

EXCITATION FUNCTIONS OF RADIONUCLIDES PRODUCED BY PROTON INDUCED REACTIONS ON GADOLINIUM TARGETS

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Abstract

Cross section study for proton induced reaction on natural Gadolinium targets were performed. Excitation functions for the reactions ${}^{\text{nat}}\text{Gd}(p,x) {}^{152\text{m}+g,154\text{m},154\text{g}}\text{Tb}$ from threshold up to $E_p = 18$ MeV have been measured employing the stacked foil activation technique, and using high resolution HPGe gamma spectrometry. Utilizing the simultaneous measurement of the excitation function of ${}^{\text{nat}}\text{Cu}(p,x) {}^{62}\text{Zn}$, ${}^{\text{nat}}\text{Cu}(p,x) {}^{63}\text{Zn}$, and ${}^{\text{nat}}\text{Cu}(p,x) {}^{65}\text{Zn}$ as monitor reactions. The theoretical analysis of the excitation functions has been done employing both ALICE-91 and EMPIRE-II codes. In general, theoretical calculations agree well with the experimental data. A significant contribution of pre-equilibrium component has been observed at these energies.

Keywords: *Protons, Cross Sections, Nuclear Reactions, HPGe-Detector, Gadolinium, and Natural Targets.*

INTRODUCTION

The cross sections data for the proton-induced reactions on natural Gadolinium in the energy range from the threshold up to 18 MeV have not previously been measured. To the best of our knowledge the excitation functions for the reactions ${}^{\text{nat}}\text{Gd}(p,x) {}^{152\text{m}+g,154\text{m},154\text{g}}\text{Tb}$ have not been reported before. For that, present measured cross section data are of importance in the radionuclide production of short lived Tb isotopes using relevant nuclear reactions. As this experimental work has been done for the first time, there is continual interest in the use of modern nuclear reaction codes for the calculation of reaction cross sections and comparison with experiments. Recent investigations have clearly indicated that in statistical nuclear reactions, at moderate excitation energies, particles are emitted prior to the establishment of thermodynamic equilibrium of the compound nucleus (CN). The process is generally known as pre-equilibrium (PE) emission. Signatures of PE emission are often found in the high energy tails of the excitation functions. The PE emission mechanism has attracted considerable attention from both the experimental and theoretical viewpoints [1-3]. Semi-classical models [4-10] have been successfully used to describe the experimental data on PE emission. The computer codes ALICE-91 [9], and EMPIRE-II [11] have been employed for

the cross section calculations.

FORMULA FOR CROSS-SECTION DETERMINATION

We used the well known induced radioactivity cross section formula

$$\sigma_{\gamma} = \frac{\lambda C_p \exp(\lambda t_d)}{N_t \varphi \varepsilon_{\gamma} I_{\gamma} (1 - \exp(-\lambda t_i))(1 - \exp(-\lambda t_c))}, \quad (1)$$

where σ (cm²) is the cross section of formation of a particular γ transition with absolute branching ratio I_{γ} , N_t is the number of target atoms/cm², φ (sec⁻¹) is the incident particle flux, λ (sec⁻¹) is the decay constant of the induced radioactive nucleus, ε_{γ} is the peak efficiency of the radiation detector for the particular E_{γ} , t_i (sec) is the irradiation time, t_d (sec) is the activity decay time, t_c (sec) is the counting time, C_p is the net counts of full energy peak of the measured gamma-transition.

Expression (1) is conveniently written in the form

$$\sigma_r = \frac{\lambda C_p \exp(\lambda t_d)}{N_t \varphi \theta K G \varepsilon I_{\gamma} [1 - \exp(-\lambda t_i)][1 - \exp(-\lambda t_c)]}, \quad (2)$$

In the above expression θ is the branching ratio of the particular radiation, $G\varepsilon$ is detector's geometry dependent efficiency, K is the γ -ray self-absorption correction for the material of the sample and is given by:

$$K = \left[\frac{1 - \exp(-\mu d)}{\mu d} \right], \quad (3)$$

where μ is the γ -ray absorption coefficient for the sample and d is the thickness of the sample.

IRRADIATION OF TARGETS AND FLUX MONITORING

The irradiation of target stacks was performed for a period of one hour with initial proton beams of energy 15 and 18 MeV, respectively at the Inshas Variable Energy Cyclotron Facility, Nuclear Research Center, AEA, Cairo, Egypt. This facility has been installed and gotten into operation since October 2000. The facility was described before [12]. Each stack was composed of target Gd, Al, and Cu foil for flux monitoring. Two stacks were irradiated separately, the targets in the stacks consisted of 9, 10 thin foils of natural gadolinium of high purity (99.99%), respectively. The Al-degraders of suitable thickness were used in between the samples in each stack to cover the broad energy range and as a catcher.

The samples of gadolinium were of 10.0 $\mu\text{m} \pm 2.0\%$ thick which is equivalent to 7.901 mg/cm² $\pm 2.0\%$. Like targets in stacks, sets of thin Al-foils (thickness 10.0 μm) and Cu-foils (thickness 10.0 25.0 μm) were used to monitor the proton flux. The inner ones of each three foils for both target and monitor were used to prevent the cross contamination from other elements due to the destruction of foils by radiation damage or heating during irradiation, where the inner foils contamination cancel out. So during the investigation we unpack every three foils of same kind with together, this is due to intensity the recoiled atoms depend on the energy of penetrating particles, of coarse the reaction cross section.

The beam current was monitored directly on the Farady cup. The off-line counting of the irradiated samples was carried out using vertical coaxial closed end 70% HPGe γ -ray detector (resolution ≈ 2 keV for 1.33MeV γ -ray of ⁶⁰Co) coupled to the ORTEC's PC based multi-

channel analyzer. The background spectrum was also recorded and was properly subtracted from the sample counting rates. The detector was pre-calibrated using various standard gamma sources including the $^{152,154,155}\text{Eu}$ point source of known strength, which was also used for the determination of the geometry dependent efficiency at various source–detector distances. The residual nuclei were identified from their characteristic gamma lines as well as from their half-lives. The nuclear data required for the calculations were taken consistently from the Table of Isotopes [13]. From the measured intensities of the identified gamma rays, the cross-sections at different incident energies were computed.

At the beginning of the measurement, the beam was positioned on an empty stack holder, a Faraday Cup measured randomly the beam current. During irradiation, the Faraday cup connected with a digital meter coupled to a PC continuously recorded the beam currents. The contributions due to the fluctuation of proton flux and the secondary particles produced in the target stack was taken into account and found to be negligible. The flux for the corresponding target positions of different stacks was measured from the nuclide $^{62,63,65}\text{Zn}$ via the reaction $^{\text{Nat}}\text{Cu}(p,x)^{62,63,65}\text{Zn}$. For this measurement, the cross-sections for the standard reaction at specific proton energies were taken from the measurement [14]. The uncertainty of proton flux was mainly determined by the uncertainties of efficiency of detectors and the mass of the monitor foil. No uncertainties due to the monitor cross-sections were included in this calculation and the reason has been explained elsewhere [15,16]. The correction for the activity due to the self-absorption of γ -rays in the targets itself was taken into account very carefully.

The energy losses in the samples as well as degrader thickness were calculated employing the SRIM program [17], giving the initial proton energy of 15.0, 18.0 MeV. The foils in stacks were arranged in such a way that Al and Cu-foils were placed in front and behind of target foils respectively. In the calculation of the uncertainty of proton energy, the uncertainty due to its initial energy just leaving the accelerator, its energy spread and straggling passing through the target stacks was taken into account attentively. The resultant uncertainty for this contribution is less than 1%.

After the end of irradiation, the samples were transported, dismantled from the stacks within 1.0h, and the individual foils separated for further measurement. Subsequently, the γ -spectrometric measurements for all irradiated samples were performed by means of HPGe detectors calibrated with standard gamma sources with certified accuracies of their activities <2.5%. The repeated measurements during the reasonable time span allowed us to get minimum spectra of four for each nuclide. To avoid problems of high dead times and pile-up effects, the distances between the sample and detector were varied to keep the dead time below 10%. The large distances were used only for the short-time measurements of nuclides those who have high activities after the irradiation. Background measurements were taken two times for about 2 hours each before, after the measurements, and its contribution was subtracted for the same counting time for the corresponding nuclides. After then these spectra were stored for further analysis.

ANALYSIS OF SPECTRA

Spectra were analyzed using the computer code APTEC for the identification of product nuclides and the corresponding activities [18]. The identification process starts with the spectra having the longest decay times. Considering the correction for the γ -rays in the targets first such lines are selected which can be unambiguously identified. Then, successively going back in time, the identified nuclides are, searched in the proceeding spectra by calculating activities and looking whether these activities should be detectable in the spectrum under

consideration. Applying this algorithm decreases the number of possible nuclides in a spectrum. In the step of identification of nuclides, the half-lives, energies and branching ratios of γ -rays were taken from the Table of Isotopes [13]. By knowing the proton flux and measuring the activity for the respective product nuclide, the cross-sections for specific proton energies at different target positions were determined using the Microsoft Excel program according to Eq. (2).

Table (1). Nuclear Data of the Produced Terbium Radionuclides

| Nuclide | Half-Life | Decay Mode (%) | E_γ (MeV) | I_γ (%) | Contributing Reaction | Threshold (MeV) |
|--------------------|-----------|----------------|--|-------------------------------------|--|---------------------------------|
| ^{152}Tb | 17.5h | EC(100) | 344.4 596.3 | 65.0 9.4 | $^{152}\text{Gd}(p,n)$ | 4.80404 |
| ^{154m}Tb | 9.4h | EC(78.2) | 247.9 540.1 649.5 996.3 1004.7 | 22.0 20.0 11.0 9.0 11.0 | $^{154}\text{Gd}(p,n)$ $^{155}\text{Gd}(p,2n)$ $^{156}\text{Gd}(p,3n)$ | 4.37283 10.84970 19.44085 |
| ^{154g}Tb | 21.5m | EC(100) | 722.1 996.3 1274.4 | 8.0 5.0 11.0 | $^{154}\text{Gd}(p,n)$ $^{155}\text{Gd}(p,2n)$ $^{156}\text{Gd}(p,3n)$ ^{154m}Tb decay | 4.37283 10.84970 19.44085 |

- Table Data were taken from [13].
- Reaction Q-values were taken from [19].

Table (1) provides the details about irradiation on two stacks of foils. The γ -ray activities of produced radionuclides, the half-lives, and reaction Q-values. The experimentally measured cross-sections for the reactions $^{nat}\text{Gd}(p,x)^{152m+g,154m,154g}\text{Tb}$ at different incident proton energies are tabulated in Tables 3-5, respectively. In these tables the first column lists the incident energy on the foil while the second column lists the corresponding measured cross-section values. The experimental technique employed in the present work and the data evaluation are described in more detail in many of publications [15,16]. The excitation functions of the used four monitor reactions were taken from [14], the recommended values over the investigation energy region.

NUCLEAR MODEL CALCULATIONS

Nuclear model calculations were performed with ALICE-91 code (Blann, 1991) [9], using the framework of the geometry dependent hybrid model Blann et al., [4-10] for the pre-equilibrium emission of neutrons and protons in combination with the compound nucleus (CN) calculations are performed using the Weisskopf-Ewing formalism [20]. While the pre-equilibrium emission component is simulated employing the geometry dependent hybrid (GDH) model (Blann, 1972) [4] for the subsequent equilibrium emission of neutrons and protons, deuterons and alpha particles. The calculations are based almost on default options in the code (Blann, 1988) [7]. The initial exciton configuration given by Blann and Vonach, (1983) [8] were used for the pre-equilibrium calculations. The initial exciton number, the initial excited neutron number and the initial excited proton number were assumed to be 3.0, 0.8 and 1.2, respectively. The exciton includes the particle and the hole in the nucleus. The initial exciton number (3) is the sum of particles and holes. The initial excited proton number (1.2) corresponds to the initial proton particle number, which includes a portion of the projectile proton. The number $3 - 0.8 - 1.2 = 1$ is the initial hole number. The number $0.8 +$

1.2 = 2 is the initial particle number. The optical model potential based on the nucleon mean free paths was used to estimate the intranuclear transition rates. Particle binding energies were internally calculated using the Myers and Swiatecki mass formula [21].

Production cross-sections for the discussed reactions were calculated using the EMPIRE II code, v.2.19 (2005) [11]. The only modification to EMPIRE's default parameter library was the extension of calculations over the level scheme of $^{nat}\text{Gd}(p,x)^{152m+g}\text{Tb}$ up to the first excitation, in order to better separate the excitation functions of $^{nat}\text{Gd}(p,x)^{154m,154g}\text{Tb}$ (EMPIRE default considers only the ground state of this nucleus). Simple spherical optical model potentials (default in EMPIRE) with suppression of direct coupling to discrete levels were found to give the best agreement with the data, compared to global spherical potentials and Re-localized coupled-channel models with direct reaction channels open. EMPIRE performs preequilibrium calculations via a combined multi-step direct (MSD) and multi-step compound (MSC) approach; MSD calculations were executed using the TRISTAN and ORION modules by M. Chadwick et al, (1994) in this version of EMPIRE-II [22], while MSC calculations were done using NVWY theory (Nishioka et al., 1986) [23,24]. Finally, decay via particle emission of the compound nucleus, continuum states to those of the residual, as well as the g cascade through the daughter's discrete excitations, is governed by a statistical Hauser–Feshbach model [25].

DISCRETE LEVEL CROSS SECTION

EMPIRE-II is a valuable tool in the spin-parity assignment for discrete levels of residual nuclei populated in nuclear reaction evolving through compound nucleus mechanism. So there are several applications illustrating the method and the code's predictive power.

A major problem in the experimental nuclear spectroscopy is the spin-parity (J^π) assignment for the nuclear levels. In direct reactions with particular probes, the transferred angular momentum can be deduced from the shape of the angular distributions of the outgoing particles. In gamma-spectroscopy, measurements of gamma-ray angular distributions and conversion electron coefficient are the main tools in establishing a certain range for (J^π) values of the nuclear levels.

Another way leading to possible (J^π) values for nuclear levels is to exploit the dependence of the compound nucleus decay probabilities on the spin-parity of the discrete levels of the residual nuclei. These decay probabilities enter the level population cross section calculated in the frame work of compound nucleus statistical theory of nuclear reactions. By comparing the measured discrete level population cross sections with calculated, and assuming different hypothesis for the (J^π) of final state a certain restricted range in the (J^π) values can be obtained. Several studies of (p,n γ), (α ,n γ), (n,n' γ) reactions applied this procedure in selecting certain (J^π) values for levels measured by γ -spectroscopical means [26-31]. In completing this task, a compound nucleus statistical model code has to be used.

There are many model codes, but they usually require an extensive input including particle transmission coefficients provide by direct interaction codes. EMPIRE-II [11], is a nuclear reaction code which has not this inconvenience because, it directly accesses the most recent References Input Parameter Library RIPL-2 [32], and contains the well-known direct interaction code system, used mainly in nuclear reaction data evaluation.

The goal of this work is to use EMPIRE-II in conjunction with RIPL-2 as a valuable spectroscopic tool for spin-parity assignment to discrete nuclear levels. On the basis of population of the computational instruments, the application of spin-parity assignment is served us in determining the discrete level cross for the predicted nuclear level values. This is one application from several applications that reveal the predictive power of EMPIRE-II.

EMPIRE-II PREDICTION OF NUCLEAR LEVEL J^π VALUES

The prediction method presented in this work is based on the dependence of the population cross-sections on the spin and parity of the discrete levels of the residual nuclei. The sensitivity of the population cross-section to the variation of the final level spin-parity values depends on the difference between the transmission coefficients entering the Hauser-Feshbach formula when different J^π values are considered. It is difficult to make universal assumptions about this sensitivity and the prediction power of this method due to their dependence on several factors such as: reaction type, energy range, odd-even character of the target, spin-parity of the target's ground state, optical potential parameters, etc.. Therefore, detailed numerical calculations of the population cross-sections have to be performed case-by-case. EMPIRE-11 is an appropriate code system for performing this task especially because it is based on the latest versions of the reaction models, takes into consideration all the open reaction channels, offers many choices for the input parameters and automatically creates the input files. Due to the short execution time of EMPIRE-11, different input parameter values and a large set of J^π hypothesis can be tested until the best agreement with the experimental data is obtained. One complete calculation for the reactions presented below on a Pentium IV machine takes only few seconds. The predictive power of EMPIRE-11 was analyzed for the case of the reaction $(p,n\gamma)$ important for discrete γ -ray spectroscopy. Induced on Gd producing radioisotopes ^{154m}Tb , ^{154g}Tb .

RESULTS AND DISCUSSION

The nuclide production cross-sections on thin target element Gd were measured by off-line γ -spectroscopy for three identified nuclides in the proton energy range of 7.0 – 18.0 MeV. The measured cross-sections for the element of Gadolinium contain 3 individual cross-sections for 2 identified nuclides. All the identified nuclides with their half-lives, decay properties are presented in Table (1). The cross-sections data for the reactions in either tabular form or graphics will contain the excitation functions measured for $^{\text{nat}}\text{Gd}(p,x)^{152m+g,154m,154g}\text{Tb}$ are given in this paper. The currently measured excitation functions are presented in Figures from 1 to 4. In the measured data, all the main sources of uncertainties and their resulting errors are presented in Table (2). The measured cross sections and their estimated errors for the considered reactions are given in Table (3). From the preliminary survey on the magnitude of production cross-sections it is seen that the large cross-sections are preferred for short-lived nuclides and on the contrary, small cross-sections are for long-lived ones. The excitation functions for both of them increase monotonically up to the highest proton energies investigated, though their absolute cross-section values are very small (in milli-barn).

Only by looking to the shape of the individual excitation functions of products, it is not enough to distinguish the mechanisms of reactions, such as direct, pre-equilibrium, and compound reactions leading to the residual nuclides. To solve this problem, the theoretical model calculation using the computer codes system based on the experimental data is suited for understanding the reaction mechanism. The shape of the present excitation functions are slightly deviated with the calculated one. The fact behind this reason was most probably due to the determination of cumulative cross-sections. The effect of varying various parameters of the codes on the calculated excitation functions is found of great importance. The high-energy tail portion of the excitation functions can be satisfactorily reproduced if the pre-equilibrium component is included in the calculations. From these figures, it is observed clearly that the measured data are lower than the theoretical predictions obtained from the use of ALICE-91 code, and it seems in agreement with EMPIRE-II. Further, all these codes give more or less

similar descriptions of the data in the peak region, which is of interest from the point of view of compound nucleus mechanism.

Table (2). Main Sources of Uncertainty with the Maximum Limit of Errors in the Present Measured Cross Section.

| Sources of Uncertainties | Error in (%) |
|--|----------------|
| Number of Target Nuclei in the Sample (Target mass) | Negligible |
| Flux Fluctuation During Irradiation Time | Negligible |
| Irradiation Time and Counting Time | Negligible |
| Half-Lives | 1.0 |
| Intensity of Gamma Rays | 1.0 |
| Self Absorption of Gamma-Rays in Target Foils | < 1.0 |
| Contamination Due to Recoil Losses | Not Considered |
| Absolute Efficiency Calibration | 2.5 |
| Quality of the Monitor Data (Proton Fluence) | 5.0 |
| Mass of Monitor Foil | < 1.0 |
| Self Absorption of Gamma-Rays in Monitor Foils | Negligible |
| Dead Time and Pile-up Losses in γ -Spectroscopy | Negligible |
| Statistical Uncertainty in Net Peak Areas | 1.0-5.0 |
| Geometry Dependent Detector Efficiency | < 1.0 |
| Secondary Particles contribution in Nuclides Production, and Beam Intensity Loss as the Beam Traverses the Stack Thickness | 7.0 |
| The Root Average of Total Error Due to all these Factors after Quadrature Sum is Estimated to Be | < 10 % |

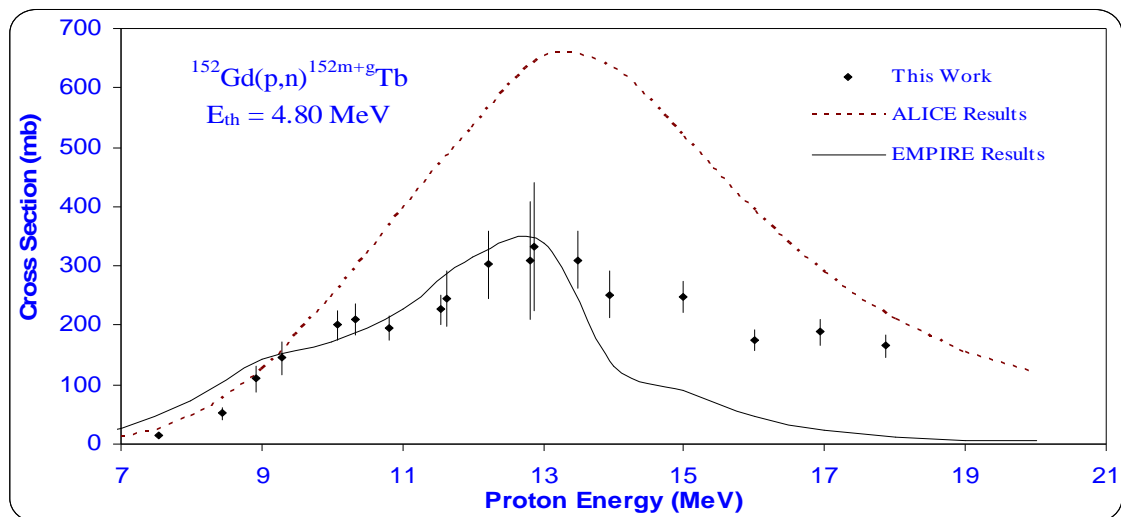


Fig. (1). Experimental Excitation Function for the Production of $^{152m+g}\text{Tb}$ Together with the Data Obtained by Theoretical Calculations.

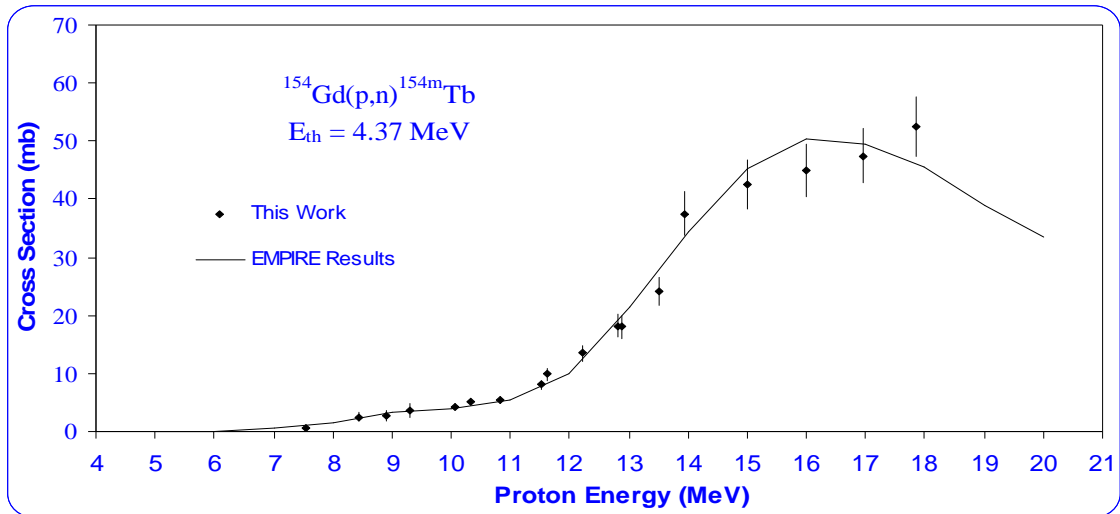


Fig. (2). Experimental Excitation Function for the Production of ^{154g}Tb Together with the Data Obtained by Theoretical Calculations.

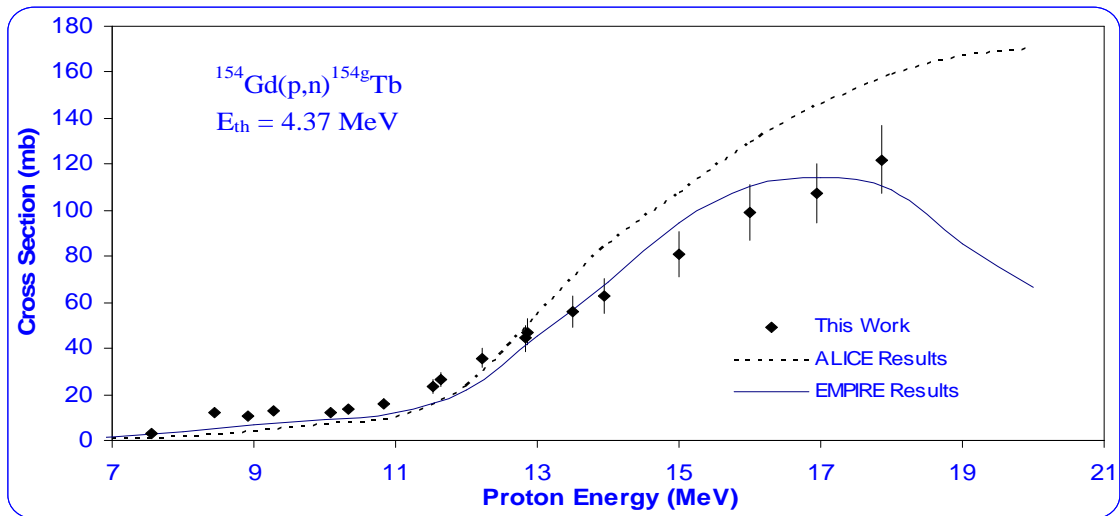


Fig. (3). Experimental Excitation Function for the Production of ^{154m}Tb Together with the Data Obtained by Theoretical Calculations.

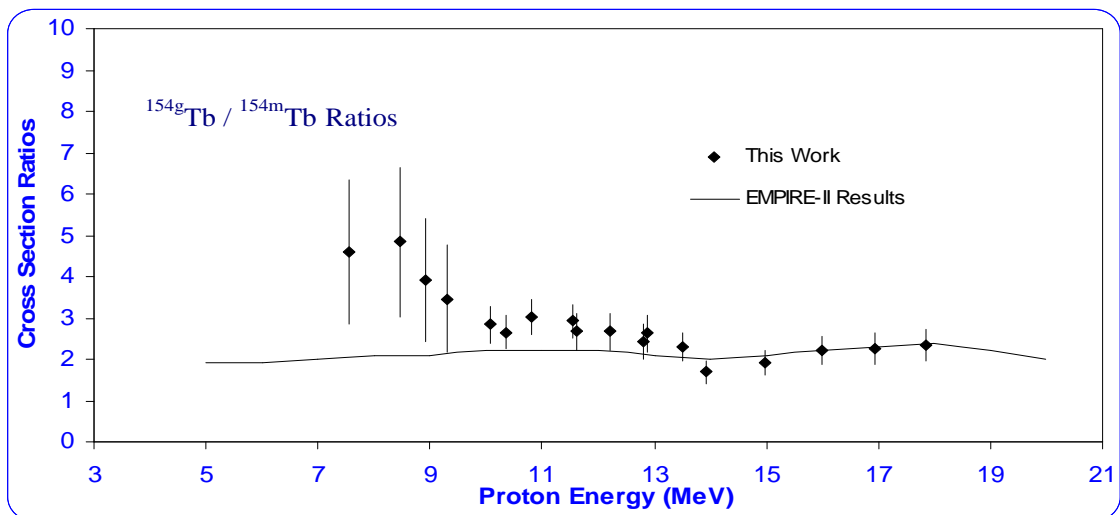


Fig. (4). Isomeric Ratios of the Experimental Excitation Functions for the Production of $^{154g}\text{Tb}(7^-)/^{154m}\text{Tb}(3^-)$ (Higher Spin State/Lower Spin State).

Table (3). Measured Cross Sections Values for Production of $^{152m+g}\text{Tb}$, ^{154g}Tb , ^{154m}Tc , and Isomeric Ratios on Natural Gd Targets at Different Energies.

| Incident Energy E_p (MeV) | $^{152}\text{Gd}(p,n)^{152m+g}\text{Tb}$ σ (mb) | $^{154}\text{Gd}(p,n)^{154g}\text{Tb}$ σ (mb) | $^{154}\text{Gd}(p,n)^{154m}\text{Tb}$ σ (mb) | Isomeric Ratios σ^g/σ^m |
|-----------------------------|---|---|---|--|
| 17.87±1.07 | 165.59±19.27 | 122.0±16.01 | 52.50±5.19 | 2.33 |
| 16.95±1.02 | 189.25±22.02 | 107.0±14.07 | 47.50±4.70 | 2.26 |
| 15.99±0.96 | 175.56±18.34 | 99.00±11.81 | 45.00±4.46 | 2.20 |
| 14.99±0.90 | 248.27±25.94 | 80.85±9.65 | 42.50±4.21 | 1.90 |
| 13.94±0.84 | 251.42±39.18 | 63.11±7.45 | 37.50±3.73 | 1.68 |
| 13.50±0.81 | 309.77±48.27 | 55.77±6.58 | 24.27±2.41 | 2.30 |
| 12.87±0.77 | 331.70±107.38 | 47.15±6.29 | 17.97±1.90 | 2.62 |
| 12.82±0.77 | 309.15±100.08 | 44.25±5.91 | 18.20±1.92 | 2.43 |
| 12.21±0.73 | 302.04±57.96 | 35.78±4.56 | 13.43±1.46 | 2.66 |
| 11.63±0.70 | 245.56±47.12 | 26.25±3.34 | 9.84±1.07 | 2.67 |
| 11.53±0.69 | 227.03±24.78 | 23.46±2.31 | 8.04±0.80 | 2.92 |
| 10.82±0.65 | 195.12±21.29 | 16.24±1.60 | 5.36±0.53 | 3.03 |
| 10.34±0.62 | 210.12±26.94 | 13.50±1.61 | 5.08±0.51 | 2.66 |
| 10.08±0.60 | 200.16±25.67 | 12.32±1.47 | 4.35±0.43 | 2.83 |
| 9.29±0.56 | 145.20±28.11 | 12.51±1.81 | 3.61±1.25 | 3.47 |
| 8.92±0.54 | 110.28±21.35 | 10.62±1.54 | 2.71±0.94 | 3.92 |
| 8.45±0.51 | 51.32±9.93 | 12.31±1.78 | 2.54±0.88 | 4.84 |
| 7.55±0.45 | 15.56±3.01 | 3.31±0.48 | 0.72±0.25 | 4.59 |

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