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GAMMA RAY PEAK AREA RATIO FOR DETERMINATION OF 15-30 MEV PROTON BEAM ENERGY

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15-30 MEV PROTON DEMETİ ENERJİSİNİN BELIRLENMESI İÇİN GAMA PİK ALANI ORANI KULLANILMASI

ABSTRACT

The energy of the proton beam produced by the cyclotron at TENMAK-PAF can be adjusted between 15 – 30 MeV by extraction systems. As the carbon stripper foil, which is part of the extraction system become closer to the center of the cyclotron, the turning radius of the beam decreases, therefore lower energy proton beam can be obtained. In this study, the proton beam energy measured by the accelerator control system was verified by using a 2 mm thick aluminum degrader, a fluorescence screen and SRIM (version 2013.00) computer program in accordance with the stopping power principle. Then a ratio table, based on the activation values of the copper foils was prepared to determine the energy of the proton beam using 5 pure copper foils of 25 μm thickness and 4 aluminum degraders of 1 mm thickness each. In the event of changes, faults and malfunctions in the accelerator control system and in the absence of expensive energy detectors, the proton beam energy can be determined practically and with a certain sensitivity by the method described in this study.

Key Words : Cyclotron, Activation, Proton Beam Energy, Stack Foil, Gamma Ray.

ÖZET

TENMAK Proton Hızlandırıcısı Tesisinde bulunan siklotron tipi dairesel hızlandırıcıdan elde edilen proton demetinin enerjisi, stripper ekstraksiyon sistemi kullanılarak 15-30 MeV aralığında ayarlanabilmektedir. Ekstarksiyon sisteminin bir parçası olan karbon folyo siklotron merkezine doğru yaklaştıkça dairesel hareketin yarıçapı azalacağından daha düşük enerjili proton demeti elde edilir. Bu çalışmada, hızlandırıcı kontrol sistemi tarafından ölçülen proton demet enerjisi, durdurma gücü prensibine uygun olarak 2 mm kalınlığında alüminyum yavaşlatıcı, floresan ekran ve SRIM (versiyon 2013.00) bilgisayar programı kullanılarak doğrulanmıştır. Daha sonra, 25 µm kalınlığında 5 saf bakır folyo ve her biri 1 mm kalınlığında 4 alüminyum yavaşlatıcı kullanılarak proton ışınının enerjisini belirlemek için bakır folyoların aktivasyon değerlerine dayalı bir oran tablosu hazırlanmıştır. Hızlandırıcı kontrol sistemindeki değişiklikler, hatalar ve arızalar durumunda ve pahalı enerji dedektörlerinin yokluğunda, proton ışını enerjisi bu çalışmada açıklanan yöntemle pratik olarak ve belirli bir hassasiyetle belirlenebilir.

Anahtar Kelimeler: Siklotron, Aktivasyon, Proton Demeti Enerjisi, Yığın Metal Folyo, Gama Işını.

1. Introduction

The cyclotron type accelerator (designed by Ion Beam Application) installed at the TENMAK Proton Accelerator Facility (TENMAK – PAF), which is operated by the Nuclear Energy Research Institute of the Turkish Energy, Nuclear and Minerals Agency (TENMAK) can provide a proton beam with the energy between 15 and 30 MeV and current up to 1.2 mA. The irradiation is performed at the solid, gas and liquid target systems placed at the end of three beam lines to produce ^{18}F, ^{123}I, ^{201}Tl, ^{67}Ga and ^{111}In radioisotopes. One of the main objectives of the utilization of TENMAK–PAF is to produce radioisotopes and radiopharmaceuticals used for diagnostic purpose in nuclear medicine. Besides, there is a separate R&D irradiation vault (~93 m^2) that is used to perform irradiations for scientific studies (Fig. 1). Since the energy resolution has great importance in the nuclear reactions, the proton energy at the reaction point must be measured precisely. The radionuclide impurity ratio of the radioisotopes and radiopharmaceuticals that should comply with the EU GMP and European Pharmacopoeia is directly related to the proton energy. Therefore, the uncertainty or incorrect measurement of proton beam energy directly effects the product quality. Any possible incompliance with the specifications given in the regulation could result in the whole batch to be wasted. In this case, delivering time of products could be delayed, which in turn could cause some problems in hospitals in timely usage of these products.

In recent years, different methods and materials have been used to measure proton beam energy by depending on the energy level. For example, semiconductor detectors are used in certain applications [1] and plastic and liquid scintillators are used in others [2, 3]. In this study, experimental measurements have been performed for the determination of the proton beam energy by foil activation method utilizing five copper foils with the dimensions of 25 µm x 2 cm x 2 cm (Thickness x Width x Length) and four aluminum energy degrader plates with the dimensions of 1 mm x 2 cm x 2 cm (T x W x L). The proton beam energies on the copper foils have been calculated by using initial energy value received from accelerator control system at the extraction point and SRIM-2013 [4]. The energy value determination has been performed by using reaction cross-section data in the literature corresponding in each copper foil's energy and the peak area of the activated copper foils' obtained from gamma spectrum of semi-conductor HPGe gamma detector.

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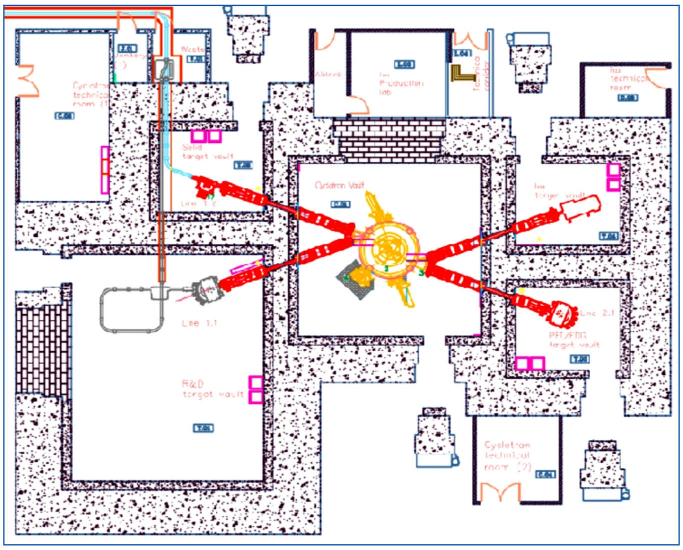


Fig. 1. The Layout of Cyclotron and Target Vaults.

2. Experimental

The proton beam energy can be produced between 15–30 MeV at the cyclotron installed at TENMAK-PAF. The adjustment is done by the stripper position in the acceleration cavity that is a part of the beam extraction system. By depending on the required beam energy, the carbon foil of the stripper system can be moved as radially into the center of cyclotron. In case of any deviation in the stripper position during this movement, the energy level required cannot be obtained precisely. In this study, the proton beam energy is verified by using SRIM-2013 based on the well-known stopping power data.

Determination of the Proton Beam Energy by Using Stopping Power

In order to verify proton beam energies monitored from the accelerator control system, computer program SRIM-2013 was used for the determination of the minimum proton beam energy level. All the protons in the beam hit on the screen after passing through 50 µm-thick Havar vacuum window and then through 2 mm-thick aluminum degrader plate. It has been determined by using the setup shown in Fig. 2 and computer program SRIM-2013. The proton energy is calculated about 20.2 MeV.

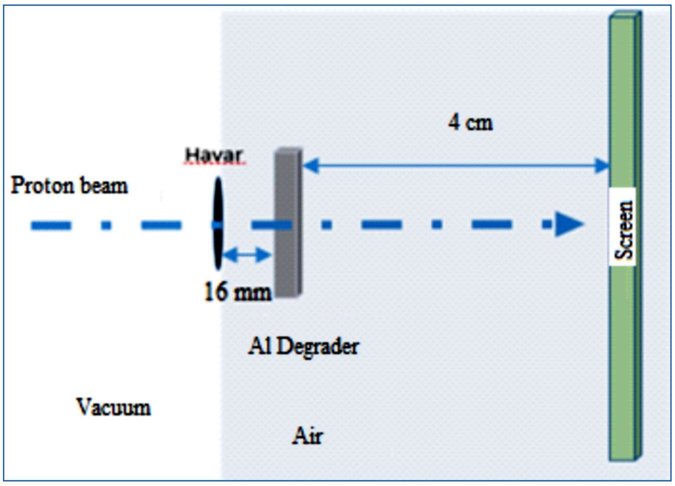


Fig. 2. Schematic view of the setup used for the determination of the stopping power of proton beam energy.

The proton beam hits on the fluorescence screen after passing through the 2 mm-thick Al energy degrader. The setup shown in the Fig 2 was irradiated by the proton beam with the 19 MeV energy and 3 µA/h current on the faraday cup just before from the exit of the beam-line. When the beam transmit from vacuum window, some current remains on the collimator. There is no beam-spot or current on the fluorescence screen at the beginning. After, the proton beam has been increased by 0.1 MeV intervals. Even in the level of 19.4 MeV and a beam current with the level of 0.5 µA/h, a beam-spot could not be observed. A 1.5 µA/h current has been monitored on the screen when the energy level has been increased to 19.8 MeV and a beam-spot shown in the Fig. 3a has appeared on the screen. When the energy level has been increased to 20.2 MeV, the current has raised to 2.1 µA/h and a beam-spot has been observed as shown in Fig. 3b. Since increasing the beam energy up to the higher levels does not lead to any change in the current anymore, all the protons in the beam are supposed to reach to the fluorescence screen. As this experimental result is in accordance with that of calculated by using computer program SRIM-2013, the proton beam energy of 20.2 MeV observed through the accelerator control system has been verified successfully.

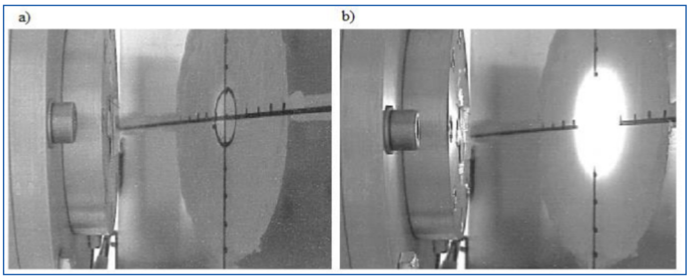


Fig. 3. a) The screen view of 19.8 MeV proton beam energy obtained from the setup shown in Fig. 2

b) The screen view of 20.2 MeV proton beam energy obtained from the setup shown in Fig. 2

Foil Activation Method

Various methods has been performed for the measurement of proton beam energy in the cyclotrons especially used for medical purpose. Different type foils, configurations, reactions and analysis techniques have been studied by using foil activation technique in the literature. In the study performed by J. H. Kim et al. [5], copper foils placed on the beam line with a certain angle has been activated by irradiation with 35 MeV proton beam energy and proton beam line energies have been verified by comparing the gamma peak areas and the cross section ratio of ^{nat}Cu(p,xn)^{62}Zn/ ^{nat}Cu(p,xn) ^{65}Zn reactions by irradiation of natural copper foils experimentally. In other literature [6, 7, 8, 9], measurements have been done by using different material types and configuration of degraders and using similar method as in this study. In these studies, more than one nuclear reaction have been utilized and gamma rays with different energy have been counted in the reactions; thus the efficiency of HPGe detector has an utmost importance. In this investigation, the gamma ray peaks belongs to ^{nat}Cu (p,n)^{63}Zn reaction by the irradiation of copper foils has been used in order to determine the proton beam energy. ^{63}Zn($T_{1/2}$=38.33 min) decays 100 % by electron capture /β^+ decay to various excited nuclear levels and the ground state of ^{63}Cu(stable). The main gamma rays of ^{63}Zn are 669.62 keV (8.2 %) and 962.06 keV (6.5 %). Since the results are very similar to each other, results calculated for only the 669.62 keV gamma peak are reported in the study.

The proton beam energy also changes with the transmission from vacuum windows of proton or any reaction through the beam line up to the target irradiation position. The foil activation method has been used at the irradiation position in order to determine the proton beam energy correctly. The experiments performed at the end of the R&D beam line, which is one of the four beam lines at TENMAK-PAF as shown in Fig. 4. Copper foils placed into the setup (shown in Fig. 4.) have been activated by irradiation at a proton beam energy between 15 – 30 MeV, which is determined by the cyclotron controlling system utilizing the stripper position. The mean proton beam energies on each of copper foils for incoming beam energy have been calculated by using computer program SRIM-2013 (Table 1). The 5th copper foil wasn't activated because of low proton energy. In accordance with the target geometry and material type to be irradiated, the elements and thicknesses of the layers were entered into the program in TRIM mode. Hydrogen element was selected for the proton beam and the angle of the beam with the target was entered into the program as 0 degrees. In the average energy calculation of the transmitted ions, the total number of ions was entered into the program as 100000. The incoming energy value between 15 and 30 MeV was defined in the TRIM program in accordance with the calculation to be made.

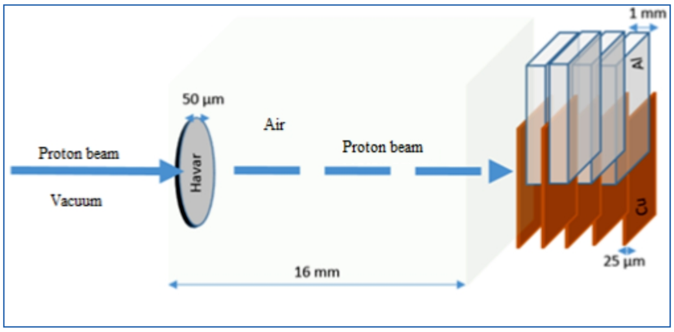


Figure 4. Schematic view of experimental setup for proton beam energy measurement.

The reaction cross-section values corresponding to the proton beam energy values given in Table 1 for ^{63}Cu (p, n) ^{63}Zn reaction has been taken from IAEA-TECDOC-1211 [10] and shown in Fig. 5. The cross- section values corresponding to the intermediate energy values have been determined by interpolation of reference values.

Table 1. The mean proton beam energies on each copper foils calculated by using SRIM.

Proton Beam Energy (MeV)	1. Cu Foil(MeV)	2. Cu Foil(MeV)	3. Cu Foil(MeV)	4. Cu Foil(MeV)
30,000	29,430	24,960	19,370	13,050
29,000	28,400	23,850	18,357	11,131
28,000	27,436	22,620	16,892	8,923
27,000	26,420	21,484	15,379	6,087
26,000	25,388	20,254	13,795	1,978
25,000	24,370	19,046	12,103	
24,000	23,352	17,758	10,241	
23,000	22,336	16,543	8,177	
22,000	21,314	15,207	5,696	
21,000	20,287	13,812	2,016	
20,000	19,247	12,404		
19,000	18,220	10,882		
18,500	17,708	10,087		
18,000	17,184	9,314		
17,500	16,664	8,477		
17,000	16,146	7,527		
16,500	15,621	6,538		
16,000	15,113	5,503		
15,500	14,582	4,259		
15,000	14,058	2,756		

The copper foils in setup (Fig. 4) have been activated by the proton beam at the energy values given in Table 1. Each irradiation have been carried out at a mean current value of 0.5 µA/h for 120 sec. The deviation for the current and irradiation duration did not change the results considerably since same quantities of particles hit on each copper foil and the ratio value is taken into consideration for the activity of copper foils.

The activation of copper foils irradiated by proton beam has been calculated by the following equation;

$$A = n\, I\, \sigma\, (1 - e^{-\lambda t_i}) \qquad (1)$$

Where;

A: The acitivity at the end of bombardment,

σ: Reaction cross section,

λ: Radioactive decay constant,

t_i: irradiation duration.

Due to the 1 mm-thick aluminum energy degraders placed around the copper foils, the proton beam energy values that falls on each foils would be different from each other. In this case, the activity value of each copper foil would be as follows;

$$A_j = n I \sigma_j\, (1 - e^{-\lambda t_i}) \qquad j{:}1,2,3,4,5. \qquad (2)$$

The activity ratio for any of two copper foils would be calculated by the following equation;

$$A_j / A_k = \sigma_j / \sigma_k \qquad j \neq k{:}1,2,3,4,5 \qquad (3)$$

Active foils have been counted by gamma spectrometry system with HPGe Detector and Genie 2000 software. So the activity at semi-conductor detector is;

$$A_j = D_j / (\varepsilon I_\gamma\, T) \qquad (4)$$

Where;

D_j:Gamma peak area,

ε:Detector efficency,

I_γ: Gamma yield,

T: Measurement time at the detector.

According to the equation (4), the activity ratio of the copper foils for any irradiation in the experimental setup would be the peak area ratio.

The cross-section ratio corresponding different proton beam energies on copper foils given in equation (3) should be in accordance with the peak area ratio of gamma rays emitted from 669.62 keV and 962.06 keV energies of ^{nat}Cu (p,n) ^{63}Zn reaction and detected by the gamma spectrometry after the activation of copper foils (Fig 6). It is not possible to get exactly the same results since the reference cross-section values and the experimental data associate with significant level of error.

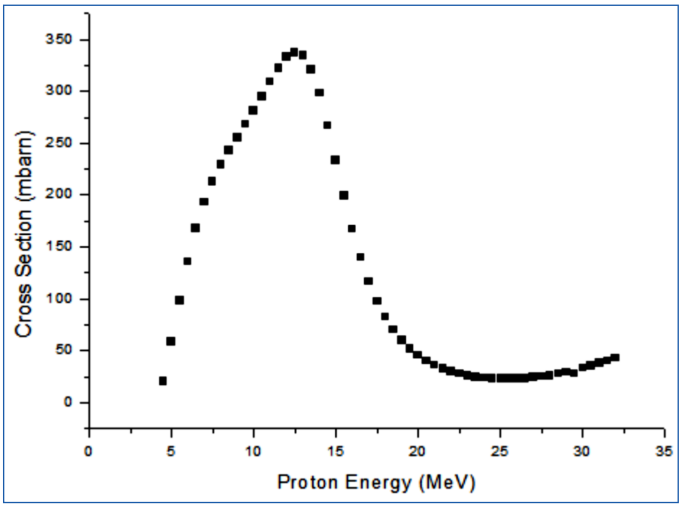


Figure 5. The cross section values for ^{nat}Cu(p, n) ^{63}Zn reaction.

The ratios of gamma peak area are obtained for proton beam energies on each copper foils (Table 2). In order to make the calculations, incident proton beam energy between 15-30 MeV is measured via accelerator control system. The calculated values are compared with the ratio of cross section values given in the literature [10]. If this conformity could be achieved, the proton beam energy would be determined with the peak area ratio obtaining from the experiments by using the experimental setup shown in Fig 4.

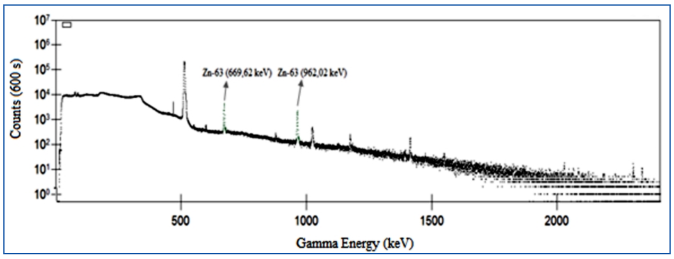


Figure 6. A gamma spectrum of an activated copper foil.

The cross section curve includes the energy values between 4.5-32 MeV (Fig. 5). To achieve cross section values corresponding the energy values given in Table 1, data taken from [10] has been divided into smaller energy level intervals by interpolation. The cross section values corresponding to the energy values of each copper foil have been assigned as σ_1, σ_2, σ_3 and σ_4. The ratio of these values has been used in the evaluation section.

Activated copper foils have been counted for 600 sec in the semiconductor HPGe detector of gamma spectrometry by using Genie 2000 software. In order to decrease the dead time during the counting, copper foils have been placed 20 cm away from the detector crystals. The peak areas of gamma rays emitting 669.62 keV which belong to the $^{nat}Cu(p,n)^{63}Zn$ reaction for 4 Copper foils has been assigned D_1, D_2, D_3 and D_4 respectively. The ratio of these peak area has been calculated for the proton beam energy values given in Table 1. The obtained peak area ratio and calculated cross section ratio have also been presented in Table 2.

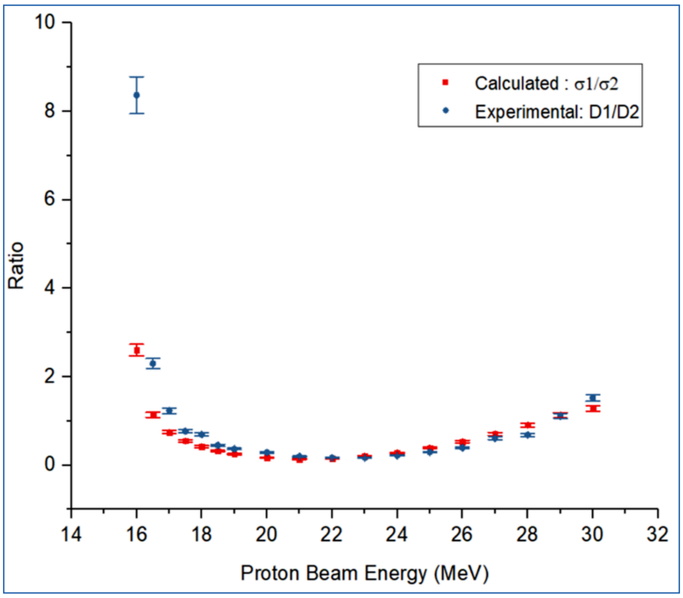


Figure 7. The peak areas ratio (D1/D2) of gamma ray at 669.62 keV emitted from of $^{nat}Cu(p,n)^{63}Zn$ reaction which occurs by irradiation of 1st and 2nd copper foils by protons energy between 15 – 30 MeV and the cross section ratios (s1/ s2) that corresponding to the energy values on each copper foils.

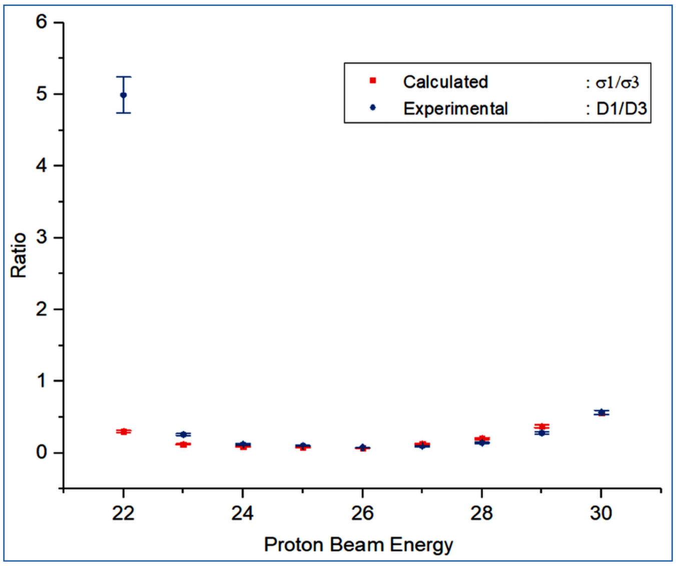


Figure 8. The peak area ratio (D1/D3) of gamma rays at 669.62 keV emitted as a result of $^{nat}Cu(p,n)^{63}Zn$ reaction which occurs by irradiation of 1st and 3rd copper foils by protons energy between 15 – 30 MeV and the cross section ratios (s1/ s3) that corresponding to the energy values on each copper foils.

Table 2. The cross section ratios (s) that corresponding to the energy values on copper foils and the peak area ratios of gamma ray at 669.62 keV emitted from activated copper foils at these energy levels.

Energy (MeV)	σ_1/σ_2	D_1/D_2	σ_1/σ_3	D_1/D_3	σ_2/σ_3	D_2/D_3	σ_2/σ_4	D2/D4	σ_3/σ_4	D3/D4
30,000	1,2783	1,51998	0,542593	0,56797	0,424444	0,37257	2,356021	0,09641	23,4676	0,25803
29,000	1,1281	1,11850	0,373973	0,28001	0,329252	0,25106	3,016529	0,12038	25,7070	0,47989
28,000	0,9113	0,68795	0,202276	0,14384	0,22541	0,21493	4,505495	0,29615	30,7235	1,39023
27,000	0,7076	0,61329	0,125323	0,09787	0,158654	0,15958	5,646036		38,1585	8,37523
26,000	0,5327	0,39526	0,07042	0,07594	0,139187	0,19213				
25,000	0,3936	0,29605	0,076189	0,10216	0,178571	0,34507				
24,000	0,2800	0,22118	0,087805	0,12066	0,313589	0,54431				
23,000	0,2065	0,17190	0,122318	0,25822	0,592275	1,50216				
22,000	0,1566	0,17005	0,300885	4,99181	1,920354	29,4425				
21,000	0,1384	0,19703								
20,000	0,1671	0,29186								
19,000	0,2525	0,36997								
18,500	0,3245	0,44888								
18,000	0,4220	0,69530								
17,500	0,5549	0,76819								
17,000	0,7465	1,23636								
16,500	1,1369	2,29798								
16,000	2,6020	8,36518								
15,500										
15,000										

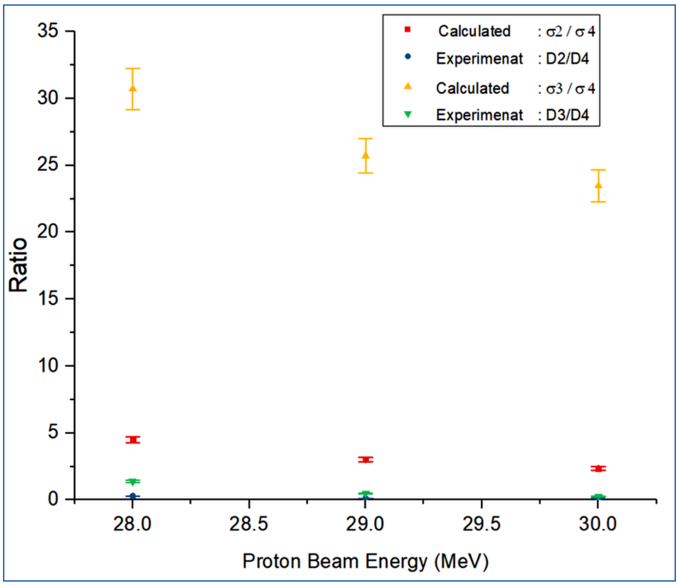


Figure 9. The peak area ratios (D2/D4, D3/D4) of gamma rays at 669.62 keV emitted as a result of $^{nat}Cu(p,n)^{63}Zn$ reaction which occurs by irradiation of 2nd and 4th copper foils and 3rd and 4th copper foils by protons energy between 15 – 30 MeV and the cross section ratios (s2/ s4, s3/ s4) that corresponding to the energy values on each copper foils.

The main sources of the errors are given in Table 3. When statistical and errors from copper foils thickness are similar for all calculated data, errors from cross section and gamma peak area change with protons energy. For the lower energy of the beam, the cross section error is higher. Furthermore the beam energy lower then 5 MeV could make low activation when hits to copper foil target. Therefore, the gamma peak area of activated target includes much more errors.

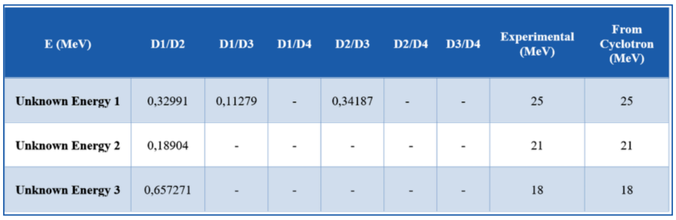
Table 3. Source of uncertainties and values.

Source of error	*Uncertainties (%)*
Statistical error	1-5
Error from copper foils thickness error(from Goodfellow)	1-15
Cross section error	≤10
Gamma peak area error	1-15

Method Verification

The irradiation has been performed by the cyclotron operator keeping the proton beam energy and using the setup given in Fig 4. Activated copper foils have been counted in HPGe gamma ray detector and after determination of energy values, the irradiation energy has been confirmed by the values read from the cyclotron control system. (Table 4).

Table 4. The constant ratio and determined energy values for the irradiated copper foils at unknown energy levels.



E (MeV)	D1/D2	D1/D3	D1/D4	D2/D3	D2/D4	D3/D4	Experimental (MeV)	From Cyclotron (MeV)
Unknown Energy 1	0,32991	0,11279	-	0,34187	-	-	25	25
Unknown Energy 2	0,18904	-	-	-	-	-	21	21
Unknown Energy 3	0,657271	-	-	-	-	-	18	18

3. Results and Discussion

The proton beam energy observed through the accelerator control system has been verified by using an aluminum degrader with the thickness of 2 mm (Fig. 2) and stopping power calculation done by SRIM computer program. The experimental set up presented in Fig. 4 was used for determination of the proton beam energy. The amount of protons on each copper foils (Fig. 4) could be assumed to be almost same due to the fact that four foils have been irradiated simultaneously and in the same position during each experiment. The gamma peaks are emitted as the result of ^{nat}Cu (p,n) ^{63}Zn reaction. ^{63}Zn peaks are located at 669,62 keV and 962 keV energy when counted on HPGe detector. The experimental ratios obtained, which are calculated by using gamma peak area are in accordance with the ratios given in the literature. Table 1 shows calculated peak area ratios and cross section ratios for proton beam energies of 15-30 MeV. The proton beam energy was changed from 30 to 20 MeV by 1 MeV intervals and from 20 to 15 MeV by 0.5 MeV intervals. The calculated peak area ratios are given in Fig. 7, 8 and 9. For proton energy Ep < 5-6 MeV on foil, a considerable deviation has been observed between calculated and experimental ratios. As the experimental error was nearly 10% in cross section values and peak areas (less activation), the difference is more significant between the results calculated by cross section values and the ratio derived from the experiment performed for the 4th foil peak area. (Fig. 9). It is shown that the proton beam energy could be determined by using the setup shown in Fig. 4 for the proton beams with energy ranges from 15 to 30 MeV produced by cyclotron. However, increase in foil number may cause more complication in deriving proton energy. The more number of foil is more control point.

It is shown that the number of activated foils and the peak area ratio of activated foils are important for the determination of the proton beam energy. In order to verify the test method, the unknown irradiation energy which is just known by the cyclotron operator was compared with the experimentally determined energy value of protons. Thus, three different irradiations ratios were calculated by activated foils peak area. The beam energies are verified easily by using Fig. 7, 8 and 9 and results are given for different energy values in Table 4.

4. Conclusions

In this study, copper foils are activated with protons having a beam energy between 15-30 MeV for determination of proton beam energy. It is very simple and practical method to check proton beam energy at the cyclotron used for both production of medical radioisotopes and research activities. This method requires four copper foils, four Al degraders and a HPGe gamma detector without any complex algorithm and computational skill. When it comes to compare with other studies carried out for the determination of proton beam energy by using foil activation method as given in literature, some additional computation and errors as the result of the efficiency curve of HPGe detector have been eliminated since the same reaction and the same peak energy have been used during the counting procedure. Proton beam energies between 15-30 MeV can be determined with sensitivity of 0.5 MeV. In order to perform much more sensitive measurements, the parameters such as foil number, materials and degrader thickness would be changed in further studies.

Acknowledgments

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