma)^{175}Hf$ reaction, while the red circles represent the corrected values, determined by the technique described above. It should be emphasized that these are preliminary results and that the errors represent only the statistical part and not the systematic errors of this method.

Table 1
Cross section Values of the ^{176}HF(n,2n)^{175}HF reaction

Neutron Energy (MeV)	Cross Sections including (n,γ) contamination (barn)	Corrected Cross Sections (barn)
8.8	0.48 ± 0.04	0.45 ± 0.04
9.0	0.76 ± 0.03	0.73 ± 0.03
10.0	1.00 ± 0.08	0.92 ± 0.08
10.5	1.92 ± 0.13	1.75 ± 0.13
11.0	2.42 ± 0.22	2.04 ± 0.24

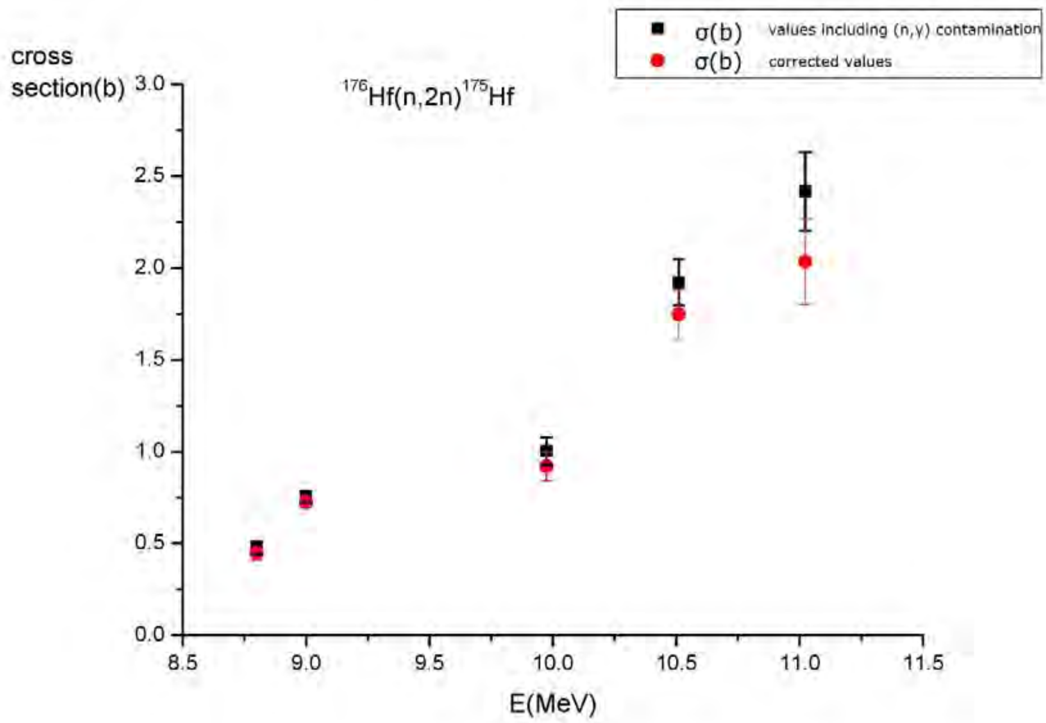

Fig. 2. Cross section Values of the ^{176}Hf(n,2n)^{175}Hf reaction

From the results in Table 1 and in Figure 2, it is apparent that the divergence between the corrected and the uncorrected cross section values increases with neutron energy. This result is consistent with the fact that the amount of the parasitic neutrons in the beam increases with increasing energy of neutron beam.

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