

Integrated Modeling of Natural Tin Photoactivation

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ABSTRACT

In this study photoactivation reaction cross-sections on natural Sn were evaluated in the GDR region for incident neutron energy up to some ten's of MeV's. For each Sn isotope, the contribution of the compound, direct and preequilibrium mechanisms were extracted. The cross-sections and isomer ratios were calculated. The results are compared with existing experimental data obtained by Bremsstrahlung source and activation methods.

Theoretical evaluations of the photoactivation cross-sections obtained with Talys are in good agreement with existing experimental data. Taking into account the satisfactory description of experimental cross-section data, the isomer ratios for incident photons energy from the neutron threshold (8÷9 MeV) up to 36 MeV were calculated.

If the isomer nuclei are obtained in final states, then the isomer ratios were evaluated using the corresponding production cross-sections (σ_m , σ_g) in the exclusive (γ , 1n) reactions. In the present work, the results of (γ , n) reaction on ¹¹⁴Sn and ¹²⁰Sn nuclei are analyzed.

1. Introduction

Nuclear reactions induced by photons with neutrons emission are of interest for fundamental and applied researches. Related to the first type of studies these reactions provide new data on the structure of nuclei, nuclear states densities, and reaction mechanisms. For applied researches, photoneutron processes are of great importance in safeguard technologies, nucleosynthesis, radiation shielding design and transport, activation analysis, isotopes production, etc (Ishkhanov & Varlamov, 2004), (Ishkhanov & Troschiev 2010).

The nucleus Sn has a magic number of protons ($Z=50$) and the highest number of stable isotopes among the nuclei from Mendeleev's Table. The atomic masses (A) and abundances (%) of the Sn isotopes are (A -%): 112-0.96%, 114-0.66%, 115-0.35%, 116-14.30%, 117-7.61%, 118-24.03%, 119-8.58%, 120-32.85%, 122-4.72%, 124-5.94% (Audi & Wapstra, 1993), (Audi & Wapstra, 1995), (Vocke, 1999).

In nuclear physics, the isomer states are considered the excited states of atomic nuclei with high spins value and consequently with a very large time of life compared with other excited nuclear states. They can be obtained in nuclear reactions and represents an efficient way to produce new artificial nuclei which are of interest for fundamental and applicative researches. The cross-sections of (γ , xn) photoneutron reactions ($x=1,2,3,4$) on Sn isotopes using Talys codes have been evaluated (Koning & all, 2020), (Koning & Rochman, 2012). When isomer

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nuclei are produced, isomer ratios, for incident photons energy from the neutron threshold ($8 \div 9$ MeV) up to 36 MeV have been calculated also. For each process, the contribution of reaction mechanisms followed by a separation into discrete and continuum states to the cross- section was extracted.

Theoretical evaluations of the photoneutron cross-sections obtained with Talys are in good agreement with existing experimental data from literature and the isomer ratios were also calculated.

2. Theory

In the case of photons induced reactions when the incident gammas are produced by a Bremsstrahlung source the experimental isomer ratio has the expression (Tran & all, 2009), (Tran & all, 2006):

$$R^{\text{exp}} = \left[\int_{E_{th}}^{E_m} N_0 \phi(E_\gamma) \sigma_m(E_\gamma) dE_\gamma \right] \cdot \left[\int_{E_{th}}^{E_m} N_0 \phi(E_\gamma) \sigma_g(E_\gamma) dE_\gamma \right]^{-1} \quad (1)$$

where: E_{th} = neutron threshold energy of incident gammas, E_m = maximum energy of gammas, N_0 = concentration of investigated isotope, ϕ = incident gamma flux; $\sigma_{m,g}$ = cross- section of isomer (m = metastable) and ground (g) states respectively production.

Relation (1) shows that for isomer ratio evaluations, the cross-sections are necessary. From neutron threshold (≈ 8 MeV) up to 36 MeV, including GDR region, are acting all known nuclear reaction mechanisms (compound, pre – equilibrium, and direct) which are considered in the calculations. A comprehensive description of nuclear reaction mechanisms can be found in (Koning & Rochman, 2012) and more details about pre-equilibrium processes can be read in (Gadioli & Hodgson, 1992). As it will be shown in the next paragraph, in the mentioned energy interval compound nucleus mechanism is dominant. In this case compound processes are described by the statistical model of nuclear reactions and the cross-section can be written using Hauser – Feshbach formalism is (Hauser & Feshbach, 1952):

$$\sigma_{\alpha\beta} = \pi \lambda_\alpha^2 T_\alpha T_\beta \left(\sum_c T_c \right)^{-1} W_{\alpha\beta} \quad (2)$$

where: T = penetrability (or transmission) coefficients, $W_{\alpha\beta}$ = width fluctuation correction factor, λ_α = reduced wavelength of incident particle, α, β, c = incident, emergent and possible outgoing channels, respectively. Sum is taken over all possible open channels c . Our case $\alpha = \gamma, \beta = xn, x = 1, 2, 3, \dots$.

In the Hauser-Feshbach formula (2) the penetrability coefficients in the ingoing and outgoing channels are necessary to be calculated and they are defined as the probability of a particle to pass a potential barrier. The transmission coefficients are increasing with energy but their values are always lower than 1. When the excitation energy of the compound nucleus involves only a few discrete states of the final nucleus then the penetrability coefficients can be calculated using approaches described in the textbooks of quantum mechanics. If the excitation energy is high leading to states in the continuum it is necessary to calculate the effective transmission coefficients (Koning & Rochman, 2012).

$$T_{\text{eff}}(U) = \int_{E_{\text{min}}}^{E_{\text{max}}} \rho(\varepsilon) T(\varepsilon) d\varepsilon \quad (3)$$

where: U = excitation energy, ρ = density levels.

The width fluctuation correction factor W represents a correlation between the incident channel (α) and outgoing channels (β). At low incident energies, this factor is equal with one indicating no correlation between ingoing and outgoing channels (Bohr hypothesis). The fluctuation factor is decreasing slowly with the energy. There are a few approaches to obtain the correction factor W and its calculation is described in detail in (Koning & Rochman, 2012).

The cross-sections and isomer ratios are evaluated using Talys codes. Talys provides precise nuclear reaction evaluations starting with 1 keV up to 1000 MeV, useful for describing of nuclear data. By free contributions of many authors the nuclear reaction mechanisms (compound, direct, pre-equilibrium), nuclear data like energy levels and density etc in a unitary friendly easy to use interface were implemented (Koning & all 2020), (Koning & Rochman 2012).

The cross-section is exclusive when in the outgoing channel, a well-defined state, with a residual nucleus and emergent particle(s) are counted. For example, in the case of $(\gamma, 2n)$ process, exclusive cross-section means when in the final state there are two neutrons coming from the same channel $(\gamma, 2n)$. By comparison, inclusive cross-sections can be understood when in the final state, emergent particles coming from other open channels are considered. In the case of a (γ, n) inclusive cross-section, registered neutrons come from other channels involving neutrons like (γ, xn) , (γ, np) , $(\gamma, 2np)$, $(\gamma, n2p)$ etc (Koning & Rochman, 2012).

3. Results and Discussions

In this work, the photoneutron cross-sections on all ten natural isotopes of the Sn nucleus were obtained. For each nucleus the inclusive (γ, n) cross-section was calculated taking into account the corresponding exclusive (γ, xn) ($x = 1, 2, 3, 4$) processes. The contribution of compound, direct and pre-equilibrium processes to the photoneutron inclusive cross-section was obtained. Separation into discrete and continuum processes was effectuated as well. If the isomer nuclei are obtained in final states then the isomer ratios were evaluated using the corresponding production cross-sections (σ_m , σ_g) in the exclusive $(\gamma, 1n)$ reactions. In the present work, the results of (γ, n) reaction on ^{114}Sn and ^{120}Sn nuclei are analyzed.

In multiple emission processes of type (γ, np) , $(\gamma, 2np)$, $(\gamma, n2p)$, $(\gamma, 3np)$, $(\gamma, n3p)$ and others with participation of charged particles a lot of isomer states are produced but they were not considered because their cross-sections usually have very low values due to the suppression of nuclear Coulomb field

In the Figures 1 and 2 the photoneutron cross-sections for the $^{114}\text{Sn}(\gamma, n)^{113}\text{Sn}$ and $^{120}\text{Sn}(\gamma, n)^{119}\text{Sn}$ are shown. In the Figures 1.a and 2.a are represented the (γ, n) inclusive reactions. The inclusive (γ, n) reactions can be interpreted like a superposition of $(\gamma, 1n)$, $(\gamma, 2n)$, $(\gamma, 3n)$ exclusive processes. The experimental data from Fig. 1.a) (Varlamov & all, 2010), (Sorokin & Yurev, 1974) and 2.a) (Varlamov & all, 2010), (Sorokin & Yurev, 1974) (curve 5) are described very well by $(\gamma, 1n)$ exclusive reactions.

In Figures 1.b) and 2.b) the inclusive (γ, n) cross-section is separated in the compound and direct processes. We have taken into account the pre-equilibrium processes, as the evaluations showed they can be neglected. In Figures 1.c) and 2.c) the exclusive (γ, n) cross-section is represented as a sum between discrete and continuum processes. At low energies the processes on discrete levels are predominant but with the increase of energy, the role of continuum processes becomes more important. In Figures 1.d) and 2.d) the cross-sections of $^{114}\text{Sn}(\gamma, n)^{m,g}{}^{113}\text{Sn}$, and $^{120}\text{Sn}(\gamma, n)^{m,g}{}^{119}\text{Sn}$ reactions which correspond to the production of isotopes in the ground and isomer states of ^{113}Sn and ^{119}Sn nuclei are shown.

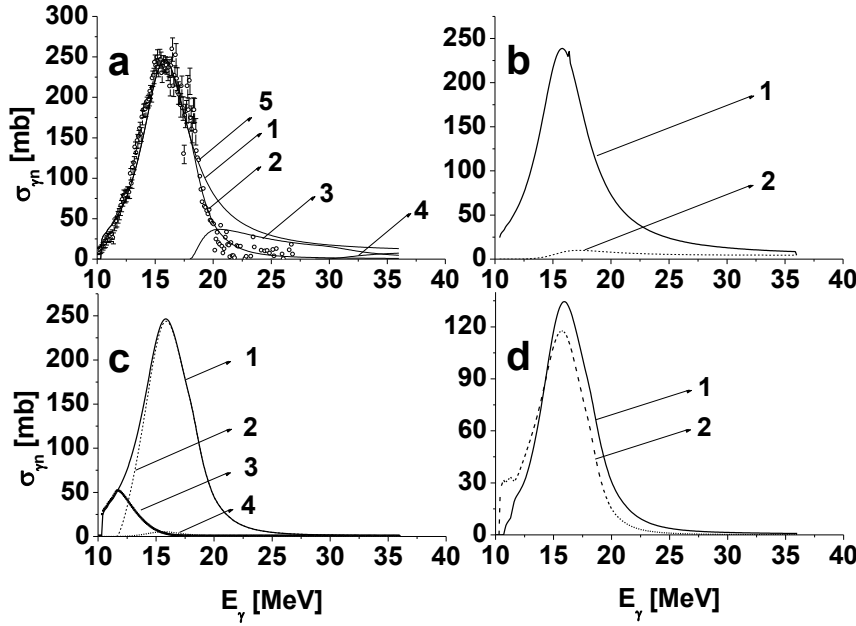


Figure 1. Cross-section of $^{114}\text{Sn}(\gamma, n)^{113}\text{Sn}$ reaction. a) 1 – inclusive cross section (γ, n); 2,3,4 – exclusive processes (γ, xn), ($x = 1,2,3$); 5 – Comparison with experimental data; b) Decomposition into (1) compound and (2) direct processes; c) Contribution of discrete (2) and continuum states (3) to exclusive ($\gamma, 1n$) process (1); d) Isotopes production of $^{113m,g}\text{Sn}$ of ground (1) and isomer states (2), respectively

At this stage, having the cross-sections of isomer and ground states production (see Figures 1.d and 2.d), the isomer ratios can be easily calculated using relation (1). In order to be closer to the experimental results, it is necessary also to use an appropriate model of the X-ray source. The isomer ratios were calculated for the isotopes from Table 1 obtained in the reactions from column three of the same Table. In Table 1 the main properties of nuclei considered in the calculation of isomer ratios are shown (Firestone & all, 1996).

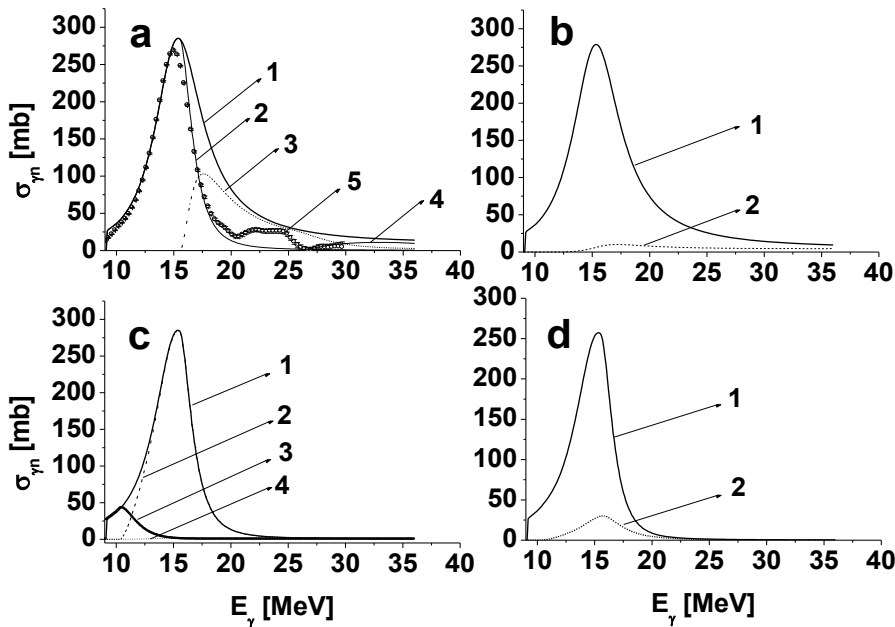


Figure 2. Cross-section of $^{120}\text{Sn}(\gamma, n)^{119}\text{Sn}$ reaction. a) 1 – inclusive cross section (γ, n); 2,3,4 – exclusive processes (γ, xn), ($x = 1,2,3$); 5 – Comparison with experimental data; b) Decomposition into (1) compound and (2) direct processes; c) Contribution of discrete (2) and continuum states (3) to exclusive ($\gamma, 1n$) process (1); d) Isotopes production of $^{119m,g}\text{Sn}$ of ground (1) and isomer states (2), respectively

Table 1.

Spin, parity, and time of life of Sn isotopes in isomer (m) and ground (g) states

Isotope	Reaction	Isomer (m)		Ground (g)	
		J^π	τ_m	J^π	τ_m
1 ^{113}Sn	$^{114}\text{Sn}(\gamma, n)^{113\text{m},\text{g}}\text{Sn}$	$(7/2)^+$	21.40 m	$(1/2)^+$	115.9d
2 ^{117}Sn	$^{118}\text{Sn}(\gamma, n)^{117\text{m},\text{g}}\text{Sn}$	$(11/2)^-$	13.60 d	$(1/2)^+$	Stable
3 ^{119}Sn	$^{120}\text{Sn}(\gamma, n)^{119\text{m},\text{g}}\text{Sn}$	$(11/2)^-$	293.1 d	$(1/2)^+$	Stable
4 ^{121}Sn	$^{122}\text{Sn}(\gamma, n)^{121\text{m},\text{g}}\text{Sn}$	$(11/2)^-$	55.00 y	$(3/2)^+$	27.06 h
5 ^{123}Sn	$^{124}\text{Sn}(\gamma, n)^{123\text{m},\text{g}}\text{Sn}$	$(3/2)^+$	40.60 m	$(11/2)^-$	129.2 d

Evaluating the incident X-ray flux is a rather complex issue. One simple realistic model, proposed a long time ago, based on accelerator data and known as Kramers relation was proposed (Kramers, 1923). According to this model, the incident photon flux is:

$$\phi(T_e, E_\gamma) \sim I_c = i_b Z (T_e - E_\gamma) E_\gamma^{-1} \quad (4)$$

where: I_c = intensity of gamma quanta; i_b = electrons beam current; T_e = energy of accelerated electrons; Z = charge of stopping element.

Many authors are using Monte Carlo simulation together with the Schiff model (Schiff, 1951). In the present evaluations of isomer ratios, the following cases have been used. In the first case, the incident flux is considered constant with different maximal energy (R_1 , R_2). In the second case an X-ray source with a flux based on the Kramers formula was considered (R_3) (Kramers, 1923). In the third case (R_4), Bremsstrahlung spectra $W(T_e, E_\gamma)$ as function electrons ($T_e[\text{MeV}]$) and gamma's ($E_\gamma[\text{MeV}]$) energies, respectively, was modeled with GEANT and confirmed by the calculations database of Moscow State University (MSU) (Varlamov & all, 2010).

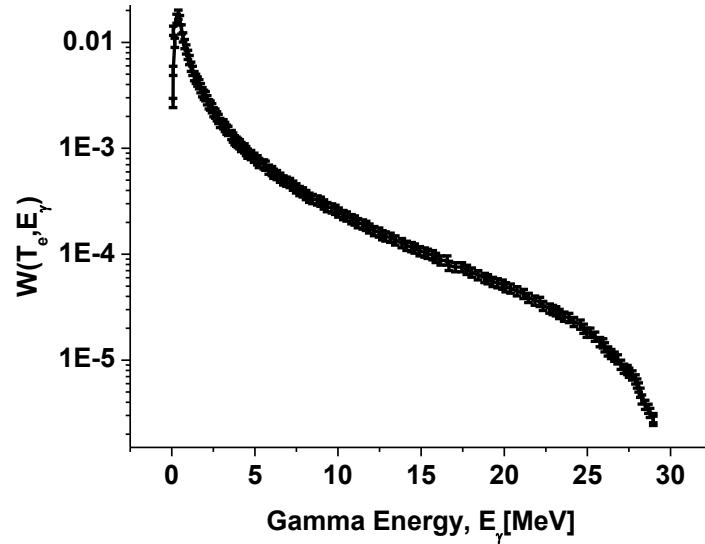


Figure 3. *Bremsstrahlung spectra as a function of gamma quanta energy, E_γ . Electrons energy, $T_e = 30 \text{ MeV}$*

The MSU database contains spectra from 10 to 80 MeV electrons energy with a step of 10 keV measured at Microton. The $W(T_e, E_\gamma)$, X-ray spectra, is represented in Figure 3 and it was obtained for the following conditions: the thickness of the Tungsten target is 2.5 mm, the maximum electron energy is $T_e = 30 \text{ MeV}$, and the intensity of electrons beam is 100 μA with an average power of the facility equal with 2.5 W. For all cases, isomer and ground production

cross-sections were obtained by using Talys software (Fig 1.d and 2.d). Results on isomer ratios are shown in Table 2.

Table 2.

Isomer ratios results. R_1) $\phi = 1; E_m = 35 \text{ MeV}$. R_2) $\phi = 1; E_m = 25 \text{ MeV}$. R_3) $\phi \sim (T_e - E_\gamma)/E_\gamma$; $E_m = 25 \text{ MeV}$; $T_e = 30 \text{ MeV}$. R_4) $W(T_e, E_\gamma)$ from Fig. 3; $E_m = 25 \text{ MeV}$; $T_e = 30 \text{ MeV}$

	Reaction	R_1	R_2	R_3	R_4
1	$^{114}\text{Sn}(\gamma, n)^{113\text{m},g}\text{Sn}$	1.142 ± 0.203	1.121 ± 0.173	0.995 ± 0.159	0.874 ± 0.23
2	$^{118}\text{Sn}(\gamma, n)^{117\text{m},g}\text{Sn}$	0.081 ± 0.015	0.0768 ± 0.012	0.062 ± 0.010	0.055 ± 0.019
3	$^{120}\text{Sn}(\gamma, n)^{119\text{m},g}\text{Sn}$	0.131 ± 0.025	0.126 ± 0.022	0.104 ± 0.018	0.098 ± 0.027
4	$^{122}\text{Sn}(\gamma, n)^{121\text{m},g}\text{Sn}$	0.258 ± 0.052	0.243 ± 0.045	0.170 ± 0.030	0.138 ± 0.038
5	$^{124}\text{Sn}(\gamma, n)^{123\text{m},g}\text{Sn}$	4.075 ± 0.532	4.187 ± 0.785	5.078 ± 0.954	4.937 ± 1.057

The isomeric ratios were obtained numerically based on relations (1-4), simulated spectra from Fig. 3 and calculated cross-sections, starting from neutron threshold up to maximal energy, using 0.1 MeV step. The errors of isomer ratios indicated in Table 2 are the results of the fact that the integrals from (1) are approximated by sums with a given step and numerically evaluated cross-sections.

All cross-sections were calculated from neutron threshold up to 35 MeV and therefore for R_1 the photons maximum energy (E_m) was chosen as 35 MeV . In real situations the photons flux is decreasing with energy as is reflected in expression (4) then R_2 and R_3 are obtained for $E_m = 25 \text{ MeV}$. From relation (4) it is easy to see that if the electrons energy, $E_0 = 35 \text{ MeV}$, then the highest value of incident gamma is also, $E_\gamma = 35 \text{ MeV}$, but the intensity of incident photons is zero. Therefore, in order to obtain reliable experimental data, the photons maximum energy must be not greater than $25 - 30 \text{ MeV}$. In the case of modeled gamma spectra from column R_4 results are close to R_3 but lower than R_1, R_2, R_3 as is expected. Higher errors from R_4 than R_1, R_2, R_3 are coming from the relative errors, of the order of $10 - 15\%$ of gamma spectra from Fig. 3.

From Table 2 we see that if the spin of isomer states is greater than the spin of the ground state then the isomer ratio is less than one. The isomer ratios are greater than one if vice versa. This is given by the selection rules because the levels with high spin are less populated than those with lower spin values. The exception seems to be the $^{114}\text{Sn}(\gamma, n)^{113\text{m},g}\text{Sn}$ reaction but in this case a) the isomer ratio has a value close to one; b) the difference between spins values of isomer and ground states are not so high and c) in the columns R_3, R_4 which are close to real measurement, the isomer ratios is lower than 1.

4. Conclusions

Developments of experimental devices, techniques, and facilities using very intense photons source of last decades give the possibilities to verify and improve old experimental data and to obtain new ones related to excited nuclei and isomer states in nuclear reactions induced by photons in the GDR region (Camera & all, 2016). A significant example is the ELI-NP facility with the corresponding scientific thematic plan.

The present results are part of nuclear data and isotopes production programs at FLNP JINR Dubna facilities. Nuclear reactions involving neutrons in the incident and emergent channels are traditionally investigated experimentally at JINR Dubna (Oprea & all, 2011).

In this work, the cross-sections and isomer ratios of photoneutron reactions on Sn isotopes in the GDR region were evaluated. The cross-sections were obtained using the standard Talys input. For isomer ratios calculations, some models for incident gamma flux were used.

For each evaluated cross-section, the contributions of compound, direct, and pre-equilibrium processes, and also the weights of discrete and continuum states of the residual nucleus were

obtained. A good agreement between cross-sections theoretical evaluations and experimental data from literature was observed. Considering the good description of experimental cross sections data, isomer ratios were calculated.

In the future, cross-sections measurements and isomer ratios from Table 2 are planned together with computer modeling of the experiments, as part of the scientific program on isotopes production, at FLNP JINR Dubna basic facilities.

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