

Measurement and estimation of isomeric cross section of $^{137}\text{Ba}(n, n')^{137\text{m}}\text{Ba}$ reaction using accelerator based neutron source



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ABSTRACT

The isomeric cross section of $^{137}\text{Ba}(n, n')^{137\text{m}}\text{Ba}$ reaction over the neutron energy from 1 keV to 4 MeV has been calculated using TALYS-1.2 nuclear code. The variations of cross section with level densities and effective imaginary potentials have been studied to obtain the best fit with the excitation function. The reaction cross section for Barium was also measured in continuous neutron energy using the Microtron accelerator based neutron source. In this case, the bremsstrahlung radiation emitted by impinging 6 MeV electrons on the $e-\gamma$ primary target (tungsten) was allowed to fall on the $\gamma-n$ secondary target (beryllium) to produce continuous neutrons and further barium was irradiated at 0° with respect to the incoming neutrons. It is observed that the experimental cross section for Barium is in good agreement with TALYS code and also evaluated data of EXFOR. Deviation factor has been determined for measuring and theoretical cross sections by TALYS-1.2 found to be around 14.71.

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1. Introduction

In concern to the theoretical and experimental studies of neutron interaction with matter, the neutron activation cross section data are having importance. Neutron passing through matter loses energy and having wide range of energy spectrum. Therefore, the cross section data of nuclear reactions induced by different energy neutrons are required for several applications in areas of applied nuclear physics, nuclear models, elemental analysis, reactor technology, etc. (Marie et al., 2006; Luo et al., 2009). Therefore, an electron accelerator based neutron source which produces continuous neutron energy and having useful for medical and industrial applications because of their compactness, easy handling, adjustable flux, no radioactive waste, less shielding requirement, etc. (Bayanov et al., 1998). However, very few attempts have been made to use these kinds of accelerators based neutron source for nuclear reaction studies (Agbemava et al., 2011; Barough et al., 2013).

Barium sulfate (BaSO_4) is having a high coefficient of absorption for gamma radiation and X-radiation, because it is being used very often in concrete (barite concrete, barite cement) that screens the nuclear reactors and in addition, it also contains in numerous radio-opaque substances especially, in radio-opaque barite. Moreover, multi-commutation in flow-injection spectrophotometric method that exploits long path length for determination of sulfate in natural water samples using a solid-phase reactor containing

barium chloranilate immobilized in a polyester resin is also reported. This procedure is based on the formation of insoluble barium sulfate (BaSO_4) in the solid-phase (Bonifácio et al., 2011). The effects of the reactor length ($\sim 4\text{--}10$ cm), particle size ($\sim 350\text{--}500$ μm), and internal diameter (~ 3 cm) of the reactor on performance of the solid-phase reactor containing barium chloranilate are important (Bonifácio et al., 2011). Therefore, purpose of the present work is to measure and calculate the (n, n') cross section in continuous neutron energy range of 1 keV to 4 MeV from electron accelerator based neutron source. To obtain continuous neutrons, bremsstrahlung radiation emitted by impinging 6 MeV electrons on the $e-\gamma$ target is allowed to fall on the $\gamma-n$ target. These neutrons have a continuous energy spectrum and have been used for the present experiment. Moreover, the cross section of $^{137}\text{Ba}(n, n')^{137\text{m}}\text{Ba}$ reaction has been calculated by TALYS-1.2 nuclear code. The experimentally measured and theoretically calculated cross section has been compared with the cross sections from EXFOR database.

2. Experimental details

2.1. Experimental setup

In the present work, a 6 MeV Race-Track Microtron accelerator based neutron source from the Department of Physics, University of Pune was used. The Microtron, Bhoraskar (1988) is a re-circulating electron accelerator in which an electron beam repeatedly passed through RF accelerating cavity to produce 6 MeV energy

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electron beam (Asgekar et al., 1980). Pulsed current of the electron beam was 1–10 mA has pulse width 2.0 μ s and pulse rate 50 PPS. Hence, an average current of the electron beam can be in the range of 0.1 and 1 μ A. The high energy electrons interact with e- γ target (tungsten) and produces bremsstrahlung radiation which further gives continuous neutron energy from γ -n target (beryllium). The electron beam having energy 6 MeV is allowed to fall on the primary cylindrical tungsten target ($r = 0.3$ cm and thickness = 0.22 cm) and generates bremsstrahlung radiations. The tungsten target is mounted on extraction window of the Microtron accelerator. Further, these bremsstrahlung radiations are allowed to fall on the secondary beryllium target having dimensions 10 cm \times 10 cm \times 5 cm, which generates continuous neutrons through the photo nuclear reaction. The experimental setup for the measurement of the cross section of (n, n') reaction is shown in Fig. 1. The angular distribution of Microtron accelerator based neutron source has been measured experimentally and compared with FLUKA simulation (Patil et al., 2010). From the results, it has been observed that the source produces a maximum neutrons flux in the forward direction. Therefore, for the present experiment sample of which the (n, n') reaction cross section to be measured was kept at 0°. The neutron energy spectrum is estimated at the sample place in FLUKA and is shown in Fig. 2. It is observed from Fig. 2 that the continuous energies of neutrons coming out from the beryllium having an energy range from 1 keV to 4 MeV and the total neutron flux was experimentally measured through $^{51}\text{V}(n, \gamma)^{52}\text{V}$ reaction around to be 1.0×10^5 n/cm²-s at 1 μ A current of the electron beam.

2.2. Samples

For the measurement of (n, n') reaction with Barium, powder of pure Barium Chloride (BaCl₂·2H₂O powder 99% purity) was used.

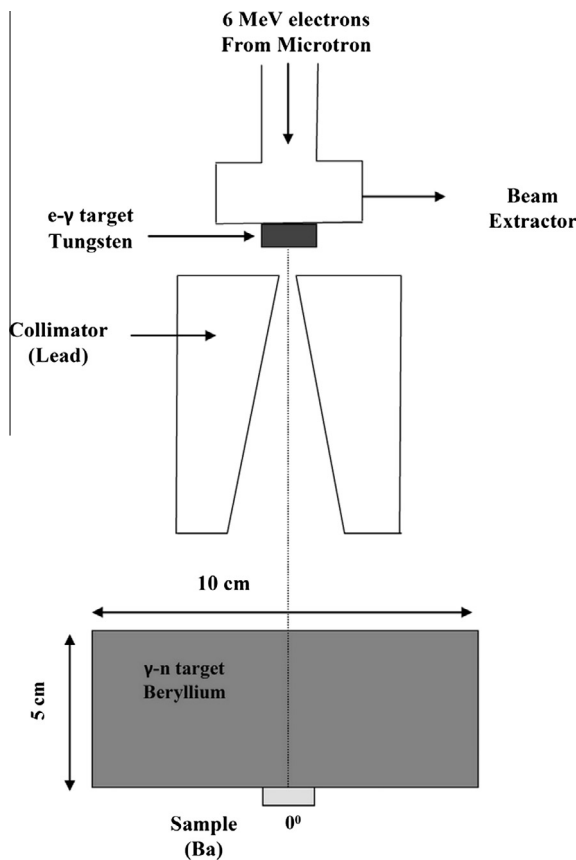


Fig. 1. Experimental setup for the measurement of nuclear reaction cross sections.

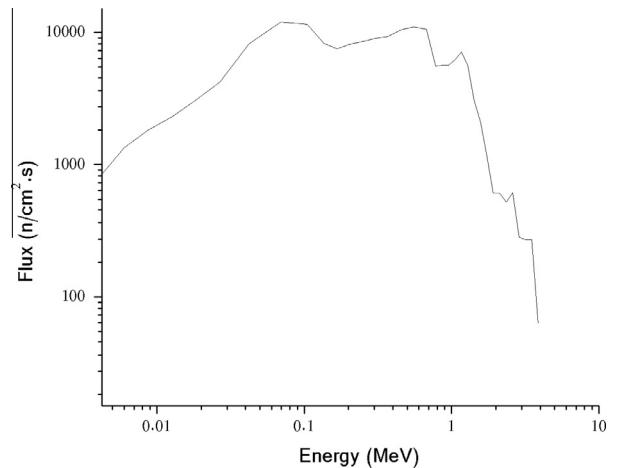


Fig. 2. Neutron spectra from the Microtron accelerator based neutron source (simulated by FLUKA).

The sample was prepared by packing a known weight in a polyethylene bag using micro-balance. The size of the sample was ~ 6 cm \times ~ 3 cm and ~ 0.1 cm thick for activation. The position of the sample for activation experiment is shown in Fig. 1.

2.3. Neutron irradiation and cross section calculations

The sample was irradiated by the accelerator based neutron source with an experimental setup shown in Fig. 1 for the measurement of nuclear reaction cross sections. In this case, 6 MeV electrons were allowed to fall on the tungsten target for bremsstrahlung production and further generated continuous neutron energy through the beryllium target. The sample of which (n, n') reaction cross section to be measured was kept at 0° with respect to the neutrons as shown in Fig. 1. After neutron irradiation, each sample was removed from the Faraday cup and was then taken it to the high purity germanium (HPGe) detector based gamma-ray spectroscopy for measurement of induced gamma activity. The HPGe detector was calibrated using standard gamma sources. The measured gamma spectrum was analyzed by Genie 2K software based multichannel analyzer (MCA). The achieved count rate can be used to determine activation integrals and other parameters of the nuclear reactions by making necessary corrections. The details of energies and branching ratios of the γ rays adopted in the cross-section estimation, the irradiation time, cooling time, and counting time are given in Table 1.

The cross section was estimated by the following activation relation (Curtiss, 1969),

$$\sigma = \frac{A\lambda}{\phi\beta N\epsilon(1 - e^{-\lambda t_1})e^{-\lambda t_2}(1 - e^{-\lambda t_3})} \quad (1)$$

where ϕ is the incident neutron flux, σ is the cross section for (n, γ) reaction, A is the total number of counts, λ is the decay constant, β is the number of gamma quanta/disintegration, N is the number of atoms in the target; ϵ is the efficiency of the detector (0.06), t_1 is the irradiation time, t_2 is the cooling time, i.e., the time between end of irradiation and start of counting, t_3 is the counting time. However, in the present case continuous neutron energy spectrum was used for the measurement of neutron reaction cross section. Therefore, the equation for continuous neutron energy can be written as

$$\sum_{i=1}^{i=n} \sigma_i \phi_i = \frac{A\lambda}{\beta N\epsilon(1 - e^{-\lambda t_1})e^{-\lambda t_2}(1 - e^{-\lambda t_3})} \quad (2)$$

Table 1

Nuclear reactions, along with their corresponding decay data of half life, energies, gamma intensity, irradiation time, cooling time, and counting time.

| Reaction | Half-life of reaction products (m) | γ energy (MeV) | γ intensity (%) | Irradiation time (s) | Cooling time (s) | Counting time (s) |
|---|------------------------------------|-----------------------|------------------------|----------------------|------------------|-------------------|
| $^{137}\text{Ba}(n, n')^{137\text{m}}\text{Ba}$ | 2.55 | 0.662 | 89 | 600 | 30 | 198.47 |

where i run from first neutron energy bin to last energy bin (1 to n).

The average cross section, $\bar{\sigma}$, over an interval, is the integral of the cross section multiplied by the flux divided by the integrated flux i.e. flux weighted average cross section, which can be written as:

$$\bar{\sigma} = \frac{\int_{E_1}^{E_2} \sigma(E) \varphi(E) dE}{\int_{E_1}^{E_2} \varphi(E) dE} \quad (3)$$

This equation in terms of summation can be written as:

$$\bar{\sigma} = \frac{\sum_{i=1}^{i=n} \sigma_i \varphi_i}{\sum_{i=1}^{i=n} \varphi_i} \quad (4)$$

Therefore, the following modified equation can be used to estimate the average cross section for continuous neutron energy spectrum (Barough et al., 2013),

$$\bar{\sigma} = \frac{A\lambda}{\varphi_{\text{integrated}} \beta N \varepsilon (1 - e^{-\lambda t_1}) e^{-\lambda t_2} (1 - e^{-\lambda t_3})} \quad (5)$$

Using Eq. (5) one can determine the experimental average cross section for Barium at continuous energy spectrum of neutrons.

2.4. Estimation of uncertainty and deviation of measured and theoretical cross section

The main uncertainty sources related to the present cross section measurement and their estimated values are given in Table 2. These uncertainties have been originated from the various sources. For example, Time: timer with 0.001 accuracy, Isotopic abundance: from table of isotope, Counting statistics: Gaussian distribution error calculation, Weight of sample: balance with 10^{-6} accuracy, Self-absorption of γ -ray: Debertin method (Jodłowski, 2006), The coincidence sum effect of cascade γ -ray: Genie software (L'Annunziata, 2012) and Detector efficiency: Poisson error method (Bevington and Robinson, 2003). Moreover, deviation factor has been applied in contrast with the results of theoretical calculation and measured cross section. In this case, following deviation factor has been used (Michel and Nagel, 1997; Broeders and Konobeyev, 2005):

$$H = \left(\frac{1}{N} \sum_{i=1}^N \left(\frac{\sigma_i^{\text{exp}} - \sigma_i^{\text{calc}}}{\Delta \sigma_i^{\text{exp}}} \right)^2 \right)^{1/2} \quad (6)$$

where σ_i^{exp} and $\Delta \sigma_i^{\text{exp}}$ are measured cross section and its uncertainty, σ_i^{calc} is calculation cross section, N is the number of experimental points. Therefore, deviation factor of cross section between measured and theoretical values has been calculated.

Table 2

Main uncertainty sources and their estimated values in the cross section measurements.

| Source of uncertainty | Uncertainty (%) |
|--|-----------------|
| Uncertainties of irradiation, cooling and counting times | 0.1 |
| Isotopic abundance | 0.1 |
| Counting statistics | 6 |
| Weight of sample | 10^{-4} |
| Self-absorption of γ -ray | ~ 0.5 |
| Coincidence sum effect of cascade γ -ray | 1 |
| Detector efficiency | 3 |

3. Theoretical calculations

In this study, by applying TALYS-1.2 nuclear code the cross section for the formation of $^{137\text{m}}\text{Ba}$ through $^{137}\text{Ba}(n, n')^{137\text{m}}\text{Ba}$ reaction over the neutron energy range from 1 keV to 4 MeV have been calculated. The basis used for TALYS nuclear structure database has been generated from the Reference Input Parameter Library RIPL (Capote et al., 2009). The local and global parameterizations of Koning and Delaroche are Optical model potentials (OMP) used in TALYS (Koning and Delaroche, 2003), Whereas, Hilaire and Goriely have been proposed the microscopic combinatorial model (Goriely et al., 2008). For the calculation of the competition of photons with other particles Gamma-ray transmission coefficients enter the Hauser–Feshbach model. In addition, in order to calculate pre-equilibrium, the two component exciton model by Kalbach (1986) is applied.

With excitation energy at least equal to the projectile separation energy in the compound system, the compound nucleus is formed in capture reactions. The number of radiative open channels is almost infinite as a result of the transmission coefficient calculation which involves all the possible states to which a photon can be emitted from the initial compound nucleus state, but each has a very small transmission coefficient.

By applying different choices of the level density parameters obtained from the Fermi gas model and Goriely's table, the cross section calculations have been carried out (Goriely et al., 2008). From the Reference Input Parameters Library (RIPL-2), the discrete energy level scheme has been adopted and such 25 discrete levels are considered to be below 2 MeV energy (Koning et al., 2009). For the RIPL database (Capote et al., 2009), on the basis of Hartree–Fock calculations Goriely has calculated level densities from drip line to drip line, which covers excitation energies up to 150 MeV and spin values I up to 30 (Goriely et al., 2008; Ericson, 1960). From the RIPL-2 developed at the IAEA, the optical model parameters of Koning et al. (2009) for neutrons (Lib. No. 1430) and protons (Lib. No. 4426) used in the study are extracted.

The Fermi gas level density is as follow under the assumption that the projections of the total angular momentum are randomly coupled (Ignatyuk et al., 1976):

$$\rho(E_x, J, \Pi) = \frac{1}{2} \frac{2J+1}{2\sqrt{2\pi}\sigma_{\text{eff}}^3} \exp\left(-\frac{(J+\frac{1}{2})^2}{2\sigma_{\text{eff}}^2}\right) \frac{\sqrt{\pi} \exp(2\sqrt{aU})}{12 a^{1/2} U^{5/4}} \quad (7)$$

where σ_{eff} is the spin cut-off parameter as a representation of the width of the angular momentum distribution and the factor $1/2$ represents the aforementioned equiparity distribution, while j is the nuclear spin and U is the excitation energy after making pairing correction.

In terms of the super-fluid models when TALYS specific parameterization of the level density 'a' parameter is selected and in addition the low-energy part of level densities is also calculated (Ignatyuk et al., 1976). Parameter 'a' is assumed to be energy (temperature) dependent which was calculated in Ignatyuk et al. (1976) as follows:

$$a(U) = \tilde{a} \left[1 + \delta w \frac{1 + \exp(-\gamma U)}{U} \right] \quad (8)$$

In which δw is the shell correction and \tilde{a} is the asymptotic value of a parameter. The asymptotic value \tilde{a} is given by

$$\bar{a} = \alpha A + \beta A^{2/3} \quad (9)$$

In addition, γ defined by

$$\gamma = \frac{\gamma_1}{\sqrt[3]{A}} + \gamma_2 \quad (10)$$

where A is the mass number, also $\alpha, \beta, \gamma_{1,2}$ are global parameters that over a whole range of nuclides have been determined to give the best average level density description.

With the pairing gap $\Delta = 12/(A)^{1/2}$, the critical temperature T_{crit} is

$$T_{crit} = 0.567\Delta \quad (11)$$

4. Nuclear models

By applying the nuclear model code TALYS-1.2, the excitation functions for reactions have been studied theoretically (Koning et al., 2009). Using a local potential proposed by Koning and Delaroche, the optical model parameters for neutrons and protons are obtained (Koning et al., 2009). The Hauser–Feshbach model is used for the calculation of the compound nucleus contribution (Hauser and Feshbach, 1952). For calculation of the pre-equilibrium contribution, the two-component exciton model developed by Kalbach (1986) is applied. In the exciton model, at any moment during reaction by the total energy and the total number of particles above and holes below the Fermi surface the nuclear state is characterized. The exciton model is a time-dependent master equation explaining the probability of transitions to more and less complex particle-hole states as well as transitions to the continuum emission (Gadioli and Hodgson, 1992).

The global parameterizations proposed by Koning and Delaroche have been applied in the optical model potentials for neutrons and protons used in TALYS-1.2 nuclear model code (Koning and Delaroche, 2003). For $12 < A < 339$ in the energy range 1 keV to 200 MeV, these optical potentials by Koning and Delaroche (2003) have been tested, whereas, Watanabe used the simple folding approach for alpha particles (Watanabe, 1958). The processes such as stripping, pickup, and knockout play important roles in deciding the cross sections in nuclear reactions involving projectiles and ejectiles with different proton and neutron numbers. The direct-like reactions are not entirely covered by the exciton model (Koning et al., 2009). The phenomenological theory developed by Kalbach (1986) has been added in the TALYS-1.2 nuclear model code in order to include the effects of these processes. The creation of neutron and proton type particles and holes are explicitly followed throughout the reaction in the two-component exciton model.

The level density parameters are calculated by applying five different choices of the level density model available in TALYS-1.2. With the only change been made in choice of the level density models, the theoretical calculations also include default parameter values. The exact angular momentum and parity coupling is observed. Along with the competing fission channel, the emission of neutrons, protons are taken into account. The full gamma-cascade in the residual nuclei is considered. Particular attention is dedicated to the determination of the level densities, which can be calculated in the non-adiabatic approach allowing for rotational and vibrational enhancements. Above certain energy, these collective effects are gradually removed. However, the dependence of the rotational enhancement on the shape of nucleus Level densities acquires dynamic features.

The model proposed by Hoering and Weidenmueller, an emission of gamma radiation in the Multi-step Compound (MSC) mechanism is treated (Hoering and Weidenmuller, 1992) through the de-excitation of the Giant Dipole Resonance (GDR) built within

classes. Following Brink-Axel hypothesis the model assumes that gamma emission occurs (Brink, 1957) each nuclear state serves as the basis of a GDR excitation with identical properties. Discrete level information is incomplete nuclear level densities and is used at excitation energies in statistical models for predicting cross sections (Herman et al., 2005). The determination of level densities can be calculated in the non-adiabatic approach allowing for the rotational and vibrational enhancements in which collective effects are gradually removed above certain energy (Ericson, 1960; Herman et al., 2005). The dynamic approach to level densities which is specific to TALYS-1.2 nuclear code takes into account collective enhancements of the level densities due to nuclear vibration and rotation. The super-fluid model below critical excitation energy and Fermi gas model is applied in the formalism (Koning et al., 2009).

5. Results and discussion

The isomeric cross section of $^{137}\text{Ba}(n, n')^{137m}\text{Ba}$ nuclear reaction induced by 1 keV to 4 MeV neutrons has been theoretically estimated using TALYS-1.2 nuclear model code. The cross sections were calculated by (i) different level density parameters, (ii) varying the effective imaginary potentials, and (iii) incorporating contributions of the negative parity energy levels. From the decay scheme of ^{137m}Ba shown in Fig. 3, it is observed that ^{137}Ba nuclei can decay from the negative parity metastable state 0.662 MeV ($11/2^-$) to stable by gamma emission involving M4 transition or of an electron whose energy is 0.617 less than the binding

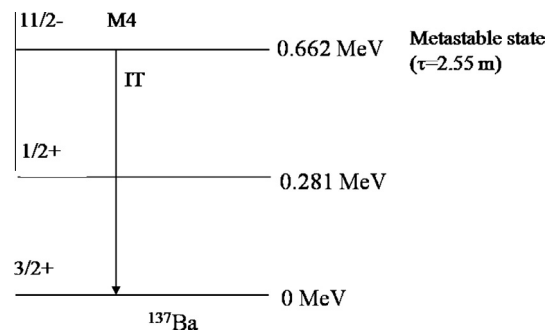


Fig. 3. Decay scheme of ^{137}Ba .

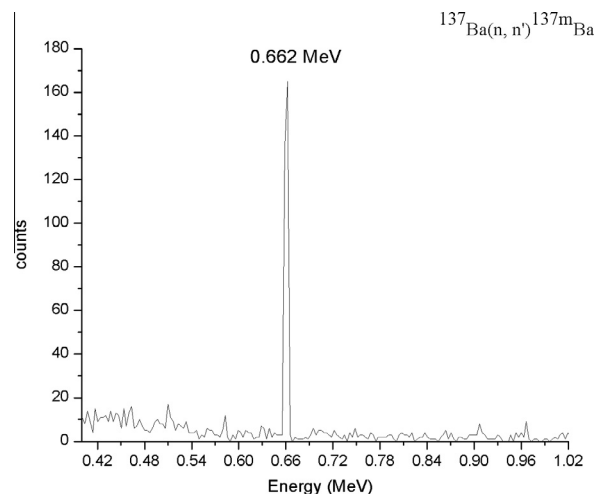


Fig. 4. Gamma-ray spectrum of ^{137m}Ba , produced through $^{137}\text{Ba}(n, n')^{137m}\text{Ba}$ reaction induced by continuous neutrons energy.

Table 3
Numerical values of theoretical and experimental cross sections for the $^{137}\text{Ba}(n, n')^{137m}\text{Ba}$ reaction.

| Reaction | Present work | |
|----------------------|--------------|---------------|
| | E_n (MeV) | σ (mb) |
| Present Experimental | 0.47 | 0.9 |
| TALYS-1.2 | 0.2485 | 0.03 |
| | 0.3045 | 0.07 |
| | 0.3707 | 0.2 |
| | 0.4528 | 0.5 |
| | 0.47 | 0.7 |
| | 0.553 | 1 |
| | 0.6754 | 4.90424 |
| | 0.7818 | 30.9023 |
| | 0.8641 | 50.0551 |
| | 0.955 | 67.3321 |
| | 1.056 | 101.382 |
| | 1.167 | 205.263 |
| | 1.289 | 264.002 |
| | 1.425 | 300.266 |
| | 1.575 | 336.081 |
| | 1.74 | 366.722 |
| | 1.923 | 415.619 |
| | 2.125 | 477.563 |
| | 2.349 | 511.356 |
| | 2.596 | 552.791 |
| | 2.869 | 622.745 |
| | 3.171 | 705.319 |
| | 3.504 | 793.042 |
| | 3.872 | 875.584 |

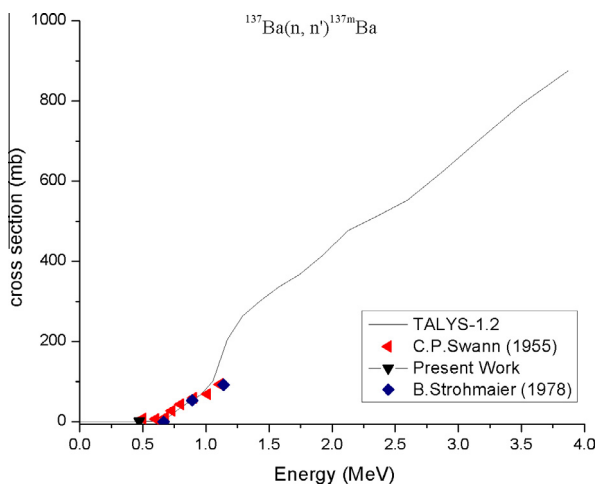


Fig. 5. Experimental and theoretical isomeric cross-sections for $^{137}\text{Ba}(n, n')^{137m}\text{Ba}$ reaction.

energy of electron in the atom. With ^{137m}Ba , about 9% of the decay events give conversion electrons with energy of 0.625 MeV and about 2% give conversion electrons with energies of 0.656 MeV. Fig. 4 shows a gamma-ray spectrum of the ^{137}Ba irradiated with continuous neutron energy up to 4 MeV in which photoppeak at 0.662 MeV due to ^{137m}Ba is observed. These results, therefore, indicate that ^{137m}Ba can be produced through $^{137}\text{Ba}(n, n')^{137m}\text{Ba}$ reaction induced by the continuous neutron energy up to 4 MeV.

The isomeric cross sections of the $^{137}\text{Ba}(n, n')^{137m}\text{Ba}$ calculated with TALYS-1.2 nuclear model code and experimentally measured values are given in Table 3. Fig. 5 shows isomeric cross section for the reaction $^{137}\text{Ba}(n, n')^{137m}\text{Ba}$ over the continuous neutron energy up to 4 MeV using TALYS-1.2 nuclear model code and further it also shows the cross section values measured by Strohmaier et al. (1978) and Swann and Metzger (1955) and the present one. There is an agreement between the measured, evaluated, and with all the

cross sections calculated using TALYS-1.2 nuclear code. The uncertainty in the measured cross section is being calculated and found to be around 6.8%. Therefore, the present measured average cross section is fairly in good agreement with the evaluated data of EXFOR. In addition, the deviation in measured and theoretical cross section has been calculated by using Eq. (6) and it is found to be around 14.71.

6. Conclusion

In this work, the isomeric reaction cross section for Barium has been measured using 6 MeV electron accelerator based neutron source and compared with the calculated cross sections by TALYS nuclear code. It is observed that the experimental cross section for Barium is in good agreement with the TALYS nuclear code. The uncertainty in measured cross section is found to be 6.8% and the deviation of measured and theoretical cross section is observed to be 14.71. The level density parameters and the adjusted value of the effective imaginary potential as input parameters to the TALYS nuclear code could yield isomeric cross section values close to the corresponding experimental values. The (n, n') reaction for ^{137}Ba has been experimentally measured and determined with relatively small uncertainties using the latest decay data. These new sets of measured data can help in fixing statistical model parameters to understand the type of reactions in terms of statistical models and to a better evaluation in the range of neutron energies from 1 keV to 4 MeV.

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