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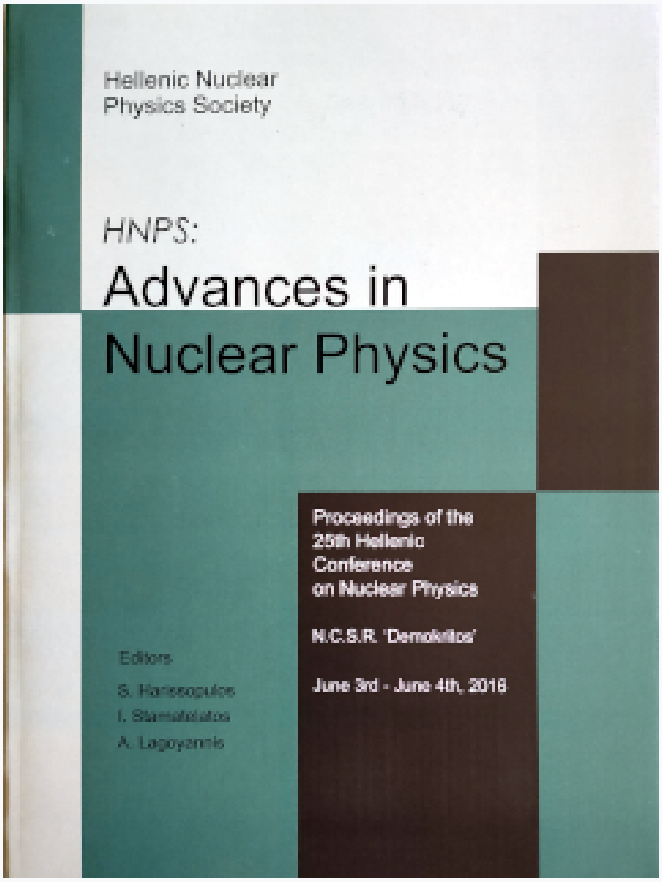

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^{197}Au(n,xn) reactions at The Svedberg Laboratory high-energy neutron facility in Uppsala

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Abstract Cross section measurements for the ^{197}Au(n,xn) reactions have been performed at The Svedberg Laboratory (TSL) high-energy neutron facility in Uppsala,Sweden. The 45.6 and 58.3 MeVquasi-monoenergetic neutron beams were produced by means of the ^{7}Li(p,n) reaction and were monitored with thin-film breakdown counters (TFBCs). After the end of the irradiations, the activity induced by the neutron beams in the targets and in reference foils, has been measured by a HPGe detector. In order to determine the cross sections of the (n,xn) reactions, the spectral neutron flux distribution is needed, thus the characterization of the beam is of major importance. Therefore, simulations that take into account the whole experimental setup of the irradiation have been performed with the use of MCNP5 code and the results are presented in this work. Currently, further analysis of the data is in progress.

Keywords quasi-monoenergetic neutron beam, Au(n,xn) cross section, MCNP5 code

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INTRODUCTION

Studies of the excitation functions of nuclear reactions are of considerable importance for testing nuclear models as well as for practical applications. The case of ^{197}Au is particularly interesting since neutron induced reactions on ^{197}Au are important as reference/monitor reactions. The (n,γ) reaction is recommended as reference reaction for astrophysical applications below 200 keV, while the (n,xn) reactions are proposed as a standard for high energy neutron dosimetry [1]. In addition, the residual nuclides from the ^{197}Au(n,xn) reaction channels haveconvenienthalf-lives and the respective activities can be measured using the activation method, providing a complete set of exit channels for testing nuclear models. Thus, the availability of complete and reliable cross-section datasets is mandatory. Up to 20 MeV, data available in literature are generally considered to be reasonably well known, whereas at higher energies, datasets exist only for the (n,3n) and (n,4n) reactions with very poor statistics and many discrepancies among them, while for the (n,5n) and (n,6n) channels, only few data points are available in literature. The Svedberg Laboratory (TSL) high-energy neutron facility in Uppsala, Sweden, is a unique place for such measurements, with neutron beams of reasonable flux (~10^{5} cm^{-2} s^{-1}) and energies up to 175MeV [2].

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Thus, a proposal has been submitted for ^{197}Au(n,xn) cross-section measurements and in November 2015 the corresponding experiment was carried out at TSL, at 45.6 and 58.3 MeV incident neutron energies.The neutron beams are quasi-monoenergetic which means that, although there is a strong dominance of neutrons over a narrow energy range, there also exist low-energy neutrons which can also activate the targets. Therefore, the beam characterization is of vital importance for the determination of the cross sectionof the (n,xn) reactions. For this purpose, simulations that take into account the whole experimental setup of the irradiation have been performed with the use of the MCNP5 code and the results are presented in this work. Currently, further analysis of the data is in progress.

NEUTRON INTERACTION ON ^{197}Au

When a neutron with an energy of 45.3 or 58.0MeV (in the CM system) impinges on a ^{197}Au nucleus, emission of as many as 7 neutrons becomes energetically possible (see Fig. 1).

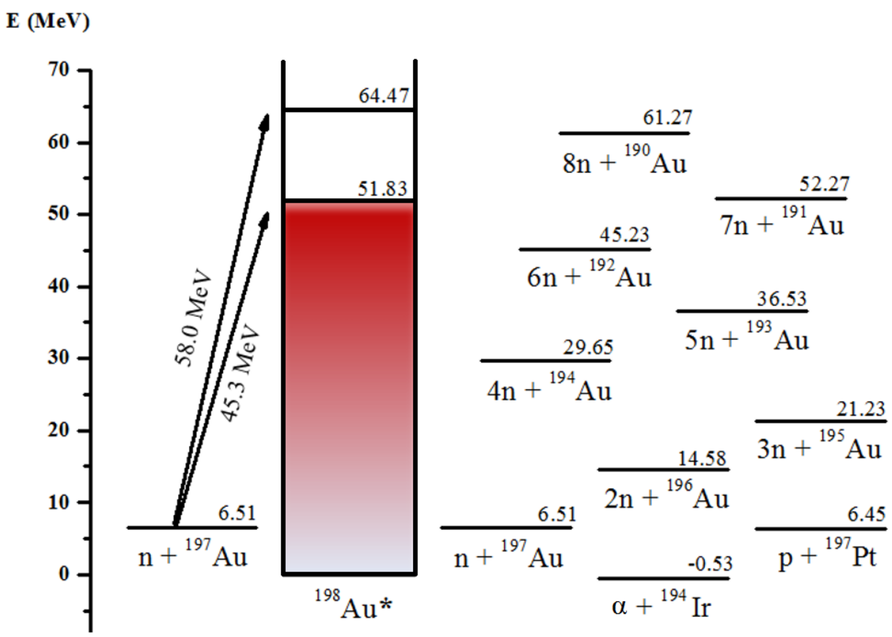


Fig. 1. Energy diagram of the n+^{197}Au interaction. The energies are given in MeV in the CM system.

Reaction	Residual Nucleus	$T_{1/2}$	E_{thr} (MeV)	E_γ (keV)
^{197}Au (n,2n)	^{196}Au	6.2 d	8.11	355.7
^{197}Au (n,3n)	^{195}Au	186.1 d	14.79	98.9
^{197}Au (n,4n)	^{194}Au	38.0 h	23.26	948.3
^{197}Au (n,5n)	^{193}Au	17.7 h	30.17	255.6
^{197}Au (n,6n)	^{192}Au	3.2 h	38.92	316.5
^{197}Au (n,7n)	^{191}Au	3.2 h	45.99	586.5

Table 1. ^{197}Au(n,xn) reactions along with their residual nuclei andenergy thresholds. The half-lives of the residual nucleialong with the characteristic γ-rays emitted during their deexcitation are also presented.

The residual nuclei from the ^{197}Au(n,xn) channels have convenient half-lives (see Table I) and therefore they can be measured using the activation technique.

The neutron beam distribution contains a long tail of low-energy neutrons, which contribute to production of the residual radionuclides (i.e. the beam energy of neutron that causes the reaction cannot be unambiguously assigned) and consequently, the number of the produced nuclei during the irradiation should be corrected for this contribution. Therefore, the characterization of the beam is of major importance in order to account for the low-energy neutrons and thus accurately determine the cross section.

EXPERIMENTAL SETUP AND ACTIVATION MEASUREMENTS

The measurements were carried out at The Svedberg Laboratory (TSL) high-energy neutron time-of-flight facility in Uppsala, Sweden. The proton beam was accelerated up to 48.5 (61.0) MeV by means of the Gustaf Werner cyclotron and the 45.6 (58.3) MeV quasi-monoenergetic, pulsed neutron beam was produced using the ^{7}Li(p,n)^{7}Be reaction. The 4 mm thickness, rectangular shaped (20x32mm^2) Li foil was situated upstream of a massive bending magnet in a concrete cave for radioprotection purposes (see Fig. 2). The starting and stopping signals for the time-of-flight measurement were defined by the RF system of the cyclotron and by means of Thin-Film Breakdown Counters (TFBCs), which trigger when neutrons are detected [3]. The function of TFBCs is based on neutron-induced fission reactions using ^{238}U, ^{209}Bi, ^{235}U targets (the details of the method are described in Ref. [4-6]) and they have been successfully used for the measurement of spectral neutron flux at several facilities [4,6,9].

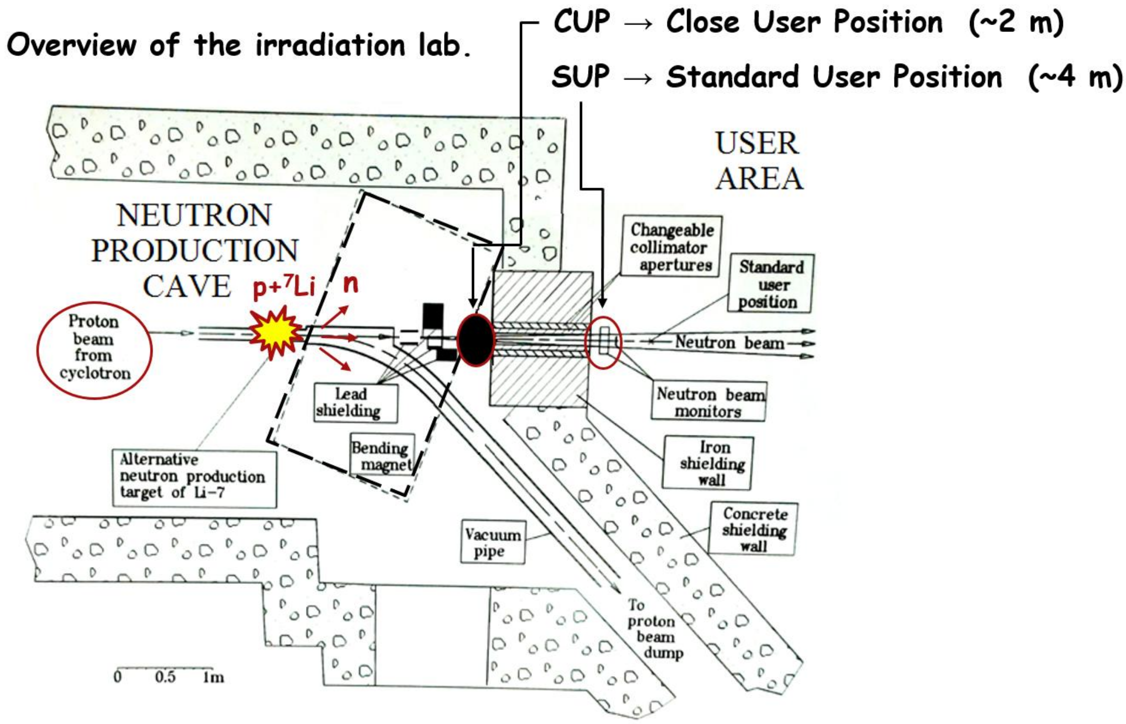
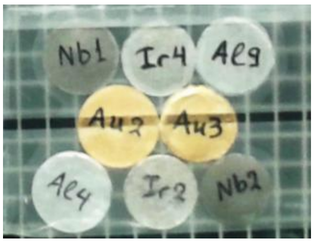


Fig. 2. A schematic layout of the quasi-monoenergetic neutron (QMN) facility at TSL with the Close User Position (CUP) and theStandard User Position (SUP) locations marked.

During the irradiations, two available positions where used, namely the Close User Position (CUP) [9] and the Standard User Position (SUP). In the CUP position, high-purity foils ofAl, Nb, Ir and Au were placed (Fig. 3a) in order to have the highest neutron flux available, while in the SUP position a Au foil was placed (Fig. 3b) in order to double-check the results of the subsequent off-line study for the neutron beam profile in the position where the neutron beam has been well characterized in the past [2,10].



(a)

(b)

Fig. 3. (a) Target assembly with Al, Nb, Ir and Au high purity foils placed at the Close User Position (CUP) and (b) Au foil placed at the Standard User Position (SUP).

Following the irradiations, the induced activity on the high purity targets was measured with a HPGe detector, properly shielded with lead blocks to reduce the contribution of ambient natural radioactivity. The samples were placed at a distance of 10 cm from the detector window. Calibrated ^{152}Eu, ^{133}Ba, and ^{137}Cs sources, placed at the same distance, were used to determine the efficiency of the detector. With this setup, corrections for true coincidence summing effects were negligible. Spectra of the characteristic γ-rays of the residual nuclei from the (n,4n), (n,5n), (n,6n), and (n,7n) channels, detected during the measurements, are presented in Fig. 4.

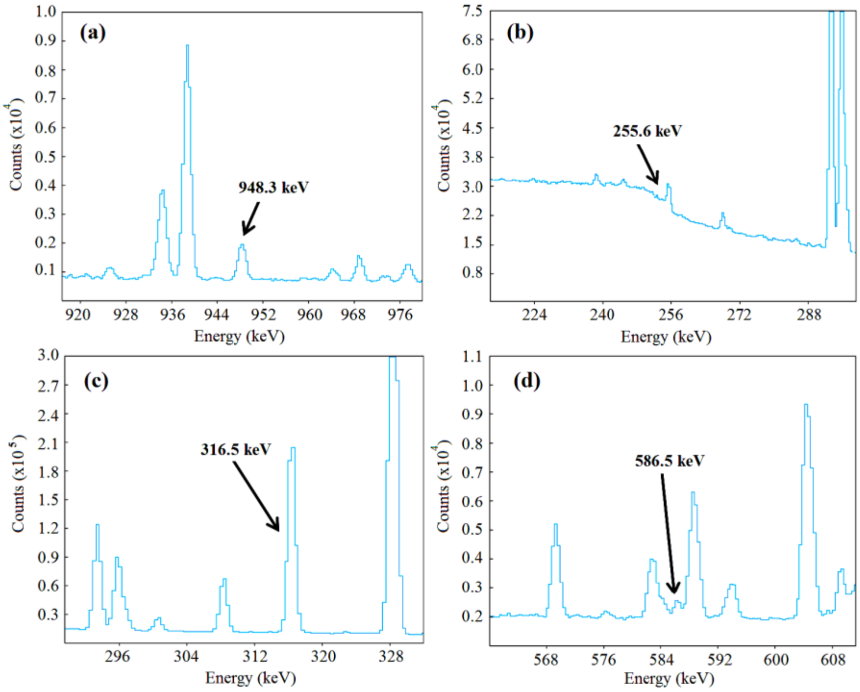


Fig. 4. Characteristic γ-rays in the spectra taken after the end of the irradiation which correspond to the (a) Au(n,4n), (b) Au(n,5n), (c) Au(n,6n) and (d) Au(n,7n) channels, respectively.

ESTIMATION OF THE ABSOLUTE NEUTRON FLUX

In order to study the neutron beam profile and the spectral neutron flux, MCNP5 [11] simulations were performed taking into account the experimental set-up in great detail (Fig. 5).

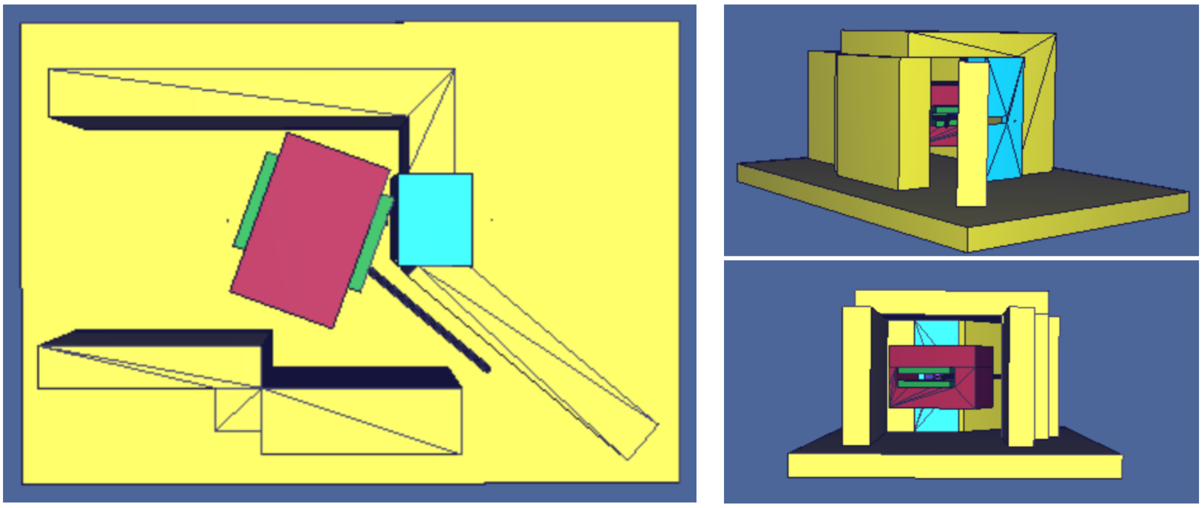


Fig. 5. Full geometry of the irradiation lab. The figures are extracted from the 3D-view plot feature of the MCNP5 visual editor.

As input for the MCNP5 simulation,a neutron distribution obtained from the systematics for neutron spectra by A. V. Prokofiev et. al. [10,12] was used and the results showed that interactions of neutrons with the surrounding materials contribute only to the low-energy neutron tail, which is not critical when studying reactions with threshold neutron energies above ~8 MeV. Moreover, as is shown in Table 2, the neutron beam is laterally homogeneous at the CUP, since the neutron flux between the foils differs by no more than 3%.

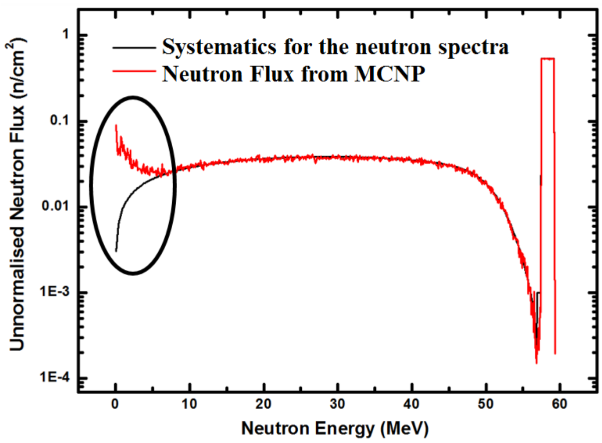


Fig. 6. Neutron flux obtained from MCNP5 compared to the systematics from Ref. [10] in order to study the contribution of the surrounding materials.

Foil	Unnormalised Neutron Flux from MCNP (a.u)
Au2	2.62E-09
Au3	2.59E-09
Nb1	2.59E-09
Nb2	2.56E-09
Al9	2.54E-09
Al4	2.54E-09
Ir2	2.60E-09
Ir4	2.62E-09

Table 2. MCNP5 results (need to be normalized) for the neutron flux in the targets placed at the CUP.

For the estimation of the absolute neutron flux, the following folding method was used. The unnormalised reaction rate for a reference reaction can be determined theoretically according to the expression:

where σ_i is the cross section of the reaction under study, retrieved from the TENDL library [13] and Φ_{mcnpi} is the spectral neutron fluence on the target according to the MCNP5 simulation (with arbitrary normalization). 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 neutronenergy and the sum

has been deduced. Further, the reaction rate for each studied reaction was determined experimentally using the following expression:

where λ is the decay constant of the residual nucleus, t_B the irradiation time, N_p the number of the produced nuclei and N_t the number of the target nuclei. By dividing the two aforementioned reaction rates one can deduce a normalization factor α (sec^{-1}) = $RR_{experimental}$ / RR_{mcnp} and then, the total absolute neutron flux can be estimated via the expression:

The results are presented in Table 2 for two reference reactions and are in excellent agreement, resulting to the total neutron flux of $8.4 \cdot 10^4$ n/cm^2 s at the target position (CUP).

Reaction	$RR_{experimental}$	RR_{mcnp}	α (sec^{-1})	Total Neutron Flux (n /cm^2 * sec)
Au (n,2n)	3.59E-20	2.22E-26	1.62E+06	8.44E+04
Ir (n,2n)	3.11E-20	1.94E-26	1.60E+06	8.35E+04

Table2. Results for both experimental and simulated reaction rates along with the normalization factors and the total neutron flux for each reference reaction.

This process, however, is only the first step in a more detailed investigation of the neutron beam energy spectrum, which is under progress. The transmission of the low-energy neutron tail at 4π geometry should be taken into account in the simulations. In addition, the information concerning the time-of-flight from the TFBCs will be used in comparison with MCNP simulations. Moreover, further tests will be carried out for more reference reactions, such as ^{93}Nb(n,2n) and ^{27}Al(n,α), along with ^{238}U and ^{209}Bi fission data from the TFBC detectors, in order to cross-check the neutron beam profile and normalization. After this neutron beam detailed analysis process, the cross sections of the (n,4n), (n,5n), (n,6n) and (n,7n) reactions on ^{197}Au at 45.6 MeV and 58.3 MeV will be deduced, implementing the above mentioned folding technique to account for the contribution of the long tail of low-energy neutrons to the activation and production of the residual nuclei ^{194}Au, ^{193}Au, ^{192}Au and ^{191}Au, respectively.

SUMMARY

Cross section measurements for the ^{197}Au(n,xn) reactions were performed at The Svedberg Laboratory (TSL) high-energy neutron facility in Uppsala, Sweden. The 45.6 and 58.3 MeV quasi-monoenergetic neutron beams were produced by means of the ^{7}Li(p,n) reaction and were monitored with thin-film breakdown counters (TFBCs). After the end of the irradiations, the activity induced by the neutron beams in the targets and reference foils, was measured by implementing a HPGe detector. Further, MCNP5 simulations that take into account the whole experimental setup of the irradiation were performed in order to study the spectral neutron flux distribution and the resulting total neutron flux at CUP, according to the ^{197}Au(n,2n) and ^{191}Ir(n,2n) reference reactions. The latter was found to be ~8.4·10^{4} n/cm^{2}s. Currently, more detailed simulations and further reference reactions such as the ^{27}Al(n,α), ^{238}U(n,f), ^{209}Bi(n,f) and ^{93}Nb(n,2n) ones are being investigated in order to verify the results concerning the absolute value of the neutron flux. The most accurate information about the neutron flux distribution will be provided by the ToF spectrum from the TFBCs. The next steps will be to deduce the cross sections of (n,4n), (n,5n), (n,6n) and (n,7n) reaction channels and to perform theoretical calculations using the EMPIRE code.

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