Isomer ratios for products of photonuclear reactions on Rh

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Abstract. Over the past several years various preequilibrium models for nuclear reaction mechanisms description were developed. Diversified detailed experimental data in the medium excitation energy region for nuclei are needed for reasonable selection among these theoretical models. Lack of experimental data in this energy region does essentially limit the possibilities for analysis and comparison of different preequilibrium theoretical models. For photonuclear reactions this energy range covers 30–100 MeV. Experimental measurements and estimations of isomer ratios for products of photonuclear reactions with multiple particle escape on antimony were performed using bremsstrahlung spectrum as projectile with end-point energies 74.9 and 85.7 MeV. Method of the induced activity measurement was applied. For acquisition of gamma spectra we used HPGe spectrometer with 20% relative efficiency. Linear accelerator of electrons LU-40 was a source of bremsstrahlung. Energy resolution of electron beam was about 1% and a mean electron current varied within (3.8 – 5.3)$\mu$A.

1 Introduction

Using high energy gamma-quanta as projectiles in nuclear reactions has some essential advantages to study the nuclear structure and nuclear reaction mechanisms. The energy region of photonuclear reactions between Giant Dipole Resonance (GDR) and pion-producing threshold (from about 30 to about 100 MeV of gamma-quanta energy) is not sufficiently studied both theoretically and experimentally [1].

During last several years an essential progress has been achieved in development of the new theoretical models for the photonuclear reactions and in refinement of the existing ones in the considered energy region. The quasi-deuteron model was further improved [2], some new preequilibrium models have been developed for description of the multi-particle emission [3]. Permanently growing interest in Accelerator Driven Systems and progress in the design of high intensity quasi mono-energetic gamma-quanta sources [4] also stimulates studying the photonuclear reactions and in refinement of the experimental data in the energy range (30-100) MeV for testing newly developed and available theoretical models was the major motivation for the present work.

The main purpose of this study is to obtain the experimental isomer ratios for nuclei $^{103}$Rh as the products of the $^{103}$Rh($\gamma,4n$)$^{99m}$Rh reaction. Bremsstrahlung source used in this work has high intensity which provided opportunity to study ($\gamma,4n$) reaction with it’s relatively low yield.

2 Methodology

De-excitation time of nuclei by the $\gamma$-cascade irradiation usually does not exceed $10^{-12}$s [5]. In some cases transitions between levels of nucleus are suppressed due to the large difference of angular momentum of these levels involved and the nucleus can live long enough in a specific state called the isomer state. Usually this isomer state doesn’t have large excitation energies and its angular momentum differs from a spin of the ground state by a few units of $\hbar$.

The isomer or ground levels with large values of spin are populated mainly from highly excited states with large spin values. Population of isomer or ground levels with smaller values of spin can occur mainly from highly excited states with small values of spins. Therefore investigations of relative populations of the isomer and ground states [4, 6] can be very useful to derive spins of highly excited levels and to study the de-excitation mechanisms via gamma emission.

For mono-energetic gamma-ray beam with energy $E$ the isomer ratio is determined as the cross sections ratio $\sigma_m(E)/\sigma_g(E)$, where $\sigma_g(E)$ is the cross section of the photonuclear reaction leading to the formation of the ground state, $\sigma_m(E)$ is the cross section for the same nucleus leading to the formation of the isomer state. Also the isomer ratio is often determined as a ratio of the cross section $\sigma_H$ for the state with higher spin to the cross section $\sigma_L$ for the state with lower spin:

$$\xi = \frac{\sigma_H}{\sigma_L} \quad (1)$$

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If a gamma-ray beam is non-monoenergetic (this is the case for experiments with bremsstrahlung sources), the isomeric yield ratio is then determined as:

\[ d(E_{\text{max}}) = \frac{Y_m}{Y_g} \quad (2) \]

where the reaction yield \( Y \) is given by expression

\[ Y_{m,g} = N_i \int \sigma_{m,g}(E) \cdot W(E, E_{\text{max}}) dE \quad (3) \]

with \( N_i \) - number of the target nuclei, \( Y_{m,g} \) - reaction yield for nucleus in the isomer (m) or ground (g) state, \( E_{\text{max}} \) - maximal gamma energy, \( W(E, E_{\text{max}}) \) - bremsstrahlung spectrum, \( \sigma_{m,g}(E) \) with \( i = m, g \) - reaction cross sections for nucleus to be formed in meta-stable (ground) state for gamma energy \( E, E_{m,g}^{i} \), \( i = m, g \) - the energy threshold of the reaction leading to the meta-stable (ground) state.

The production of isomeric pair and its decay can be described by the following differential equation system:

\[
\begin{align*}
\frac{dN_i}{dt} &= Y_m - \lambda_m \cdot N_i \\
\frac{dN_g}{dt} &= Y_g - \lambda_g \cdot N_g + p \cdot \lambda_m \cdot N_m,
\end{align*}
\]

(4)

where \( N_i \) - population of i-state \( i = m \) - isomer state, \( i = g \) - ground state; \( Y_i \) - reaction yield according to (3), \( \lambda_m, \lambda_g \) - decay constants for isomer and ground state, \( p \) - branching factor (transition probability from isomer to ground state) \[7, 8\].

The differential equations system is valid under the following conditions: gamma-quanta flux is time invariable; the isomer level decays by gamma transition to the ground state in competition with \( \beta \)-decay and the ground state decays by \( \beta \)-decay branch. Solution of system (4) is as follows:

\[
\begin{align*}
S_m &= Y_m \Lambda_3 \Lambda_6 \Lambda_9 \\
S_g &= Y_g \Lambda_2 \Lambda_4 \Lambda_8 + Y_m(\Lambda_1 \Lambda_3 \Lambda_8 + \Lambda_1 \Lambda_2 \Lambda_8 + \Lambda_1 \Lambda_4 \Lambda_7) \\
\end{align*}
\]

(5)

where \( S_i, \ i = g, m \) – photo-peak area (in the gamma spectrum of the activation products), coefficient \( C \) includes self-absorption factor, true coincidence effects of cascade gammas and other effects, \( \epsilon \) - full efficiency of gamma detection for the analysed gamma-line; \( f_i, i = g, m \) – quantum yield of gamma-line for \( i \)-state decay (transition probability for this line); coefficients \( \Lambda_j, j = 1, 9 \) are defined by \( t_1, t_2, t_3 \) - irradiation time, cooling time and measurement time, respectively and determined in the ref \[9\].

As a result, the following expression is obtained

\[ F = Y_m X + Y_g \quad (6) \]

where \( F \) and \( X \) are defined as

\[ X = \frac{(\Lambda_1 \Lambda_3 \Lambda_6 \Lambda_9 + \Lambda_1 \Lambda_2 \Lambda_6 \Lambda_9 + \Lambda_1 \Lambda_4 \Lambda_6 \Lambda_9) + 2 \Lambda_1 \Lambda_6 \Lambda_9}{\Lambda_2 \Lambda_4 \Lambda_8} \quad (7) \]

with \( S = S_g + S_m \) peak area sum, \( Y_m,g \) - values, proportional to reaction yields.

The isomer yield ratio was calculated by fitting the experimental data \( (X, F) \) using the expression (6).

Experimentally, the method of the induced activity was applied to obtain the isomer ratios. Similar approach already was used by our group for some other target nuclei and showed very good results \[10–13\]. Experimental measurements and calculations of isomer ratios for the products of photonuclear reactions on rhodium with escape of several particles were performed with application of bremsstrahlung with 74.9 and 85.7 MeV endpoint energies. Metallic rhodium targets to study the reaction \(^{103}\)Rh(\(\gamma,4n\))\(^{99m}\)Rh were irradiated. Linear accelerator LU-40 (Research and Development Complex “Accelerator” NSC KIPT) was used as a source of fast electrons \[14\]. Instability of electron beam intensity was within 2%. Inner monitor of electron beam was calibrated by values from Faraday cup of the magnetic analyzer, placed at the accelerator outlet. The tantalum converter with 1.05 mm thickness was installed on the exit window of the accelerator facility, closely to which the cylindrical aluminium gamma absorbers (thickness 5.5 and 10 cm) were installed. Diameter of beam spot on the conversion target was less than 9 mm. Energy of electron beam was determined using the magnetic analyzer and was double checked in the low energy region by detailed considering the photonuclear reaction thresholds. A distance between tantalum converter and absorber was 2 cm, between tantalum converter and target – 20 cm. Irradiation time for every sample was 20 and 60 min. Then within (3 – 8) seconds the irradiated sample was moved with pneumatic transfer system to the counting area. HPGe detector with the energy resolution <2.0 keV for \(^{60}\)Co \( \gamma \)-line 1332 keV was used to acquire the instrumental gamma-ray spectra of the activation products as a set of serial measurements with various time periods. Cooling times varied from 5 seconds and a few days. Distances between sample and detector (dozens centimetres just after irradiation and few centimetres at the end of counting period) were chosen to optimize both statistics and time restrictions when large contribution of interfering reactions took place. A minimal distance was limited by condition of negligible contribution of cascade summing effects. Efficiency calibration of spectrometer was carried out for each detector-to-sample distance. The efficiency-energy dependence in double logarithmic scale showed a good quality and linearity in the energy range of interest with deviations between experimental data and linear fitted values not exceeding 2%.

3 Results and discussion

The isomer ratios were calculated as \( IR(E_i) = \frac{Y_i(E_i)}{Y_0(E_i)} \), where \( Y_i(E_i) \) is the reaction yield for the state of fi-
nal nucleus with larger angular momentum (meta-stable state), \( Y_i(E) \) is the reaction yield for the state of final nucleus with smaller angular momentum (ground state). Gamma transitions and corresponding lines with the energy 618 keV, EC+β+ decay of the isomer state (common line for decay of the ground and isomer states) were used to calculate the isomer ratio for \(^{99m}\)Rh from the \(^{103}\)Rh(γ,4n) nuclear reaction.

While decaying, the both states of \(^{99m}\)Rh irradiate γ-quanta with 618 keV energy. Peaks, corresponding to these quanta were used in our research. One of the obtained spectra is shown in Fig. 1. peak on 618 keV is clearly distinguishable. After peak areas determination, \( X \) and \( F \) variables were calculated for each of the obtained spectrum. They are presented in Fig. 2. We used the decay scheme of the \(^{99m}\)Rh nucleus from \([5]\).

The examples of the instrumental gamma-ray spectra due to the induced activities counting for the rhodium target are shown in Fig. 1. The gamma line 618 keV (common line EC+β+ decay of the ground and isomer state) was used to obtain the isomer ratio for \(^{99m}\)Rh \((^{103}\)Rh(γ,4n)^{99m}\)Rh\).

One can see Fig. 2 the fitting result of the decay line in presentation \((X, F)\) according to Eqs. (6, 7).

The experimental values of the isomer ratios, obtained by our group and other authors are presented in the Table 1. The spins of the target nucleus are \(1/2\), the spins of the meta-stable and ground states are 9/2+ and 1/2−, correspondingly.

The uncertainties, given in table 1 include contributions from photopeak efficiency calibration (up to 1.5%), abundance, geometry configuration (up to 3 % along the detector axis and up to 1 % in transvers direction) and intensities of gamma-rays (photopeak areas). Statistical uncertainties of photopeak areas made the main contribution to total uncertainty of result. Also number of experimental points of \( F, X \) dependency and range of \( X \) determination influenced total experimental error. These values were limited by experimental conditions.

**Table 1.** Reactions, the bremsstrahlung energy end-points \( E_{\gamma, \text{max}} \) and the experimental isomer ratios \( IR = \frac{Y_{\text{g}}}{Y_{\text{m}}} \) for nucleus, obtained in our experiments.

| \( E_{\gamma}, \text{MeV} \) | \( IR(E_{\gamma}) = \frac{Y_{\text{g}}(E_{\gamma})}{Y_{\text{m}}(E_{\gamma})} \) | Reference |
|-------------------------|---------------------------------|------------|
| 55                      | 1.59 ± 0.29                     | [15]       |
| 60                      | 1.39 ± 0.24                     | [15]       |
| 65                      | 1.43 ± 0.20                     | [16]       |
| 74.9                    | 1.60 ± 0.19                     | Present work|
| 85.7                    | 1.44 ± 0.30                     | Present work|

We used the code TALYS \([17]\) for theoretical calculations of isomer ratios for investigated nucleus Fig. 3. Bremsstrahlung spectrum was used from \([18]\).

Calculations of isomer ratios using code TALYS with various model approaches: LD1 - Constant temperature + Fermi gas model; LD2 - Back-shifted Fermi gas model; LD3 - Generalised superfluid model; LD4 - Microscopic level densities (Skyrme force) from Goriely’s tables; LD5 - Microscopic level densities (Skyrme force) from Hilaire’s combinatorial tables; LD6 - Microscopic level densities (temperature dependent HFB; Gogny force) from Hilaire’s combinatorial tables.
Figure 3. Solid line – calculation with using of code TALYS, red filled circles – experimental data obtained in this work, black circles – experimental values from [15][16]

4 Conclusion

The experimental values of the isomer ratios are obtained for $^{99m}_{1,2,3}$Rh nuclei as products of the photonuclear reaction $^{103}$Rh($\gamma,4n)^{99m}_{1,2,3}$Rh, using bremsstrahlung endpoint energies in the region from 74.9 to 85.7 MeV.

Experimental values of isomer ratios for $^{99m}_{1,2,3}$Rh from the reaction $^{103}$Rh($\gamma,4n)^{99m}_{1,2,3}$Rh are in a fairly good agreement with theoretical calculation results, especially for LD1, LD2 and LD3 models and also other experimental data.

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