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(ԵՐԵՎԱՆԻ ՖԻԶԻԿԱԿԻ ԻՆՏԵՐՆԱԿԱՆ)

Մկրտչյան Հայկուհի Ավագի

ՅԻՎԼՈՏԻՈՆ C18/18-Ի ՎԸՆ ՊՐՈՏՈՆ ՀԱՐՈՆՑՎԱԾ ՆՈՒԿԼԻԴՆԵՐԻ ՍՏԱՅՄԱՆ
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մասնագիտությամբ ֆիզիկամաթեմատիկական գիտությունների թեկնածուի գիտական
աստիճանի հայցման ատենախոսություն

ՍԵՂՄՍԳԻՐ

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A.I. ALIKHANYAN NATIONAL SCIENCE LABORATORY

(YEREVAN PHYSICS INSTITUTE)

Haykuhi Avag Mkrtchyan

STUDY OF THE PRODUCTION OF PROTON-INDUCED NUCLIDES ON NATURAL

GADOLINIUM ON CYCLOTRON C18/18

SYNOPSIS

of Dissertation in 01.04.16 - "Nuclear, elementary particles and cosmic ray physics" for the degree
of Candidate in Physical and Mathematical Sciences

YEREVAN – 2026

Ատենախոսության թեման հաստատվել է Ա.Ի Ալիխանյանի անվան Ազգային գիտական Լաբորատորիայի (Եր.ՖԻ) գիտական խորհրդում:

Գիտական ղեկավար՝
Ֆիզմաթ. գիտ. թեկնածու

Քերոբյան Բվետաա Արամավազի (ԱԱԳԱ)

Պաշտոնական ընդմիջություն՝

Ֆիզմաթ. գիտ. դոկտոր

Բալաբեկյան Սևակիտ Ռաֆիկի (ԵՊՀ)

Ֆիզմաթ. գիտ. թեկնածու

Մարգարյան Աննա Թեատրոսի (ԱԱԳԱ)

Առաջատար կազմակերպություն՝

Երևանի պետական համալսարան (ԵՊՀ), Երևան, Հայաստան

Ատենախոսության պաշտպանությունը կայանալու է 2026 թ. օգոստոսի 25-ին ժամը 14:00-ին, ԱԱԳԱ-ում գործող ԲԿԳԳ-ի 024 «Ֆիզիկայի» մասնագիտական խորհրդում (Ալիխանյան եղբայրների փ. 2, Երևան, 0036):

Ատենախոսությանը կարելի է ծանոթանալ ԱԱԳԱ-ի գրադարանում:

Մեղմագիրն առաքված է 2026 թ. հունիսի 15-ին:

Մասնագիտական խորհրդի գիտական քարտուղար՝
Ֆիզմաթ. գիտ. դոկտոր

Հրաչյա Մարուկյան

The subject of the dissertation is approved by the scientific council of the A.I. Alikhanyan National Science Laboratory (YerPhI).

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The defense will take place on August 25 2026 at 14:00, during the "Physics" professional council's session of HESC 024 of acting within AANL (2 Alikhanyan Brothers str., Yerevan, 0036).

The dissertation is available at the AANL library.

The synopsis is delivered on June 15, 2026.

Scientific secretary of the professional council
Doctor of ph-math sciences

Hrachya Marukyan

Abstract

This work is devoted to the investigation of proton-induced production of terbium radionuclides using a natural gadolinium target irradiated with the C18/18 cyclotron beam. Terbium (Tb), a lanthanide, is unique among the elements in providing four medically important radionuclides for nuclear medicine applications, i.e. ^{149}Tb , ^{152}Tb , ^{155}Tb and ^{161}Tb applicable for therapeutic and/or diagnostic (theranostic) purposes. ^{155}Tb ($T_{1/2} = 5.32$ d) and ^{152}Tb ($T_{1/2} = 17.5$ h) decay via positron emission or electron capture and hence mostly by emitting γ -rays detectable by gamma cameras: SPECT or PET scanners. While ^{161}Tb ($T_{1/2} = 6.9$ d) decays by emitting β -particles that damage the surrounding cells over a few millimeters, ^{149}Tb ($T_{1/2} = 4.1$ h) produces even shorter-range alpha α -particles that allow much more localized destruction of the cancerous cells. The ^{160}Tb ($T_{1/2} = 72.3$ d) isotope of great practical importance is used in nuclear examination (forensics). The ^{161}Tb emits beta particles, which are effective for treating larger tumors. Additionally, it also emits low-energy conversion and Auger electrons, which are particularly effective at targeting smaller tumors, micro-metastases, and even individual cancer cells. This makes it potentially more effective in treating cancers at earlier stages and reducing the risk of recurrence. Proton-induced reactions on natural gadolinium for producing the radioisotopes ^{152}Tb , ^{155}Tb , ^{156}Tb , ^{160}Tb were investigated to determine the excitation functions across energy ranges from the respective reaction thresholds up to 18 MeV. The experiment was carried out by an external proton beam of Cyclotron C18/18, located at AANL, using activation technology and subsequent spectrometric analysis. The experiment was carried out using stacked-activation technique. The proton beam irradiated targets were subsequently analyzed by gamma-ray spectrometry using HPGe detector CANBERRA coupled to a multichannel digital analyzer. The experimentally obtained data were compared with earlier published data and the model calculations using codes TALYS 1.96, EMPIRE 3.2, and ALICE library. For isomeric triplets $^{154\text{g}}, ^{154\text{m}}, ^{154\text{m}2}\text{Tb}$ and $^{156\text{g}}, ^{156\text{m}}, ^{156\text{m}2}\text{Tb}$ the isomeric cross-sections ratio was measured.

The aims of this work are summarized below:

- Measurement of excitation functions for the production of residual nuclides ^{135}Tb , ^{134}Tb , and ^{156}Tb via proton-induced reactions on natural gadolinium;
- Measurement of cross-sections and integral activities for the formation of the medically relevant radionuclide ^{153}Tb , which has potential use in targeted radiotherapy;
- Measurement of cross-sections for the formation of ^{160}Tb , a nuclide of interest in nuclear forensics and safeguards;
- Measurement of cross-sections for total and ground states of ^{156}Tb , ^{156g}Tb and ^{156m}Tb ;
- Measurement individually of cross-sections for the metastable and ground states of ^{154}Tb , $^{154m1}\text{Tb}$, $^{154m2}\text{Tb}$, ^{154g}Tb , and ^{154o}Tb .

The importance and novelty of work

- At the first time in the same experiment were measured cross-sections of formation ^{156g}Tb and ^{156m}Tb ;
- At the first time was measured thin-target activities of ^{135}Tb ;
- At the first time was measured ICR for triplet 134m1 , 134m2 , ^{134g}Tb ;
- At the first time was measured ICR for triplet 156m1 , 156m2 , ^{156g}Tb .

The significance of the obtained results:

- Contribute to the extension of the nuclear reaction database for rare earth elements;
- Support the development and validation of nuclear reaction simulation codes;
- Open new possibilities for medical radionuclide production using compact cyclotrons;
- Offer reference data for nuclear forensics and safety-related applications;
- Provide experimental benchmarks for refining theoretical models in nuclear science.

Structure of the dissertation

The dissertation comprises 100 pages, including figures and tables. The list of references contains 47 titles. The dissertation is composed of: List of papers; published based on dissertation data; List of figures; List of tables; Abbreviations; Abstract; The importance and novelty of work; Introduction; Five chapters; Conclusion; References.

Content of the dissertation

The "Introduction" is present the detailed description of the measurement of excitation functions for proton-induced reactions on natural gadolinium and the determination of the activity of medically relevant radionuclides produced. A particular feature of this study is to establish the suitability of a commercial cyclotron C18/18 with energy 18 MeV. The expansion of the experimental database contributes to the refinement of existing theoretical models and supports the development of new ones. The results of the measurements have been compared with theoretical calculations using computer codes TALYS 1.96, EMPIRE 3.2 and MENDL-2P data library. Program packages GEANT4 and SRIM/TRIM, have been used in the initial stages of experimental planning.

The CHAPTER 1 "Theoretical programs" provides an overview of the theoretical calculations performed as part of this study. Sections 1.1 to 1.5 detail the use of the nuclear reaction codes TALYS 1.96, EMPIRE 3.2, and ALICE/IPPE (MENDL-2P) for the theoretical evaluation of reaction cross-sections based on various nuclear models. The GEANT4 and SRIM/TRIM simulation packages were employed to model proton transport through the target stack, ensuring accurate representation of energy loss and particle interactions.

The CHAPTER 2 "Theoretical calculations" presents a comparative analysis of the theoretical excitation functions for proton-induced reactions on natural gadolinium calculated using the inherent models of the TALYS 1.96 and EMPIRE 3.2 codes, as well as data from the MENDL-2P library. The calculations cover the proton energy range from the respective reaction thresholds up to 70 MeV and are

compared with available experimental data. The predictive capabilities of each model are evaluated and discussed in detail

The CHAPTER 3 “Experimental equipment” provides a detailed description of the experimental setup at the AANL (Yerevan Physics Institute). The natural gadolinium target was irradiated using the extracted proton beam with 18 MeV energy of the C18/18 cyclotron. The technical specifications of the cyclotron and the Nirta target module are presented. The appearance of the target holder and stack that was made at AANL is described. The experiment was carried out using the activation analysis method, and the induced activity was measured through high-resolution γ -ray spectrometry employing a high-purity germanium HPGe detector CANBERRA coupled with a multichannel analyzer. The procedures for detector efficiency and energy calibration are also described in detail.

The CHAPTER 4 “Experiment” discusses the fundamental principles of activation analysis used to investigate the excitation functions of nuclides produced through the $^{nat}\text{Gd}(p,xn)$ reaction. Natural gadolinium has seven stable isotopes with abundances of: ^{152}Gd - 0.20%; ^{154}Gd - 2.18%; ^{155}Gd - 14.80%; ^{156}Gd - 20.47%; ^{157}Gd - 15.65%; ^{158}Gd - 24.84%; ^{160}Gd - 21.86%. The spectrometric data for the radionuclides under investigation are presented. The stacked-foils activation technique is an experimental method widely used in nuclear physics to measure nuclear reaction cross-sections as a function of incident particle energy. A key advantage of the stacked-foil activation method is that it enables the determination of the complete excitation function curve from a single proton beam irradiation. The described stacked-foil activation technique was employed to measure cross-sections of proton-induced reactions on natural gadolinium $^{nat}\text{Gd}(p,x)$ over an energy range extending from the threshold energy of contributing reactions up to 18 MeV. In this method, a sequence of thin-target foils is arranged to form a stack, as illustrated in

Fig. 1.

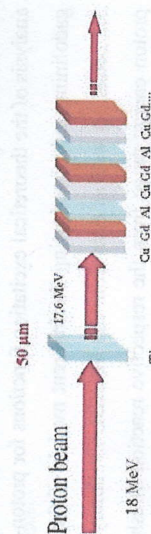


Fig.1. General view of the stack target.

Table 1. Spectrometric properties of produced nuclides Tb

Nuclides	Half-Life	Decay mode	Spin	E_γ , keV	Intensity, %
^{153}Tb	2.34 d	ε (100%)	5/2+	102.25 109.75 212	6.4 6.8 31
^{154g}Tb	21.5 h	ε (100%)	0	123.07 722.12 873.19	26 7.7 5.3
$^{154m1}\text{Tb}$	9.994 h	ε (78.2%) IT (21.8)	3-	123.07 247.92 649	30 21.1 10.9
$^{154m2}\text{Tb}$	22.7 h	ε (98.2%) IT (1.8%)	7-	247.94 346.70 1419.81	78.81 69.13 46.15
^{155}Tb	5.32 d	ε (100%)	3/2+	86.55 105.31 180.08	32 25.1 7.5
^{156g}Tb	5.35 d	ε (100%)	3-	88.97 199.19 534.29	18 41 67
$^{156m1}\text{Tb}$	24.4 h	IT (100%)	7-	49.63	74.1
$^{156m2}\text{Tb}$	5.3 h	IT <(100%) ε > (0.%)	0+	88.4	1.15
^{160}Tb	72.3 d	β^- (100%)	3-	86.78 298.57 879.37 966.16	13.2 26.1 30.1 25.1
^{65}Zn	243.93 d	$\varepsilon^+\beta^+$ (100%)	5/2-	1115.54	50.12

The spectrometric data of the under study nuclides, such as the half-life ($T_{1/2}$), the γ -ray energy (E_γ), and the γ -ray emission intensity (I_γ), are taken from NuDat 3 and collected in Table 1. Prior to the experiment, the irradiation setup was simulated using the Monte-Carlo method. The aim of the simulation was to determine the average proton energies in each natural gadolinium (^{nat}Gd) and copper (Cu) foil within the stacked-foil assembly, as well as the corresponding energy degradation and energy straggling. Each natural gadolinium foil was followed by a natural aluminum (^{nat}Al) foil, which served as a proton energy degrader. A Cu foil was placed in front of each Gd foil to monitor the beam intensity. The energy loss of protons in the

stacked foils was calculated using the GEANT4 and SRIM/TRIM packages in Table 2. The simulation took into account the presence of Ti technological window foil with 50 μm thickness.

Table 2. Protons energy distribution in Cu and Gd each foils, simulated by TRIM and GEANT4 packages.

Cu foil		Proton energy, MeV		Gd foil	
TRIM	GEANT4	TRIM	GEANT4	TRIM	GEANT4
4.56 $\pm$ 0.85	4.84 $\pm$ 1.30	3.77 $\pm$ 0.80	4.16 $\pm$ 1.41		
6.66 $\pm$ 0.74	6.84 $\pm$ 1.03	6.05 $\pm$ 0.69	6.31 $\pm$ 1.11		
8.33 $\pm$ 0.72	8.47 $\pm$ 0.83	7.83 $\pm$ 0.63	8.02 $\pm$ 0.89		
9.79 $\pm$ 0.67	9.88 $\pm$ 0.67	9.35 $\pm$ 0.57	9.48 $\pm$ 0.72		
11.10 $\pm$ 0.69	11.17 $\pm$ 0.54	10.69 $\pm$ 0.57	10.80 $\pm$ 0.58		
12.31 $\pm$ 0.59	12.34 $\pm$ 0.43	11.94 $\pm$ 0.44	12.00 $\pm$ 0.47		
13.44 $\pm$ 0.50	13.44 $\pm$ 0.34	13.07 $\pm$ 0.53	13.11 $\pm$ 0.37		
14.48 $\pm$ 0.68	14.47 $\pm$ 0.27	14.16 $\pm$ 0.41	14.16 $\pm$ 0.29		
15.48 $\pm$ 0.64	15.45 $\pm$ 0.22	15.16 $\pm$ 0.50	15.16 $\pm$ 0.24		
16.44 $\pm$ 0.52	16.37 $\pm$ 0.19	16.14 $\pm$ 0.39	16.10 $\pm$ 0.20		
17.37 $\pm$ 0.28	17.26 $\pm$ 0.14	17.06 $\pm$ 0.38	16.99 $\pm$ 0.17		

Consequently, proton energy values were calculated for each individual foil within the stack. Additionally, this method allows for a more precise determination of the protons amount in each foil. The graphical interface of the created program for the first and last Cu foils is shown in Fig. 2.

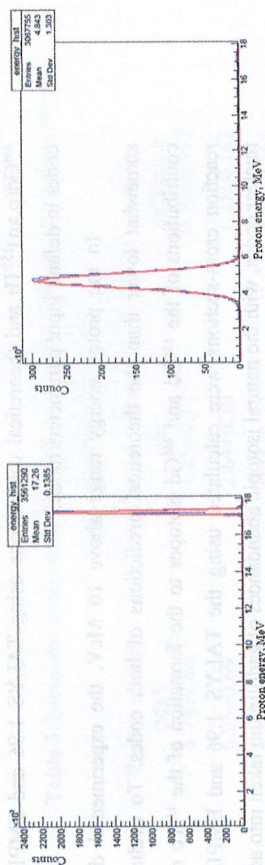


Fig 2. Graphical interface, proton energy distribution in the first and last Cu foils.

The collected target was irradiated with an external proton beam from the cyclotron with energy 18 MeV at a beam current of 1 μA for 10 minutes. The emitted γ -rays were measured by the high-resolution HPGe detector CANBERRA connected to the analyzer for spectral processing by program GENIE 2000.

The CHAPTER 5 "Results" discusses the measured excitation function values for the $^{155}\text{Gd}(p,xn)$ reactions. The analysis of theoretical calculations performed using the TALYS 1.96 and EMPIRE 3.2 codes up to proton energies of 70 MeV revealed that, within the proton energy range up to 20 MeV, the results from both codes show good agreement across different models. Therefore, in the present study, calculations performed using default parameters of TALYS 1.96 and EMPIRE 3.2 codes were used for comparison with the experimental data. The overall uncertainties are coming from both statistic and systematic errors. The statistical errors are attributed to counting statistics and are estimated to be around 3-7%. The systematic errors are due to uncertainties in the irradiation time - about 1%, the detection efficiency error - about 3%, the uncertainties in proton beam intensity - about 10%. The overall uncertainties of the cross-section measurements were in the range of 11-13%.

Production of ^{155}Tb

Nuclide ^{155}Tb (half-life $T_{1/2} = 5.3$ d) is a long-lived analog of technetium $^{99\text{m}}\text{Tc}$ (half-life $T_{1/2} = 6.0067$ h), due to which it is applicable in SPECT. In Fig. 3, the measured excitation function is given together with the data [1, 2] for the reaction

$^{155}\text{Gd}(p,xn)^{155}\text{Tb}$ and theoretical calculations using TALYS 1.96 and EMPIRE 3.2 codes in default input parameters mode.

In the proton energy range above 10 MeV, the experimental data are somewhat lower than the theoretical predictions of both codes. To evaluate the contributions of the ^{155}Gd and ^{156}Gd isotopes to the formation of the ^{155}Tb nuclide, reaction cross-sections were calculated using the TALYS 1.96 and EMPIRE 3.2 nuclear codes, with the natural isotopic abundances of gadolinium taken into account.

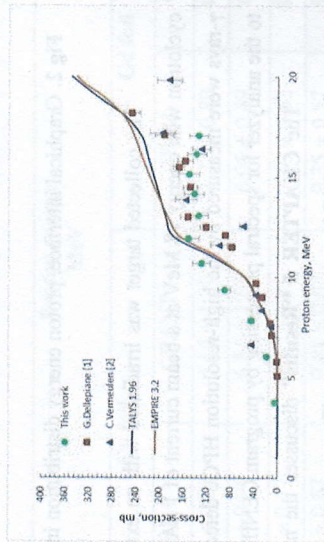


Fig. 3. Measured excitation function for $^{nat}\text{Gd}(p,xn)^{155}\text{Tb}$ reaction along with data from [1, 2] and calculated by TALYS 1.96 and EMPIRE 3.2 codes.

The calculation results show that in the proton energy range from the threshold to 10.2 MeV, the formation of ^{155}Tb occurs through the (p,n) channel. In the energy range exceeding 10.2 MeV, the predominant source of the ^{155}Tb isotope is the $^{156}\text{Gd}(p,2n)$ reaction, clearly depicted in Fig. 2. Table 3 presents the measured cross-sections and calculated thin-target activities as a function of 11 discrete proton energy values.

Table 3. Measured cross-sections and thin-target activities for formation ^{155}Tb in reaction $^{nat}\text{Gd}(p,xn)$.

Proton energy, MeV	Cross-section, mb	Activity, $\mu\text{Ci}/\mu\text{A h}$
3.77 ± 0.80	1.05 ± 0.12	0.50 ± 0.057
6.05 ± 0.69	2.64 ± 0.30	1.25 ± 0.142
7.83 ± 0.63	6.75 ± 0.77	3.21 ± 0.366
9.35 ± 0.57	69.57 ± 7.86	33.1 ± 3.74
10.69 ± 0.57	112.23 ± 12.63	53.3 ± 6.0
11.94 ± 0.44	141.16 ± 15.81	67.1 ± 7.52
13.07 ± 0.53	136.15 ± 15.18	64.7 ± 7.21
14.16 ± 0.41	145.23 ± 16.12	69.0 ± 7.66
15.16 ± 0.50	151.34 ± 16.72	71.4 ± 7.89
16.14 ± 0.39	139.23 ± 15.32	66.1 ± 7.27
17.06 ± 0.38	132.71 ± 14.53	63.0 ± 6.89

The simplest and most effective ways to obtain the ^{155}Tb nuclide for medical purposes is to use the reaction $^{155}\text{Gd}(p,n)$. The integral activity of the ^{155}Tb formation on enriched gadolinium ^{155}Gd in the proton energy range up to 10.8 MeV, is presented in Fig. 4, together with data from [1, 2] and the results of theoretical calculations using the TALYS 1.96 code. The experimental results are in good agreement with the theoretical predictions.

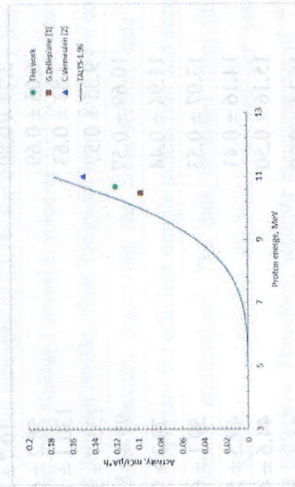


Fig. 4. Measured integral activity formation ^{155}Tb on enriched target ^{155}Gd along with data from [1, 2] and calculated by TALYS 1.96 code.

When using an enriched ^{155}Gd target at proton energies above 10.8 MeV, the reaction $^{155}\text{Gd}(p,2n)$ occurs, resulting in the formation of ^{154}Tb . Thus, the use of an enriched ^{155}Gd target and protons with energies below 10.8 MeV potentially provides significant yields of ^{155}Tb with high radionuclide purity. With such a limitation of proton energy for the integral activity, a value of 0.12 mCi/($\mu\text{A h}$) was obtained, which is consistent with the calculated estimation of 0.15 mCi/($\mu\text{A h}$) for proton energy from threshold up to 11 MeV presented in Ref. [2] and 0.097 mCi/($\mu\text{A h}$) for proton energy range up to 10.5 MeV given in Ref. [1].

Production of ^{160}Tb

The function of ^{160}Tb is critical in post-detonation analysis, particularly in nuclear forensics. In nuclear forensics, post-detonation debris analysis is an important procedure for determining an exploded nuclear weapon's origin, features, and history. The $^{160}\text{Gd}(p,n)^{160}\text{Tb}$ is an important reaction in the field of technical nuclear forensics used in post-detonation debris analysis to understand the characteristics of a detonated nuclear weapon. The ^{160}Gd has seven stable isotopes; however, the ^{160}Tb in proton-induced reactions formed only on ^{160}Gd . The experimental data obtained for a natural target can be extended to an enriched target, taking into account the abundance of ^{160}Gd in a natural target (21.86%). The experimental measured cross-sections for reaction $^{160}\text{Gd}(p,n)^{160}\text{Tb}$ [3] are presented in Table 4.

Table 4. Measured cross-sections for $^{160}\text{Gd}(p,n)^{160}\text{Tb}$ reaction.

Proton energy, MeV	Cross-section, mb
3.77 ± 0.80	10.7 ± 1.28
6.05 ± 0.69	58.2 ± 6.98
7.83 ± 0.63	130.1 ± 15.61
9.35 ± 0.57	135.7 ± 15.94
10.69 ± 0.57	99.8 ± 11.48
11.94 ± 0.44	78.3 ± 9.00
13.07 ± 0.53	59.1 ± 6.50
14.16 ± 0.41	49.5 ± 5.20
15.16 ± 0.50	44.6 ± 4.68
16.14 ± 0.39	36.3 ± 3.74
17.06 ± 0.38	30.8 ± 3.17

Figure 5 shows the comparison of the measured excitation function for $^{160}\text{Gd}(p,n)^{160}\text{Tb}$ reaction and experimental data from [2, 4, 5] along with theoretical calculated values using TALYS 1.96, EMPIRE 3.2 codes and MENDL-2P library.

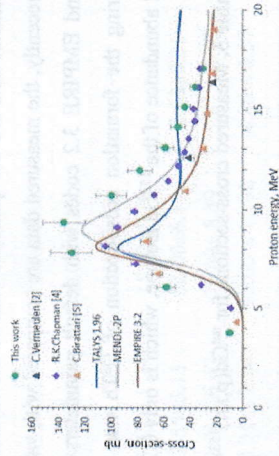


Fig 5. Measured excitation function for $^{160}\text{Gd}(p,n)^{160}\text{Tb}$ reaction along with data from [2, 4, 5] and calculated by TALYS 1.96, EMPIRE 3.2 codes as well MENDL-2P library.

In the investigated energy range of the protons in Ref. [2] there are only two values, and they agree with results measured in frame of this study. Our measurements are in agreement with the theoretical calculations using TALYS 1.96, EMPIRE 3.2 codes, and MENDL-2P library within the error range. However, the measured data are higher and has wider range than the data from [2, 4, 5]. The reason for the difference between our experiment and data of [5] could be because they employed a partial intensity of 32.1% for the 879 keV γ -line in their work. However more recent data NuDat 3 reveal a value of 30.1%. The data from Ref. [4] may have lower data than our results, since the gadolinium foils were encapsulated with Kapton tape in their research. Improving the cross-section data for the $^{160}\text{Gd}(p,n)^{160}\text{Tb}$ reaction is important for efficient sample production and supports the use of ^{160}Tb in nuclear forensic applications. The data obtained from these measurements will contribute to the experimental database required for precise analysis of post-detonation materials. Therefore, accurate measurement of ^{160}Tb is indispensable for ensuring precise analysis.

The numerical data of excitation functions for formation ^{153}Gd , ^{156}Gd and $^{156\text{m}}\text{Gd}$ are listed Table 5 and Table 6, respectively. The total errors of the experimental data are the quadratic sum of the statistical and systematic errors. As noted recently, the measured data are compared with values calculated using TALYS 1.96 and EMPIRE 3.2 codes with default parameters as well MENDL-2P library. Measuring the formation cross-section of ^{152}Tb was not feasible due to the low natural abundance of the ^{152}Gd isotope, which is only 0.2%.

Table 5. Measured cross-sections for $^{153}\text{Gd}(p,xn)^{153}\text{Gd}$, ^{156}Gd reactions.

Proton energy, MeV	Cross-sections, mb		
	^{153}Tb	^{156}Tb	$^{156\text{m}}\text{Tb}$
3.77±0.80		2.78±0.32	4.20±0.49
6.05±0.69	-	15.24±1.79	15.40±1.81
7.83±0.63	-	40.12±4.71	46.70±5.49
9.35±0.57	-	89.89±10.56	90.71±10.66
10.69±0.57	-	129.31±15.19	157.50±18.51
11.94±0.44	4.00±0.47	155.13±18.22	171.22±20.12
13.07±0.53	9.60 ± 1.12	125.71±14.77	168.69±19.82
14.16±0.41	12.60±1.48	122.69±14.41	183.64±21.58
15.16±0.50	15.60±1.83	123.64±14.52	188.31±22.13
16.14±0.39	14.80±1.73	111.07±13.05	163.36±19.19
17.06±0.38	14.70±1.72	103.60±12.17	147.28±17.31

Table 6. Measured cross-sections for $^{154}\text{Gd}(p,xn)^{154}\text{Gd}$, $^{154\text{m}}\text{Gd}$, $^{154\text{m}2}\text{Gd}$ reactions

Proton energy, MeV	Cross-section, mb		
	^{154}Tb	$^{154\text{m}}\text{Tb}$	$^{154\text{m}2}\text{Tb}$
3.77±0.80	-	-	-
6.05±0.69	1.52±0.17	1.00 ± 0.11	-
7.83±0.63	3.76±0.44	3.00±0.35	-
9.35±0.57	8.94 ± 1.05	7.00±0.82	-
10.69±0.57	15.25 ± 1.79	15.00±1.76	-
11.94±0.44	43.55±5.11	52.00±6.11	2.68±0.31
13.07±0.53	54.71 ± 6.42	72.00± 8.39	3.67 ± 0.43
14.16±0.41	63.91 ± 7.50	93.00 ± 10.92	4.86 ± 0.57
15.16±0.50	72.35 ± 8.50	110.00 ± 12.92	5.89±0.69
16.14±0.39	65.60 ± 7.70	102.00± 11.98	5.85±0.68
17.06±0.38	62.97 ± 7.39	104.00± 12.22	5.59±0.65
			161.77±13.53
			188.24±15.74
			173.45±14.50
			172.56±14.51

The comparison of experimental results for the $^{154}\text{Gd}(p,xn)^{154}\text{Gd}$, $^{154\text{m}}\text{Gd}$, $^{154\text{m}2}\text{Gd}$ reactions with calculated values using the TALYS 1.96 and EMPIRE 3.2 codes indicates the need for further investigations over a wider range of proton energies to resolve the observed discrepancies between measured and predicted cross-sections. Such studies are essential not only for validating existing nuclear reaction models but also for improving their predictive accuracy and guiding the development of more advanced theoretical approaches.

Isomeric Cross-Section Ratio

Important insights into the mechanism of nuclear reactions can be gained through analyzing the ICR in the creation of isomeric pairs in nuclear reactions. It explains how the nuclear reaction develops and provides crucial information regarding the transfer of energy and angular momentum throughout a reaction.

The ICRs for the isomeric triplets $^{154\text{m}}\text{Gd}$, $^{154\text{m}2}\text{Gd}$ and $^{156\text{m}}\text{Gd}$ were investigated. Since both ^{154}Tb and ^{156}Tb exist as triplets comprising one ground and two metastable states - the ICR is defined as $\text{ICR} = \sigma_g/\sigma_{\text{total}}$ to avoid ambiguity in the

calculations. Theoretical ICRs in the proton energy range from the reaction thresholds up to 18 MeV were calculated using the TALYS 1.96 code. Comparisons of the measured ICRs for $^{154}\text{m}_1;^{154}\text{m}_2;^{154}\text{gTb}$ and $^{156}\text{m}_1;^{156}\text{m}_2;^{156}\text{gTb}$ nuclides with the calculated values using TALYS 1.96 code are presented in Table 7 and Figs. 6-7. The errors in the ICRs values listed in the Table 5.6 are determined using the errors in the cross-section measurements.

Table 7. Measured ICRs for $^{154}\text{m}_1;^{154}\text{m}_2;^{154}\text{gTb}$ and $^{156}\text{m}_1;^{156}\text{m}_2;^{156}\text{gTb}$ triplets with calculated by TALYS 1.96 code.

Proton energy, MeV	Isomeric Cross-section Ratios			
	$^{154}\text{m}_1;^{154}\text{m}_2;^{154}\text{gTb}$		$^{156}\text{m}_1;^{156}\text{m}_2;^{156}\text{gTb}$	
	Experimental	TALYS 1.96	Experimental	TALYS 1.96
3.77 ± 0.80	-	-	0.66 ± 0.11	0.750
6.05 ± 0.69	-	-	0.99 ± 0.16	0.746
7.83 ± 0.63	-	-	0.86 ± 0.14	0.832
9.35 ± 0.57	-	-	0.99 ± 0.16	0.854
10.69 ± 0.57	-	-	0.82 ± 0.14	0.862
11.94 ± 0.44	0.44 ± 0.07	0.328	0.91 ± 0.15	0.852
13.07 ± 0.53	0.41 ± 0.07	0.338	0.75 ± 0.12	0.831
14.16 ± 0.41	0.39 ± 0.06	0.353	0.67 ± 0.11	0.810
15.16 ± 0.50	0.38 ± 0.06	0.367	0.66 ± 0.11	0.792
16.14 ± 0.39	0.37 ± 0.06	0.379	0.68 ± 0.11	0.774
17.06 ± 0.38	0.36 ± 0.06	0.395	0.70 ± 0.12	0.757

In Table 7, the measured ICRs are generally consistent with the theoretical predictions obtained using the TALYS 1.96 code within the experimental uncertainties. However, notable differences are observed in the behavior of energy dependence of the ICR for ^{154}Tb and ^{156}Tb . It is well established that the population probability of high-spin states increases with increasing incident particle energy. For ^{154}Tb , both metastable states correspond to high-spin configurations (see Table 1).

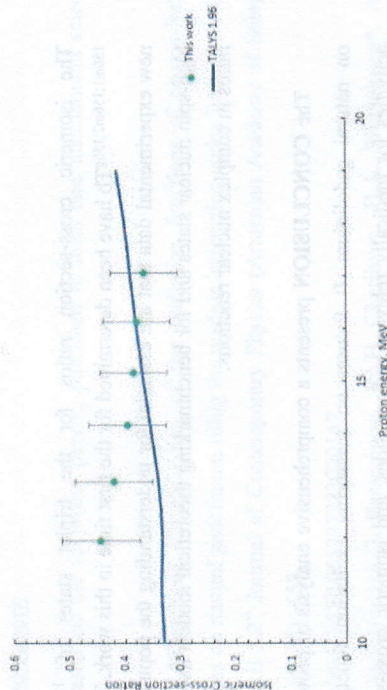


Fig. 6. ICR for $^{\text{nat}}\text{Gd}(p,xn)^{154}\text{Tb}$ reaction along with calculated by TALYS 1.96 code.

Since the ICR is defined as the ratio of the ground-state formation cross-section to the total cross-section, an increase in the population of high-spin states results in a decrease of the ICR with energy, in agreement with the experimental data (see Fig. 6). However, the ICR values calculated using the TALYS 1.96 code exhibit a slight increasing trend with rising proton energy, which contradicts the experimental data. This discrepancy indicates that the theoretical model does not adequately describe reaction mechanisms involving multiple isomeric states.

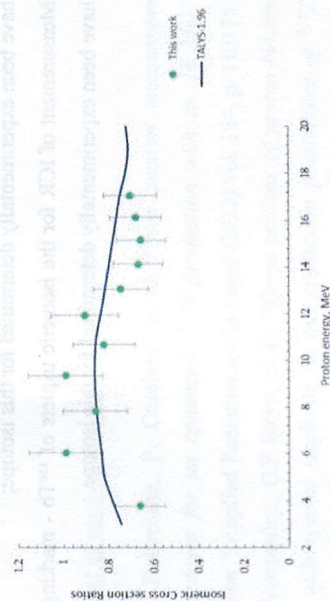


Fig. 7. ICR for the $^{\text{nat}}\text{Gd}(p,xn)^{156}\text{Tb}$ reaction along with calculated by TALYS 1.96 code.

The isomeric cross-section ratios for the triplet states of $^{154m1}, ^{154m2}, ^{154g}\text{Tb}$ and $^{156m1}, ^{156m2}, ^{156g}\text{Tb}$ have been determined for the first time in this work. These results provide new experimental data that are essential for understanding the population mechanisms of high-spin nuclear states and for benchmarking theoretical models used to predict isomeric ratios in complex nuclear reactions.

The **CONCLUSION** presents a comprehensive analysis of proton-induced reactions on natural gadolinium. It focuses on measuring excitation functions, cross-sections, activities for medically relevant radionuclides, and isomeric cross-section ratios (ICRs) across a range of proton energies up to 18 MeV. This research significantly contributes to the development of nuclear data for medical, scientific, and applied purposes.

The key accomplishments of this work are summarized below:

- Measurement of cross-sections and integral activities for the formation of the medically relevant radionuclide ^{157}Tb , which has potential use in targeted radiotherapy;
- Measurement of cross-sections for the formation of ^{160}Tb , a nuclide of interest in nuclear forensics and safeguards;
- Measurement of cross-sections for total and ground states of ^{156}Tb , ^{156g}Tb and $^{156m1}\text{Tb}$;
- Measurement of ICR for the isomeric triplets of ^{154}Tb - marking the first time ICRs have been experimentally determined for this isotope;
- Measurement of ICR for the isomeric triplets of ^{156}Tb - marking the first time ICRs have been experimentally determined for this isotope.

List of published papers

This work is based on the following papers, published in local and international scientific journals:

1. **H. A. Mkrtchyan** "Investigation of the excitation functions of proton-induced reactions on natural gadolinium using the nuclear interaction codes TALYS 1.96 and EMPIRE 3.2" *Journal of Contemporary Physics* (Armenian Academy of Sciences) 2023, 58, 151–158.
DOI: 10.1134/S1068337223020147
2. **R.V. Avetisyan, A.G. Barseghyan, Yu.H. Gharibyan, H.A. Mkrtchyan*, A.Yu. Petrosyan, I.A. Kerobyan** "Investigation of production possibility of medical isotope ^{155}Tb on cyclotron C18/18" *Journal of Contemporary Physics* (Armenian Academy of Sciences) 2024, 59, 26–34.
DOI: 10.1134/S1068337224700038
3. **R.V. Avetisyan, A.G. Barseghyan, Yu.H. Gharibyan, I.A. Kerobyan, H.A. Mkrtchyan*, A.Yu. Petrosyan** "Excitation function of proton-induced production of ^{160}Tb ", *The European Physical Journal Plus*, 2025, 140, 27.
DOI: 10.1140/epjp/s13360-024-05911-8
4. **H.A. Mkrtchyan, R.V. Avetisyan, Yu.H. Gharibyan, A.Yu. Petrosyan, W.Tan, I.A. Kerobyan** "Excitation functions of proton induced reactions on natural gadolinium", *Nuclear Inst. and Methods in Physics Research B*, 2026, 576.
DOI: 10.1016/j.nimb.2026.166146

Shortlist of used references

1. G. Dellepiane, P. Casolaro, et al. "Cross section measurement of terbium radioisotopes for an optimized ^{155}Tb production with an 18 MeV medical PET cyclotron", *Applied Radiation and Isotopes* (2022) vol. 184, p. 110175.
2. C. Vermeulen, G.F. Steyn, et al. "Cross sections of proton-induced reactions on ^{nat}Gd with special emphasis on the production possibilities of ^{152}Tb and ^{155}Tb ", *Nuclear Instruments and Methods in Physics Research B* (2012) vol. 275, p. 24.
3. **R.V. Avetisyan, H.A. Mkrtchyan, et al.** "Excitation function of proton-induced production of ^{160}Tb ", *The European Physical Journal Plus* (2025) vol.140, p. 27.

4. R.K. Chapman, A.S. Voyles, et al "Measurement of the $^{160}\text{Gd}(p,n)^{160}\text{Tb}$ excitation function from 4–18 MeV using stacked-target activation", Applied Radiation and Isotopes (2021) vol. 171, p. 109647.

5. C. Bibrattari, E. Gadioli, E et al. "Pre-Equilibrium processes in (p,n) reactions", Nuclear Physics A (1973) vol. 201, p. 579.

Ցիկլոտրոն C18/18-ի վրա արտոն-հարուցված նուկլիդների ստացման

ուսումնասիրումը բնական գադոլինիումի վրա

Ամփոփագիր

Աշխատանքը նվիրված է բնական գադոլինիումի թիրախի վրա արտոններով հարուցված ռեակցիաների միջոցով տերբիումի ռադիոնուկլիդների ստացման ուսումնասիրությանը: Տերբիումը (Tb), որը պատկանում է լանթանիդների խմբին, առանձնահատուկ տեղ է զբաղեցնում այդ տարրերի շարքում, քանի որ այն պարունակում է միջուկային բժշկության համար կարևոր չորս ռադիոնուկլիդներ՝ ^{149}Tb , ^{152}Tb , ^{155}Tb և ^{160}Tb : Այս ռադիոնուկլիդները կիրառվում է հիվանդությունների ախտորոշման և բուժման համար: Բնական գադոլինիումի վրա արտոններով հարուցված ռեակցիաները ուսումնասիրվել են ^{153}Tb , ^{154}Tb , ^{155}Tb , ^{156}Tb և ^{160}Tb ռադիոնուկլիդներ ստացման նպատակով: Գրգռման ֆունկցիաները որոշվել են համապատասխան ռեակցիաների շնային էներգիաներից մինչև 18 ՄէՎ էներգիական միջակայքում: Փորձը իրականացվել է ԱՄՆԳԼ-ում տեղակայված ցիկլոտրոն C18/18-ի արտաքին արտոնային փնջով: Գիտափորձում կիրառվել է ակտիվացիոն անալիզի մեթոդը: Պրոտոնային փնջով ճառագայթված թիրախները հետազայում վերլուծվել են զանազան սպեկտրոմետրիայի միջոցով՝ օգտագործելով CANBERRA տեսակի բարձր մաքրության գերմանիումի (HPGe) դետեկտոր, որը միացված էր թվային վերլուծիչ համակարգին: Փորձարարական եղանակով ստացված տվյալները համեմատվել են ավելի վաղ հրապարակված արդյունքների, ինչպես նաև տեսական մոդելային հաշվարկների հետ, որոնք իրականացվել են TALYS 1.96, EMPIRE 3.2 և ALICE ծրագրային փաթեթների միջոցով: Բժշկության մեջ կիրառվող ^{155}Tb նուկլիդի համար ստացվել է ինտեգրալ ակտիվությունը, ինչը վկայում է այն մասին, որ օգտագործելով հարստացված ^{155}Gd թիրախ, պրոտոնային փնջի բարձր հոսանք և սահմանվել արտոնների էներգիան 10.8

ՄէՎ, կարելի է ստանալ բարձր ռադիոնուկլիդային մաքրությամբ նուկլիդ ^{155}Tb : Միջուկային դատաբժշկության մեջ օգտագործվող ^{160}Tb նուկլիդի համար ստացվել են լայնական կտրվածքի արժեքներ, որոնք կրնալան են շրջակա միջավայրի ճառագայթային աղտոտվածության մոնիթորինգի հետազոտական տվյալների բազան: Առաջին անգամ որոշվել են իզոմեր հարարերությունները $^{154\text{m}}\text{Tb}$, $^{154\text{m}2}\text{Tb}$, $^{154\text{g}}\text{Tb}$ և $^{156\text{m}}\text{Tb}$, $^{156\text{m}2}\text{Tb}$, $^{156\text{g}}\text{Tb}$ եղանակների համար: $^{154\text{m}}\text{Gd}(p,xn)^{155,154,155,156,160}\text{Tb}$ ռեակցիաների համար ստացված փորձարարական արդյունքները ցույց են տալիս արտոնային էներգիաների ավելի լայն միջակայքում հետագա հետազոտությունների անհրաժեշտությունը՝ չափված և կանխատեսված լայնական կտրվածքների միջև դիտարկված անհամապատասխանությունները լուծելու համար:

Изучение получения протон-индуцированных нуклидов на природном гадолинии на циклотроне C18/18

Резюме

Диссертационная работа посвящена исследованию получения радионуклидов тербия в протон-индуцированных реакциях на природном гадолинии. Тербий, принадлежащий к группе лантанидов, занимает особое место в их ряду, поскольку четыре его радионуклида ^{149}Tb , ^{152}Tb , ^{155}Tb и ^{160}Tb имеют широкое применение в терапии и диагностике. Целью исследования протон-индуцированных реакций на природном гадолинии являлось определение функций возбуждения ^{153}Tb , ^{154}Tb , ^{155}Tb , ^{156}Tb и ^{160}Tb радионуклидов. Функции возбуждения были определены в области энергии протонов от соответствующих порогов реакций до 18 МэВ. Эксперимент был проведен на выведенном пучке протонов циклотрона C18/18, расположенного на территории ННПЯ. Эксперимент был выполнен с использованием активационного метода стопок. Активность облученных протонным пучком мишеней измерялась посредством германиевых детекторов HPGe CANBERRA, оснащенных цифровым анализатором. Полученные экспериментальные результаты были сравнены с ранее опубликованными данными других научных центров, а также с теоретическими вычислениями посредством кодов TALYS 1.96, EMPIRE 3.2 и пакета ALICE. Для представляющего интерес для медицины тераностического нуклида ^{155}Tb была определена интегральная

активность. Полученные данные относительно применяемого в медицине радионуклида ^{157}Tb позволяют заключить, что при использовании обогащенной мишени, высокого тока протонного пучка и ограниченном значении энергии протонов 10.8 МэВ, возможно получение радионуклида ^{157}Tb для медицины с высокой радионуклидной чистотой. Для применяемого в ядерной криминалистике нуклида ^{160}Tb получены значения сечения, которые пополнят базу исследователей данных при контроле радиационного загрязнения окружающей среды. Впервые были получены изомерные отношения для триплетов $^{154\text{m}1}\text{Tb}$, $^{154\text{m}2}\text{Tb}$, $^{154\text{g}}\text{Tb}$ и $^{156\text{m}1}\text{Tb}$, $^{156\text{m}2}\text{Tb}$, $^{156\text{g}}\text{Tb}$. Полученные экспериментальные результаты для реакций $^{153}\text{Gd}(\text{p},\text{xn})^{153,154,155,156,160}\text{Tb}$ указывают на необходимость дальнейших исследований в более широком диапазоне энергий протонов для разрешения наблюдаемых расхождений между измеренными и предсказанными сечениями.

