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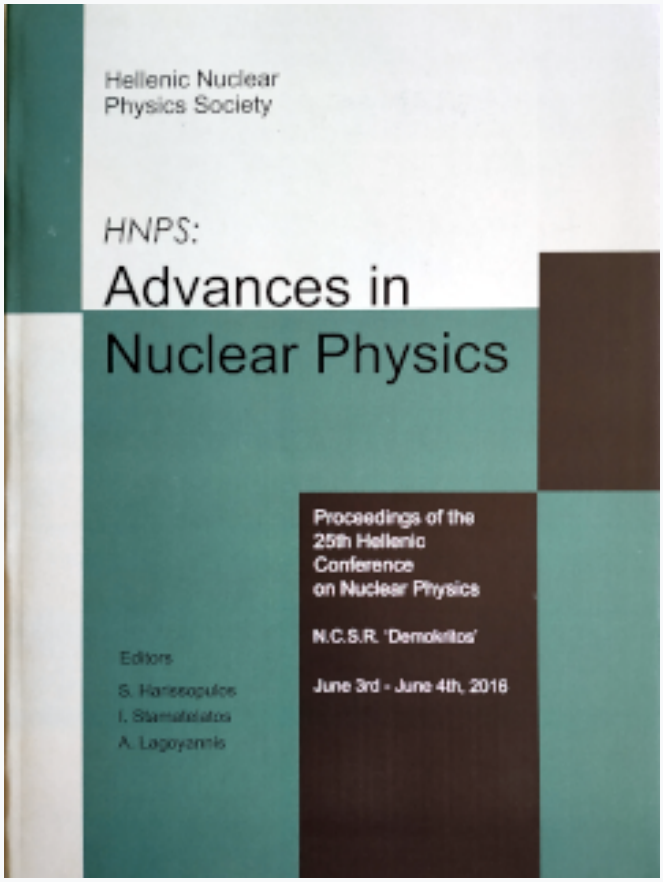

Investigation of (n,2n) reactions using the high energy neutron facility at NCSR "Demokritos"

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Abstract In the high energy neutron facility at the 5.5MV tandem T11/25 Accelerator of NCSR "Demokritos", the neutron beam is produced by means of the $^{3}H(d,n)^{4}He$ reaction in the energy range ~15-21 MeV and at a flux of the order of 10^{5} -10^{6} n/cm^{2}s. The beam has been characterized by means of the NeuSDesc and MCNP5 Monte Carlo simulations along with the multiple foil activation technique at various neutron energies. The neutron flux has been deduced using reference reactions, such as the $^{27}Al(n,\alpha)$ reaction, while the flux variation of the neutron beam was monitored by using a BF3 detector. After the characterization, the neutron beam has been used for (n,2n) reaction cross section measurements on Hf, Au and Ir isotopes via the activation technique. The irradiations were carried out at 15.3 and 17.1 MeV.

Keywords Nuclear Reactions, (n,2n) reaction cross section, neutron activation, Hafnium, Gold, Iridium

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INTRODUCTION

Investigation of neutron threshold reactions is of considerable importance not only for testing and improving nuclear models but also for many practical applications in nuclear technology, dosimetry, medicine and industry. All these tasks require improved nuclear data and accurate cross sections for neutron induced reactions at specific energies. The nuclei investigated in the present work, are of particular interest for various applications. Hf 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]. Neutron induced reactions on ^{197}Au are very important as reference/monitor reactions and more complete and reliable data are necessary for this purpose. The need, especially at high energies, of accurate reference/monitor reactions for advanced reactor system studies and other applications, has been emphasized in several IAEA meetings. Ir is a unique case among the elements relevant for radiochemical diagnostics based on different nuclear reactions which allow sampling of various components of a neutron energy spectrum [3]. The (n,2n) threshold reactions on ^{191}Ir and ^{193}Ir are

sensitive to the high energy neutrons (~14 MeV), while the ^{193}Ir(n, n') inelastic reaction is sensitive to lower energy neutrons (below 1 MeV).
Furthermore, the formation of a high spin isomeric state in the residual nucleus of a reaction is of considerable importance for testing nuclear models, as it is governed by the spin distribution of the level densities and the level scheme of the nuclei involved [4-6]. The ^{197}Au(n,2n) and ^{191}Ir(n,2n) reactions present interesting cases since the residual nuclei ^{196}Au and ^{190}Ir can be produced either in their ground state or in one of their high spin isomeric states, thus offering great sensitivity for such studies.

These reactions have been investigated in the past by many groups, including our group [1, 7, 8], from ~8 to ~24 MeV, with many discrepancies among the data, differing by as much as 50% in some cases. In the present work, the ^{174}Hf(n,2n), ^{197}Au(n,2n) and ^{191}Ir(n,2n) reactions have been measured at 15.3 and 17.1 MeV. In addition, the ^{193}Ir(n,2n)^{192}Ir and ^{176}Hf(n,2n)^{175}Hf threshold reaction cross sections have also been determined, taking into account the contribution from the contaminant ^{191}Ir(n,γ)^{192}Ir, ^{174}Hf(n,γ)^{175}Hf and ^{177}Hf(n,3n)^{175}Hf reactions. It should be emphasized that the (n,γ) reaction channels are open to low energy parasitic neutrons, which are always present in the low energy part of the main neutron beam. The correction method is based on the existing data in ENDF for the contaminant reactions, convoluted with the neutron spectra which have been extensively studied by means of simulations using the NeusDesc and MCNP codes.

EXPERIMENTAL PROCEDURE

The ^{174,176}Hf(n,2n), ^{197}Au(n,2n) and ^{191,193}Ir(n,2n) reaction cross sections have been measured at the 5.5 MV tandem T11/25 Accelerator Laboratory of NCSR "Demokritos" at 15.3 and 17.1 MeV. The neutron beam was produced via the ^{3}H(d,n)^{4}He reaction. A new Ti-tritiated target of 373 GBq activity has been used, consisting of a 2.1 mg/cm2 Ti-T layer on a 1mm thick Cu backing. Two collimators of 5 and 6 mm in diameter were used and the deuteron beam current was measured both at the collimators and the target and was kept at ~ 1 μA. During the irradiations, the flux variation of the neutron beam was monitored by a BF3 detector placed at a distance of ~3 m from the neutron production. The spectra of the BF3 monitor were stored at regular time intervals (~200 sec) by means of 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. The absolute flux of the beam was obtained with respect to the cross section of the ^{27}Al(n,α) reference reaction.

High purity Hf, Au, Ir and Al natural samples of 1.3cm in diameter, having a thickness of ~0.5 mm, were placed as a sandwich at a distance of ~2 cm from the tritium target and were irradiated for up to 96 h. The induced activity of product radionuclides was measured with two HPGe detectors of 56% and 100% relative efficiency, properly shielded with lead blocks. The efficiency of the detectors at the position of the activity measurements (10 cm) was determined via a calibrated ^{152}Eu point source. Corrections for self-absorption of the sample, coincidence summing effects of cascading gamma rays and counting geometry 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. During the activity measurements the resulted decay spectra of all samples were saved at regular time intervals in order to compare the evolution of the photopeak net-area counts with the theoretical curve for the expected half-life time.

Apart from the primary-energy neutrons, low energy neutrons are present in almost all cases, due to neutron production and scattering near the source as well as to scattering effects in the experimental area. A comprehensive understanding of the energy dependence of the neutron flux is of major importance for the reliability of reaction cross section measurements. In the absence of time-of-flight capabilities, for the investigation of the quasi-monoenergetic neutron beams produced via the 3H(d,n) reaction at the tandem Laboratory of NCSR "Demokritos", the multiple foil activation method has been applied along with Monte Carlo simulations implementing the NeuSDesc and MCNP codes [9, 10]. 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 $\Phi\iota(E)$ at the targets was estimated. In order to validate the simulated $\Phi\iota(E)$, the simulated reaction rates (R.R) of the reactions used in the multiple foil activation (namely ^{58}Ni(n,p)^{58}Co, ^{93}Nb(n,2n)^{92m}Nb, ^{197}Au(n,γ)^{198}Au, ^{56}Fe(n,p)^{56}Mn, ^{59}Co(n,α)^{56}Mn, ^{115}In(n,n')^{115m}In, ^{46}Ti(n,p)$^{46m+g}$Sc, ^{47}Ti(n,p)^{47}Sc, ^{48}Ti(n,p)^{48}Sc, ^{64}Zn(n,p)^{64}Cu and ^{27}Al(n,α)^{24}Na), have been deduced using the expression :

$$R.R = \int_{Eth,i}^{\infty} \sigma_i\ E\ \cdot \Phi_i\ E\ \ dE$$

where $\sigma_i(E)$ are the excitation functions of the reactions taken from the ENDF/B-VII.1 library and $\Phi_i(E)$ the function of the neutron fluence normalized to the experimental fluence on each foil. In fact, the normalized neutron spectral distribution has been cut in energy slices ΔE starting from the threshold of each reaction up to the maximum neutron energy and the sum

$$R.R = \sum_{\Delta E} \sigma\ E\ \cdot \Phi\ E \qquad (1)$$

has been deduced for each reaction. The resulting simulated reaction rates have been compared with the experimental ones and seem to agree well, thus verifying the reliability of the simulations [11]. The tests have been carried out at 15.3±0.5 MeV and 17.0±0.3 MeV neutron energies, which have subsequently been used for the cross section measurements of the present work.

MEASUREMENTS AND RESULTS

The (n,2n) reaction on Hf isotopes

Of all the isotopes contained in natural Hf, only the ^{176,174}Hf(n,2n) reactions can in principle be studied using the activation technique. ^{174}Hf is the lowest abundant isotope (0.162%), but the ^{174}Hf(n,2n)^{173}Hf reaction has a high cross-sectional value, so it can easily be measured. The residual nucleus ^{173}Hf decays by electron capture, with a half-life of 23.6 h, to ^{173}Lu and the two most intense characteristic γ-ray transitions of 123.7 keV and 297.0 keV have been used for the determination of the reaction cross section. As for the ^{176}Hf(n,2n)^{175}Hf reaction the residual nucleus decays by β^+, with a half-life of 70 d, to ^{175}Lu, but the characteristic γ-ray transition 343.4 keV cannot directly be used for the determination of the reaction cross section, due to the contamination from the ^{174}Hf(n,γ)^{175}Hf and ^{177}Hf(n,3n)^{175}Hf

reactions. In order to estimate the correction for this contribution, the reaction rate *RR* of the contaminant reactions has been deduced using the *σ(E)* excitation function from the ENDF-VII library and the simulated Φ(E), normalized to the experimental fluence on the first Al foil, following expression (1). The results showed that the contribution of the ^{174}Hf(n,γ)^{175}Hf was irrelevant, due to the low (n,γ) cross section, combined with low flux of parasitic neutrons. Regarding the ^{177}Hf(n,3n)^{175}Hf contribution, the corrections turned out to be the major ones due to the fact that the energy threshold of the (n,3n) reaction is just above 15 MeV. In more detail, for the 15.3 MeV irradiation the correction was 4%, while at 17.1 MeV the correction reached 46% [12]. The experimental cross sections for the ^{176,174}Hf(n,2n) reactions at 15.3 and 17.1 MeV are presented in Fig. 1 along with data from literature and ENDF evaluations.

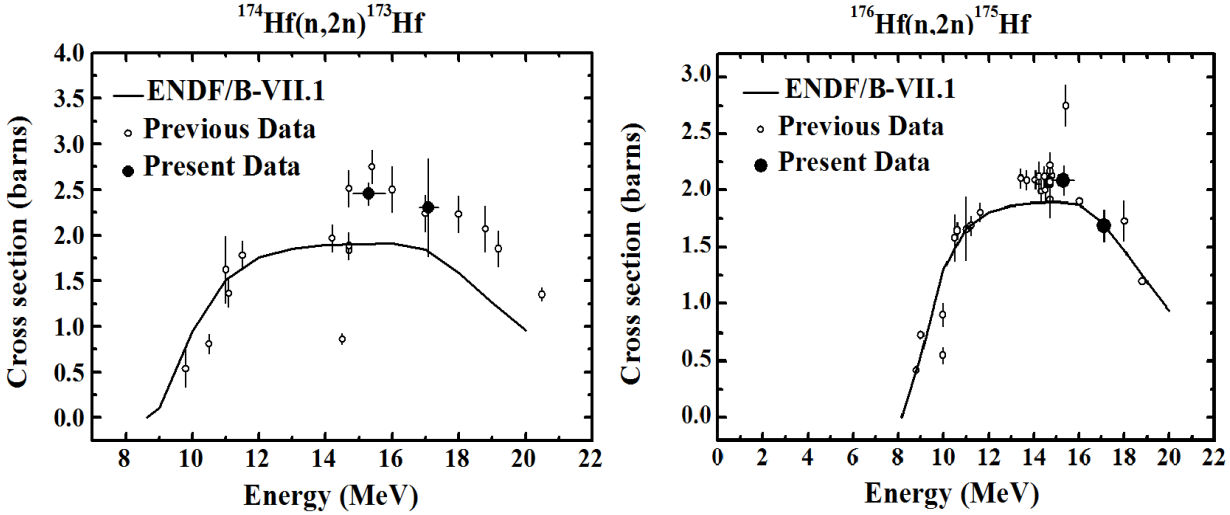

Fig. 1. Cross section data from the present work and EXFOR database of the ^{174}Hf(n,2n)^{173}Hf and ^{176}Hf(n,2n)^{175}Hf reactions. The solid lines represent evaluations from ENDF/B-VII.1.

The (n,2n) reaction on Au

The ^{197}Au(n,2n)^{196}Au reaction presents an interesting case since the residual nucleus ^{196}Au can be populated either in its high spin 12$^-$ second isomeric state (m2) or in its low spin 2$^-$ ground state (g). The cross sections for the population of the second isomeric state m2 (J^π=12$^-$ and half-life 9.7 h) of ^{196}Au and the sum of the ground g (J^π=2$^-$ and half-life 6.2 d) and first isomeric state m1 (J^π=5$^-$ and half-life 8.1 s), were independently determined through their characteristic gamma-rays. The 148 keV transition was used from the deexcitation chain of the second isomeric state and the 356 keV transition for the sum of the ground and first isomeric state. These gamma-rays were the most intense and non-contaminated transitions from the decay of ^{196}Au [13]. The experimental cross sections for the population of the m2 and g+m1 states at 15.3 and 17.1 MeV are presented in Fig. 2 along with data from literature and ENDF evaluations.

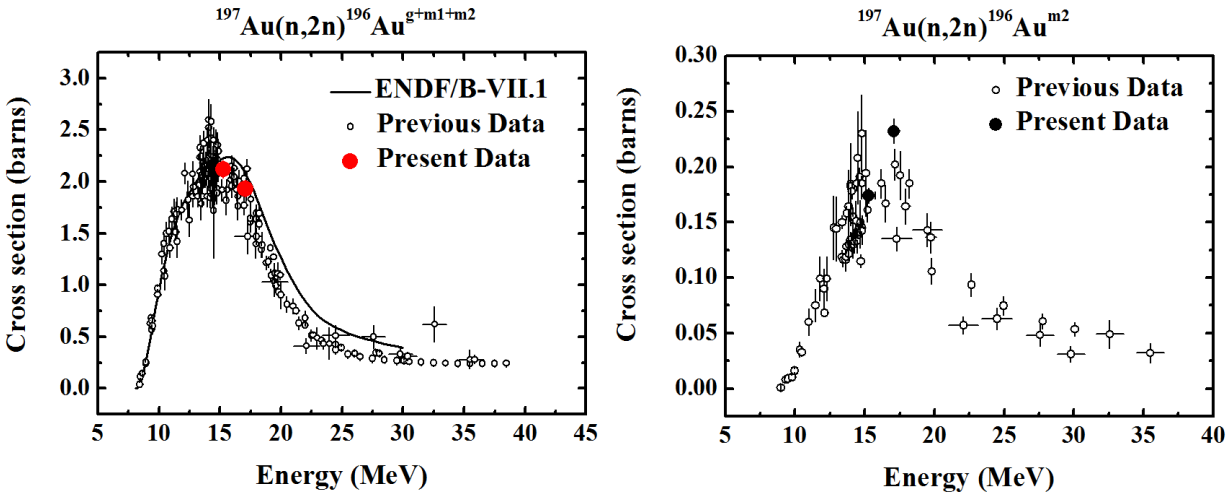

Fig. 2. Cross section data from the present work and EXFOR database of the ^{197}Au(n,2n)^{196}Au$^{g+m1+m2}$ and ^{197}Au(n,2n)^{196}Au^{m2} reactions. The solid line represents evaluations from ENDF/B-VII.1.

The (n,2n) reaction on Ir isotopes

Natural Ir consists of two isotopes ^{191}Ir and ^{193}Ir having 37.3% and 62.7% abundances, respectively. The ^{191}Ir(n,2n) reaction leads to the formation of ^{190}Ir in its ground 4$^-$ state ($T_{1/2}$=11.78 d), as well as its metastable m1 1$^-$ state ($T_{1/2}$=1.12 h) and m2 11$^-$ state ($T_{1/2}$=3.087 h), which decay to ^{190}Os. Due to the short half-life of m1, the sum of the metastable m1 and the ground state cross sections was determined via the most intensive 518.5 keV transition of ^{190}Os. The population of the second isomeric state m2 was measured independently through the 616.5 keV transition of ^{190}Os. The experimental cross sections for the ^{191}Ir(n,2n)^{190}Ir^{g+m1}, ^{191}Ir(n,2n)^{190}Ir^{rm2} cross sections at 15.3 and 17.1 MeV are presented in Fig. 3 along with data from literature and ENDF evaluations.

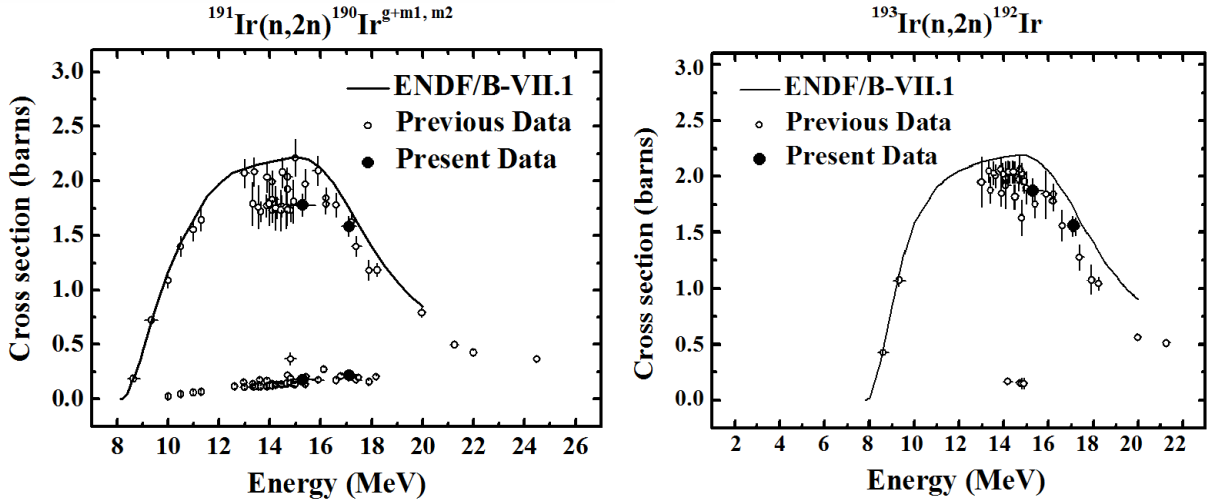

Fig. 3. Cross section data from the present work and EXFOR database of the ^{191}Ir(n,2n)^{190}Ir^{g+m1}, ^{191}Ir(n,2n)^{190}Ir^{m2} and ^{193}Ir(n,2n)^{192}Ir reactions. The solid lines represent evaluations from ENDF/B-VII.1.

As for the ^{193}Ir(n,2n)^{192}Ir reaction, the residual nucleus ^{192}Ir decays to ^{192}Pt with a half-life of 74.2 d. The characteristic γ-ray transitions 308.5, 316.5 and 468.1 keV from the de-excitation of ^{192}Pt can be used for the determination of the cross section of this reaction. The ^{192}Ir nucleus however, can also be produced by the ^{191}Ir(n,γ) reaction channel, which is always present and open to low energy parasitic neutrons. The contamination from the ^{191}Ir(n,γ) reaction has been deduced using the $\sigma(E)$ excitation function from the ENDF-VII library and the simulated $\Phi(E)$, normalized to the experimental fluence on the first Al foil, following the same procedure described before [14]. The experimental cross sections for the population of the m2 and g+m1 states at 15.3 and 17.1 MeV are presented in Fig. 3 along with data from literature and ENDF evaluations.

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,α) 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, ^{176}Hf, ^{197}Au, ^{191}Ir and ^{193}Ir isotopes. In the determination of the ^{176}Hf(n,2n)^{175}Hf reaction cross section the contamination from 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 was important in the case of corrections for the ^{193}Ir(n,2n)^{192}Ir reaction, contaminated by the ^{191}Ir(n,γ)^{192}Ir one. The population of isomeric states has also been measured in the ^{197}Au(n,2n)^{196}Au^{m2} and ^{191}Ir(n,2n)^{190}Ir^{m2} reactions. Additional cross section measurements are planned in the 18-20 MeV region in the near future and statistical model calculations are in progress by using the EMPIRE code.

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