

Cross-sections for formation of $^{178m2}\text{Hf}$ and $^{179m2}\text{Hf}$ through reactions on natural hafnium at neutron energy 14.8 ± 0.2 MeV

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Abstract The cross sections for formation of metastable state of ^{178}Hf ($^{178m2}\text{Hf}$, 574.215 keV, 31 y) and ^{179}Hf ($^{179m2}\text{Hf}$, 362.55 keV, 25.05 d) through reactions induced by 14.8 ± 0.2 MeV neutrons on natural hafnium were measured for the first time. The monoenergetic neutron beam was produced via the $^3\text{H}(d, n)^4\text{He}$ reaction on ZF-300-II Intense Neutron Generator at Lanzhou University. Induced gamma activities were measured by a gamma-ray spectrometer with high-purity germanium (HPGe) detector. Measurements were corrected for gamma-ray attenuations, random coincidence (pile-up), dead time and fluctuation of neutron flux. The neutron fluence were determined by the cross section of $^{93}\text{Nb}(n, 2n)^{92m}\text{Nb}$ reaction. The neutron energy in the measurement were by the cross section ratios of $^{90}\text{Zr}(n, 2n)^{89m+g}\text{Zr}$ and $^{93}\text{Nb}(n, 2n)^{92m}\text{Nb}$ reactions.

Keywords (n, x) Reaction · Cross section · Neutrons · Activation technique · Hafnium

Introduction

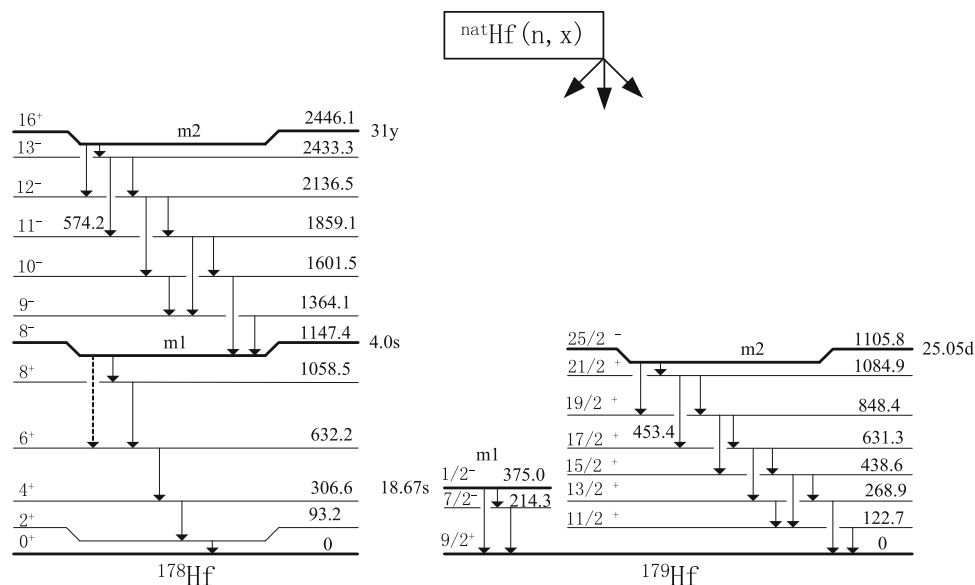
Experimental data of neutron-induced reactions in the energy range around 13 to 15 MeV are needed to verify the accuracy of nuclear models used in the calculation of cross sections. Furthermore, the data are of considerable importance for practical applications, such as for integral calculations on the first wall, blanket and shield of a conceptual fusion power reactor. The data for gas production via neutron induced reactions are of great importance in the domain of fusion reactor technology, particularly of nuclear transmutation rates, nuclear heating and radiation damage due to gas formation. A lot of experimental data on neutron induced cross sections for fusion reactor technology applications have been reported and great efforts have been devoted to compilations and evaluations [1, 2]. In general, cross sections for the formation of isomeric states are more difficult to be predicted than those for the total reaction channels, since more details on the structure of the residual nucleus have to be taken into account (cf. Ref. [3]). The relative probability of forming isomeric states in a nucleus is mainly governed by the spin state values of the levels involved, and the spin distribution of the excited states of the compound nucleus. The high spin value 16^+ and $25/2^-$ of the second isomeric state (m2) of ^{178}Hf and ^{179}Hf (Fig. 1) relative to the corresponding values 0^+ and $9/2^+$ of the ground state (g), offers great sensitivity for the study of the spin distribution of the residual nucleus. The cross sections of (n, 2n), (n, γ), (n, p), (n, d), (n, t) ... reactions for the isotopes of hafnium have been measured by many authors. Here, we adduced several authors [4–33], but the cross sections of $^{\text{nat}}\text{Hf}(n, x)^{178m2}\text{Hf}$ and $^{\text{nat}}\text{Hf}(n, x)^{179m2}\text{Hf}$ reactions have been not reported. Most of the authors neglected the effect of (n, n'), (n, γ) reactions or deducted the effect indirectly when they calculated the cross section of (n, 2n)

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Fig. 1 Simplified representation of formation and decay of the ground state and isomeric states of ^{178}Hf [34] and ^{179}Hf [35]. All energies are in keV



reaction. Thus, we investigate them in order to give the cross section of the element producing certain radioactive product nuclide.

In the present work, the cross-sections of the above mentioned reaction were measured in at neutron energy 14.8 ± 0.2 MeV and a gamma-ray counting technique was applied using gamma-ray spectrometer and data acquisition system. Pure HfO_2 was used as the target material. The reaction yields were obtained by absolute measurement of the gamma activities of the product nuclei using a coaxial high-purity germanium detector. The neutron energies in this measurement was determined by cross section ratios for the $^{90}\text{Zr}(n, 2n)^{89\text{m}+g}\text{Zr}$ and $^{93}\text{Nb}(n, 2n)^{92\text{m}}\text{Nb}$ reactions [36].

Experimental

Cross sections were measured by activation and identification of the radioactive products. This technique is very suitable for investigating low-yield reaction products and closely space low-lying isomeric states, provided their lifetimes are not too short. The details have been described over the years in many publications [37–41]. Here we give some salient features relevant to the present measurements.

Samples and irradiations

About 3 g of HfO_2 powder of natural isotopic composition (>99.95% pure) was pressed at 10 ton/cm^2 , and a pellet, 0.3 cm thick and 2.0 cm in diameter was obtained. One such pellet was prepared. Monitor foils of Nb (99.999% pure, 0.20 mm thick) of the same diameter as the pellet were then attached in front and at the back of sample.

Irradiation of the samples was carried out at the ZF-300-II Intense Neutron Generator at Lanzhou University and lasted about 22 h with a yield ~ 0.8 to $2.4 \times 10^{11} \text{ s}^{-1}$. Neutrons were produced by the $\text{T}(d, n)^4\text{He}$ reaction with an effective deuteron beam energy of 125 keV and beam current of 20 mA. The tritium-titanium (T-Ti) target used in the generator was 0.9 mg/cm^2 thick. The neutron fluence rate was monitored by the accompanying alpha-particle so that corrections could be made for small variations in the yield. The accompanying α particle monitor is shown in Fig. 2. The Au–Si surface barrier detector used in 135° accompanying α -particle tube was at a distance of 119.7 cm from the target. The groups of samples were placed at 0° angle relative to the beam direction and centered about the T-Ti target at distances of 5 cm. Cross section for $^{93}\text{Nb}(n, 2n)^{92\text{m}}\text{Nb}$ reaction [42] was selected as monitors to measure the reaction cross section on several Hf isotopes.

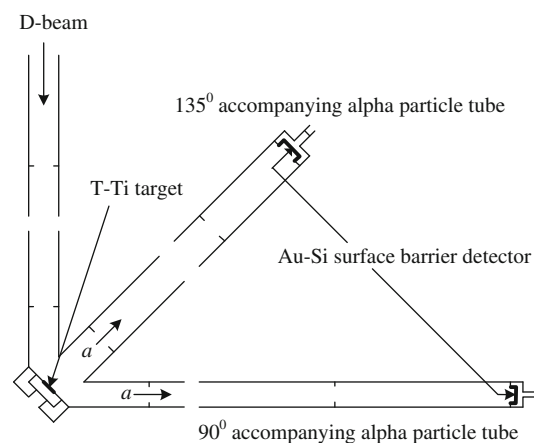


Fig. 2 ZF-300-II Intense Neutron Generator neutron flux monitor

Determination of the incident neutron energy

In the D-T reaction (Q value of 17.6 MeV), induced by deuterons of energy E_d , the kinetic energy E_n of the neutrons emitted at angle θ can be estimated [43] from the following expression (1);

$$(E_n)^{\frac{1}{2}} = \frac{(M_d M_n E_d)^{\frac{1}{2}} \cos \theta + (M_d M_n E_d \cos^2 \theta + \{M_x + M_n\} [M_x Q + E_d (M_x - M_n)])^{\frac{1}{2}}}{M_x + M_n} \tag{1}$$

where M_d , M_n and M_x are the masses of deuteron, neutron and alpha particle, respectively. The effective D-T neutron energy at irradiation position was determined by the Nb/Zr method [36, 43–45]. The measured neutron energy was shown in Fig. 3 together with the calculation using Eq. (1). The uncertainty in the neutron energy at ~ 5 cm was estimated to be 200 keV from a consideration of the sample sizes, d^+ beam diameter of about 3–4 mm, and the uncertainty in the Nb/Zr method [36].

The neutron energies were obtained both by the Eq. (1) and the Nb/Zr method (Lewis and Zieba [36]). The results are shown in Fig. 3. It can be seen, measured result is somewhat lower than the calculated value at neutron energy of 14.8 MeV. This discrepancy is probably due to the samples corresponding to the solid angle geometry, the neutron transport and scattering processes in the target and integrates over the sample volume. But measured result is consistent with the calculated value of Eq. (1) within the experimental uncertainty.

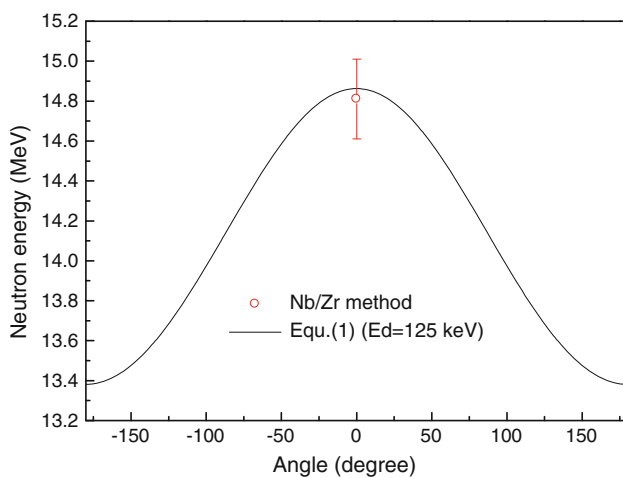


Fig. 3 Angular dependence of d-T neutron energy. The neutron energies were calculated by choosing the incident deuteron energy $E_d = 125$ keV. The open circle show experimental data determined by the Nb/Zr method (Lewis and Zieba, [36])

Measurement of radioactivity

The gamma ray activity of $^{178m2}\text{Hf}$, $^{179m2}\text{Hf}$, and ^{92m}Nb was determined by a CH8403 coaxial high-purity germanium detector made in the People’s Republic of China with a relative efficiency of 20% and an energy resolution of 3 keV at

1332 keV. The efficiency of the detector was calibrated using the standard gamma source, Standard Reference Material 4275 from the National Institute of Standards and Technology, Washington, D.C., USA. An absolute efficiency calibration curve was obtained at 20 cm from the surface of the germanium crystal. At this distance the coincidence losses can be considered to be negligible. In our case, however, we needed to calibrate the efficiency at 2 cm, the actual counting position used because of the weak activity of the sample. Therefore, we selected a set of single γ -line sources (^{137}Cs , ^{54}Mn) and placed them at two positions (20 and 2 cm) successively to measure their efficiency ratios to be able to evaluate the efficiency ratio curve as a function of energy. The absolute efficiency calibration curve at 2 cm was obtained from the calibration curve at 20 cm and the efficiency ratio curve. The uncertainty in the absolute efficiency curve at 2 cm was estimated to be $\sim 2\text{--}3\%$, while the uncertainty of the activity of the standard source is $\sim 1.0\%$.

The decay characteristics of the product radioisotopes and the natural abundances of the target isotopes under investigation are summarized in Table 1, [34, 35, 46].

Calculation of cross sections and their uncertainties

The measured cross sections can be calculated by the following formula [cf. 47]:

$$\sigma_x = \frac{[S\varepsilon I_\gamma K \eta M D]_m [\lambda A F C]_x}{[S\varepsilon I_\gamma K \eta M D]_x [\lambda A F C]_m} \sigma_m \tag{2}$$

where the subscript m represents the term corresponding to the monitor reaction and subscript x corresponds to the measured reaction, ε full-energy peak efficiency of the measured characteristic gamma-ray, I_γ gamma-ray intensity, η abundance of the target nuclide, M mass of sample, $D = e^{-\lambda t_1} - e^{-\lambda t_2}$ counting collection factor, t_1, t_2 time intervals from the end of the irradiation to the start and end of counting, respectively, A atomic weight, C measured full energy peak area, λ decay constant, F total correction factor of the activity:

Table 1 Reactions and associated decay data of activation products

Reaction	Abundance of target isotope (%)	Mode of decay (%)	Half-life of product	E_γ (keV)	I_γ (%)	Reference
$^{nat}\text{Hf}(n, x)^{178m2}\text{Hf}$	100	IT (100)	31 y	574.219	89.9	[34]
$^{nat}\text{Hf}(n, x)^{179m2}\text{Hf}$	100	IT (100)	25.05 d	362.55	41.1	[35]
$^{93}\text{Nb}(n, 2n)^{92m}\text{Nb}$	100	EC (100)	10.15 d	934.4	99.07	[46]

$$F = f_s \times f_c \times f_g \tag{3}$$

where f_s , f_c and f_g are correction factors for the self-absorption of the sample at a given gamma-energy, the coincidence sum effect of cascade gamma-rays in the investigated nuclide and in the counting geometry, respectively. Coincidence summing correction factor f_c was calculated by the method [48]. In turn, the gamma ray attenuation correction factors f_s in the HfO_2 foil and the geometry correction f_g were calculated by the following Eqs. (4) and (5), respectively.

$$f_s = \frac{\mu h}{1 - \exp(-\mu h)} \tag{4}$$

$$f_g = \frac{(D + h/2)^2}{D^2} \tag{5}$$

Here μ (in cm^{-1}) is the linear attenuation coefficient in HfO_2 for gamma rays at each of the photon energies E , h (in cm) the thickness of the sample and D is the distance from the measured sample to the surface of the germanium crystal.

K neutron fluence fluctuation factor:

$$K = \left[\sum_i^L \Phi_i (1 - e^{-\lambda \Delta t_i}) e^{-\lambda T_i} \right] / \Phi S \tag{6}$$

where L number of time intervals into which the irradiation time is divided, Δt_i duration of the i th time interval, T_i time interval from the end of the i th interval to the end of irradiation, Φ_i neutron flux averaged over the sample during Δt_i , $S = 1 - e^{-\lambda T}$ growth factor of the product nuclide, T total irradiation time, Φ neutron flux averaged over the sample during the total irradiation time T .

One can easily derive the expression of the Eq. (1) $\eta_x \sigma_x$ as

$$\eta_x \sigma_x = \frac{[S e I_\gamma \eta K M D]_m [\lambda A F C]_x}{[S e I_\gamma K M D]_x [\lambda A F C]_m} \sigma_m \tag{7}$$

Since the samples we selected are natural abundance, furthermore, the hafnium has many isotopes (^{174}Hf , 0.162%; ^{176}Hf , 5.206%; ^{177}Hf , 18.606%; ^{178}Hf , 27.297%; ^{179}Hf , 13.629%; ^{180}Hf , 35.100%) [46], it is obvious that a radioactive nuclide can be produced by several reactions, such as the reaction $^{nat}\text{Hf}(n, x)^{178m2}\text{Hf}$ is made up of

$^{179}\text{Hf}(n, 2n)^{178m2}\text{Hf}$, $^{178}\text{Hf}(n, n')^{178m2}\text{Hf}$, $^{177}\text{Hf}(n, \gamma)^{178m2}\text{Hf}$; $^{nat}\text{Hf}(n, x)^{179m2}\text{Hf}$ is made up of $^{180}\text{Hf}(n, 2n)^{179m2}\text{Hf}$, $^{179}\text{Hf}(n, n')^{179m2}\text{Hf}$, $^{178}\text{Hf}(n, \gamma)^{179m2}\text{Hf}$ and so on.

Surely, each of the reactions producing certain radioactive product has different contribution. So in the calculation we just gave the cross section of the element and didn't import the abundance of the target nuclide into the formula. For the reaction $^{nat}\text{Hf}(n, x)^{178m2}\text{Hf}$, there are three routes to produce $^{178m2}\text{Hf}$, the first via the $^{179}\text{Hf}(n, 2n)^{178m2}\text{Hf}$ reaction, the second through the $^{178}\text{Hf}(n, n')^{178m2}\text{Hf}$ reaction, and the third via $^{177}\text{Hf}(n, \gamma)^{178m2}\text{Hf}$ reaction. So the cross section of $^{nat}\text{Hf}(n, x)^{178m2}\text{Hf}$ reaction can be calculated by the following formula:

$$\begin{aligned} \sigma(^{nat}\text{Hf}(n,x)^{178m2}\text{Hf}) &= 0.13629\sigma(^{179}\text{Hf}(n,2n)^{178m2}\text{Hf}) \\ &+ 0.27297\sigma(^{178}\text{Hf}(n,n')^{178m2}\text{Hf}) \\ &+ 0.18606\sigma(^{177}\text{Hf}(n,\gamma)^{178m2}\text{Hf}) \\ &= \frac{[S e I_\gamma \eta K M D]_m [\lambda A F C]_x}{[S e I_\gamma K M D]_x [\lambda A F C]_m} \sigma_m \end{aligned} \tag{8}$$

For the reaction $^{nat}\text{Hf}(n, x)^{179m2}\text{Hf}$, there are three routes to produce $^{179m2}\text{Hf}$, the first via the $^{180}\text{Hf}(n, 2n)^{179m2}\text{Hf}$ reaction, the second through the $^{179}\text{Hf}(n, n')^{179m2}\text{Hf}$ reaction, and the third via $^{178}\text{Hf}(n, \gamma)^{179m2}\text{Hf}$ reaction. So the cross section of $^{nat}\text{Hf}(n, x)^{179m2}\text{Hf}$ reaction can be calculated by the following formula:

$$\begin{aligned} \sigma(^{nat}\text{Hf}(n,x)^{179m2}\text{Hf}) &= 0.35100\sigma(^{180}\text{Hf}(n,2n)^{179m2}\text{Hf}) \\ &+ 0.13629\sigma(^{179}\text{Hf}(n,n')^{179m2}\text{Hf}) \\ &+ 0.27297\sigma(^{178}\text{Hf}(n,\gamma)^{179m2}\text{Hf}) \\ &= \frac{[S e I_\gamma \eta K M D]_m [\lambda A F C]_x}{[S e I_\gamma K M D]_x [\lambda A F C]_m} \sigma_m \end{aligned} \tag{9}$$

The main contributions to the uncertainty of measurement in our work result from counting statistics (0.1–15%), standard cross sections uncertainties (1%), detector efficiency (2–3%), weight of samples (0.1%), self-absorption of gamma-ray (0.5%) and the coincidence sum effect of cascade gamma-rays (0–5%), the uncertainties of irradiation, cooling and measuring times (0.1–1%), etc. And some other uncertainty contribution form the parameters of the measured nuclei and standard nuclei, such

as, uncertainties of the branching ratio of the characteristic gamma rays, uncertainties of the half life of the radioactive product nuclei and so on all are considered.

Results and conclusions

Results

Using the relative activation technique of neutron, the sample was sandwiched between two Nb foils which were used as monitor. Because the cross section of $^{93}\text{Nb}(n, 2n)^{92m}\text{Nb}$ reaction has been measured by many authors, we can select it as a standard. With the irradiation taking place, the neutron flux for the sample was the same as the flux for the monitors. So we can find an equation of neutron flux between them, and avoid importing the neutron flux into the calculation.

After having been irradiated, the samples were cooled for 5–24 days, and one sample was measured with a 20% relative efficiency HPGe detector. One of the gamma-ray spectra is shown in Fig. 4. Cross-section values for $^{\text{nat}}\text{Hf}(n, x)^{178m2}\text{Hf}$ and $^{\text{nat}}\text{Hf}(n, x)^{179m2}\text{Hf}$ reactions were obtained relative to those of the $^{93}\text{Nb}(n, 2n)^{92m}\text{Nb}$ reaction. The cross sections for the $^{93}\text{Nb}(n, 2n)^{92m}\text{Nb}$ reaction, which is 459.7 ± 5.0 mb at 14.8 ± 0.2 MeV incident neutron energies, was obtained by interpolating the evaluated values of the literature [42]. The cross-sections measured in the present work are summarized in Table 2.

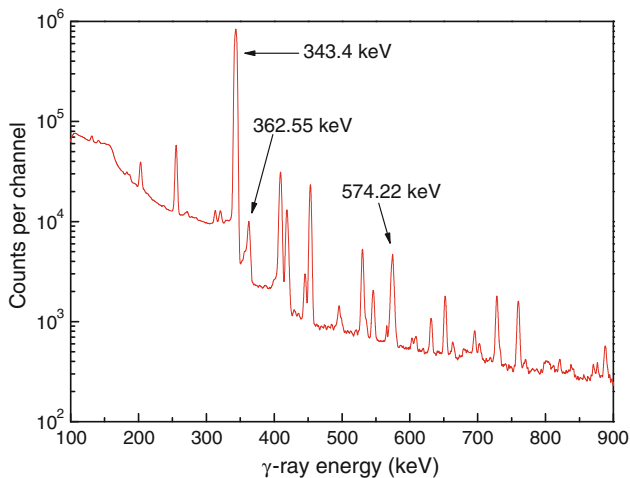


Fig. 4 The γ -ray spectra of about 24 days after the end of irradiation

Table 2 Summary of cross-section measurements

Reaction	Cross-sections (mb) at neutron energy 14.8 ± 0.2 (MeV)	Expanded standard uncertainty	Coverage factor k
$^{\text{nat}}\text{Hf}(n, x)^{178m2}\text{Hf}$	7.2	3.0	2
$^{\text{nat}}\text{Hf}(n, x)^{179m2}\text{Hf}$	18.6	3.4	2

Conclusions

We have measured the activation cross sections for $^{\text{nat}}\text{Hf}(n, x)^{178m2}\text{Hf}$ and $^{\text{nat}}\text{Hf}(n, x)^{179m2}\text{Hf}$ reactions on hafnium isotopes induced by 14.8 ± 0.2 MeV neutrons. It should be mentioned that this work presents the first cross-sections for formation of $^{178m2}\text{Hf}$ and $^{179m2}\text{Hf}$ through reactions on natural hafnium. The high precision of the cross-sections obtained in the earlier measurement was limited, because there are ^{174}Hf , ^{176}Hf , ^{177}Hf , ^{178}Hf , ^{179}Hf , and ^{180}Hf isotopes in the natural hafnium and overlapping of the gamma-peaks from different isotopes could not be neglected in the gamma-spectrum measurements with the use of NaI(Tl) detectors. The decay data such as half-life and branching ratio of the radioactive product nuclei were imprecise at that early time. In recent years, the reliabilities of the results have improved with the use of the Ge semiconductor detectors and the more accurate above-mentioned parameters. A good plan of irradiation and cooling time is also important and in our experiment we selected different irradiation time according to the half-life of the product nuclei. In addition, the present measurements were performed in the Low Background Laboratory of Lanzhou University and disturbance from environmental radiation was reduced to a very low level. In conclusion, our data would improve the quality of the neutron cross-section database.

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