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COMPARISON OF ENERGY RESPONSE FUNCTION OF STILBENE, BC501 AND EJ309 NEUTRON GAMMA DETECTION SYSTEM

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Abstract. *The paper discusses the energy response of a single crystal stilbene and two liquid scintillator detectors, BC501 and EJ309 to a range of neutrons and gamma energies generated using a 1.7MV Tandetron accelerator at IIT Kanpur. Stilbene is a solid-state composite organic detector can be used as an alternative choice for combined neutron-gamma detection. Studies have shown that stilbene's light output response is similar to BC501. Works have also claimed a linear response of stilbene to neutrons for energies less than 5 MeV. In this work, neutrons are generated using the IIT-Kanpur 1.7MV Tandetron using C(Li^7,n) reaction. The threshold energy of the reaction and the target thickness are determined by Monte Carlo simulations. Next, we measure the pulse height distribution of various neutron energies incident on stilbene, BC501 and EJ309 of the same dimensions. The response of all the organic crystals of the study to neutrons using the Tandetron is performed on energy spanning the fission neutron energy range to fast neutron energy range. A general-purpose Monte Carlo simulation kit, GEANT4, is used for simulating the reaction and detector response behaviour. Stilbene shows 38% lower energy response than that of EJ309 and BC501 shows 11% lower energy response from EJ309 for the entire neutron spectrum. These responses are consistent as the number density of hydrogen of the same mass of stilbene, BC501 is 38% and 11% lower than EJ309, respectively. GEANT4 simulation allows a detailed analysis of detector response physics for the advancement of detector development for nuclear security applications.*

Keywords: *Scintillation detector, stilbene, BC501, EJ309, Monte Carlo, GEANT4, energy response, pulse height distribution*

1. INTRODUCTION

Measurements of neutrons are of great importance in multiple fields of studies such as neutron diagnostic systems, reactors dynamics, accelerator reactions etc. Fast neutron production utilizes reactions triggered by charge particle such as protons as a projectile on low Z material [1]-[2] can be employed for ion-low Z material interaction. The neutrons emitted from these processes are of great importance for the study of nuclear detector response for developing fast neutron detection system for nuclear security application such as shielding studies, a medical application like boron neutron capture treatment (BNCT) and linear energy cancer therapy (LET) [3].

This study primarily emphasizes generating fast neutrons till 10 MeV by scattering Li-7 ions on Teflon sheet using 1.7 MV Tandetron at IIT-Kanpur. At Li-7 ion energy of 4.5 MeV, a set of numerous reaction possibilities opens up and generates a wide range of neutron spectra and its associated gamma spectrum. In this study, C (Li^7, n) reaction is simulated with a Teflon target using the Monte Carlo method, GEANT4. This study enabled us to identify all the different reactions using 4.5 MeV Li^7 ion scattered over a 350-micron thick Teflon sheet target. The thickness of the Teflon target was determined by simulations. The neutron yield peaks at lower energy and ranges all the way to 10 MeV with decreasing yield at high energy ranges. Gamma spectrum is also simulated for the same source. Further pulse height distribution is calculated and compared for 3 different scintillation detectors stilbene, BC501 and EJ309 for the same dimensions.

This paper is organized as follows: Section 2 contains the theory of the Tandetron, a method to generate neutron sources using particle accelerators and details about stilbene, BC501 and EJ309 organic scintillation detectors. Section 3 describes the modelling of Tandetron in Geant4 using detector construction. This section also describes the modelling of C(Li^7,n) reaction and the scintillation detectors using Monte Carlo Methods, Geant4 simulations. Section 4 contains a description of simulation results for generating the neutron and gamma spectrum and compares the pulse height distributions of spectrums for stilbene, BC501 and EJ309 detectors. In section 5 we describe the limitations of the study to present date and suggestions for future endeavours.

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2. THEORY

2.1. 1.7 MV IIT-Kanpur Tandetron

IIT-Kanpur Tandetron [4] is a linear tandem type ion beam accelerator with a terminal voltage of 1.7 MV. The charged particle can be accelerated to a maximum energy of 4.5 MeV. The length of the accelerating tube is 1 m and it is made of stainless steel. The interior of the accelerating chamber is maintained at an ultra-high vacuum. The accelerator produced a square beam of dimensions 2x2 mm² and an average beam strength is 300 μA. Typically 1 μA is approximately 10^{10} accelerated charged particle. The acceleration of ions in this Tandetron occurs in two steps. First, the positive electric field on the terminal in the middle of the accelerating tube accelerates the injected negative monovalent ions. One or more electrons are stripped of the ion through the process of charge exchange. The resulting positive beam is further accelerated to ground potential. The final energy of the emerging particle depends on the charged state q of the ion and is relatable by

$$E = (q+1) \times 1.7 MeV \tag{1}$$

2.2 The C (Li⁷,n) reaction

Production of neutrons using particle accelerators is the fusion of light nuclei. While other reactions exist, present reactions as represented in Table1 are among the most useful ones due to the ease of execution [5]. All reactions shown in Table 1 can be employed in open vacuum accelerating systems except for the reactions involving deuterium and tritium. The reactions involving deuterium and tritium are not preferable from the perspective of radiation safety. Hence utilizing protons and ions as a projectile to be scattered on a target is preferable. Interactions of ions with atoms or nucleus occur by predominantly three mechanisms i) inelastic interactions, ii) deflection of ion trajectories by repulsive elastic scattering with nuclei, iii) non-elastic interactions [5]. In this study, neutron generation is made feasible by scattering lithium-7 ions over a Teflon target. The predominant ion interaction is by inelastic nuclear reactions. In this reaction, the projectile lithium ion enters the nucleus of carbon leading to the emission of deuterons, protons, tritium, one or more neutrons in addition to heavy ions. The study includes modelling of the nuclear reactions with lithium-7 ions to produce neutrons using particle generator [6]. The reaction for this study is C (Li^7, n)

$$E_n^{1/2} = \frac{(M_{Li}M_nE_{Li})^{1/2}\cos\theta \pm \left[M_{Li}M_nE_{Li}\cos^2\theta + (M_{Li}+M_C)\{M_CQ + (M_C - M_{Li})E_p\}\right]^{1/2}}{M_n + M_C} \tag{2}$$

where subscript n, Li, C refers to neutron, Lithium-ion, and carbon respectively, θ is the neutron emission angle and Q represents the Q-value of the reaction.

The reaction chosen for our study C (Li^7, n) has an advantage over all the aforementioned reactions in table 1 due to the easy availability of target Teflon tape and its superior mechanical property to handle high power densities and incorporate forced cooling and the ease of lithium-ion production using the 1.7 MV IIT-Kanpur Tandetron. Reactions which occur when Lithium-7 ions scatter onto the Teflon target are shown in Table 2.

Table 1. Common nuclear reactions for neutron generation using particle accelerator

Reactions	Q-value (MeV)	Threshold Energy (MeV)	Minimum product nuclei energies (MeV)	Minimum Neutron Energy (MeV)
$H^2(d,n)He^3$	+3.269	NA	He^3:0.82	2.45
$H^2(d,n)He^4$	+17.589	NA	He^4: 3.45	14.05
$Li^7(p,n)Be^7$	-1.644	1.880	Be^7: 0.21	0.30
$H^2(d,n)Be^8$	+15.031	NA	Be^8: 0.21	13.35
$Be^9(d,n)B^9$	-1.850	2.057	B^9: 1.68	0.023
$Be^9(d,n)B^{10}$	+4.361	NA	B^{10}: 0.40	3.96

Table 2. Nuclear reactions for lithium-7 ion interaction with a Teflon target

Reaction	Q-value	Type of Reaction
$Li^7 + C^{12}$ --> $Li^7 + C^{12}$	276.52 eV	Elastic
$Li^7 + C^{12}$ --> N gamma + F^{19}	17.926 MeV	Inelastic
$Li^7 + C^{12}$ --> N gamma + alpha + N^{15}	13.914 MeV	Inelastic
$Li^7 + C^{12}$ --> N gamma + deuteron + O^{17}	4.1121 MeV	Inelastic
$Li^7 + C^{12}$ --> N gamma + neutron + F^{18}	7.4941 MeV	Inelastic
$Li^7 + C^{12}$ --> N gamma + neutron + alpha + N^{14}	3.0802 MeV	Inelastic
$Li^7 + C^{12}$ --> N gamma + proton + O^{18}	9.9331 MeV	Inelastic
$Li^7 + C^{12}$ --> gamma + triton + O^{16}	6.2264 MeV	Inelastic
$Li^7 + C^{12}$ --> neutron + alpha + N^{14}	3.0801 MeV	Inelastic
$Li^7 + C^{13}$ --> $Li^7 + C^{13}$	315.55 eV	Elastic
$Li^7 + C^{13}$ --> gamma + 2 neutrons + F^{18}	2.5478 MeV	Inelastic
$Li^7 + C^{13}$ --> gamma + alpha + N^{16}	11.456 MeV	Inelastic
$Li^7 + C^{13}$ --> gamma + deuteron + O^{18}	7.2111 MeV	Inelastic
$Li^7 + C^{13}$ --> gamma + neutron + F^{19}	12.98 MeV	Inelastic
$Li^7 + C^{13}$ --> gamma + neutron + alpha + N^{15}	8.9672 MeV	Inelastic
$Li^7 + C^{13}$ --> gamma + proton + O^{19}	8.9424 MeV	Inelastic
$Li^7 + C^{13}$ --> gamma + proton + neutron + O^{18}	4.9868 MeV	Inelastic
$Li^7 + C^{13}$ --> gamma + triton + O^{17}	5.4231 MeV	Inelastic

2.3. Stilbene, BC501 and EJ309 Scintillation Detectors

Organic scintillation detectors like stilbene BC501 and EJ309 are non-hygroscopic in nature. These detectors have fast neutron detection efficiency and inherent pulse shape discrimination capabilities for neutrons and gamma-ray events [7]. Stilbene is a solid-state scintillator crystal whose chemical composition is $C_{14}H_{12}$ and has a density of 0.971 g/cm³ [8]. BC501 is a liquid organic scintillator with a chemical formula of C_9H_{12} and a density of 0.901g/cm² [9]. Likewise, EJ309 is a low flashpoint pulse shape discrimination liquid scintillator based on solvent xylene as an alternative to the toxic EJ-301 (NE-213) manufactured by Eljen Technology. The H:C ratio for EJ309 is 1.28 and a flashpoint of 144°C which reduces the fire risks hazards associated with low flashpoint liquid scintillation material like EJ301 or NE213 [10]. BC501 and EJ309 being liquid scintillators are less prone to radiation damage, thermal and mechanical shock when compared to stilbene crystal. Studies have shown that light output response for liquid scintillators is greater than that of the solid-state single crystal stilbene detectors [7]. Also, the performance of n-γ separation property of both solid-state detectors and liquid scintillation detectors demonstrates relative linearity for monoenergetic neutrons. [11]

3. Methods

3.1. Modelling of IIT-Kanpur 1.7 MV Tandetron Accelerator

The purpose of this paper is to simulate the C(Li^7,n) reaction in the IITK-Tandetron using the Monte Carlo simulation kit GEANT4. The maximum energy to which Li-ion can be accelerated to is 4.5 MeV and can be seen from Eq. (1). For simulation purposes, the acceleration tube constructed is a stainless steel 1meter tube filled with ultra-high vacuum. The vacuum chosen material is G4_Galactic imported from the NIST database [6]. To minimize the computation time, the dimension of the Teflon target is the same as that of the source. The detector is placed at a distance of 0.5 meters from the target to obtain the detector energy response and eventually pulse height distribution. All geometries are modelled in the G4DetectorConstruction class of the application code. To simplify the simulation, monoenergetic Li-7 beams with 2mm dimensions are modelled using G4PrimaryGeneratorAction class where G4GeneralParticleSource (GPS) is used. GPS enables the user to control particle selection, its initial coordinates, its direction, energy, and distribution of the particle.

3.2. Modelling the C(Li^7, n) Reaction

A 2×2 mm^2 Teflon sheet is modelled in the user-defined class of "DetectorConstruction". The model "QGSP_BIC_HP" [12] is used to model the physics of the Li-7 ion interaction with matter. The model is known as the Quark-Gluon String Pre-compound binary cascade. This model is used for primary ions, protons and neutrons (projectiles/sources) below the initial energy of 10GeV. This physics list is mainly used for modelling radiation protection, shielding design and medical application related problems.

The primary ion source consists of a 2-mm diameter cylindrical beam of Li-7 ions starting at a distance of 5 cm from Teflon target. Teflon target has a chemical formula of C_2F_4 with a density of 2.2g/cm^3. The reaction for generating neutrons happens with Carbon atoms of Teflon. This reaction is modelled in "PrimaryGeneratorAction" class with modular "GeneralParticleSource" (GPS) class. Using this class, for a given particle its energy can be manipulated via a user input file linked to the "PrimaryGeneratorAction" class. This class modifies the particle's direction and energy with each collision thus utilizing the mean free path. Mean free path is the average distance travelled by a particle with certain energy between successive collisions. In this simulation, the total absorption length (mean free path) is computed as the average track length of the Li-7 ion. As a test, to ensure that interaction takes place at any event, the target is made infinite. Once confirmed, the target and source are modelled according to the dimensions. The results are tallied with the cross-section available in G4HadronicProcessStore and used by GEANT4. The list of nuclear reactions that occur are taken as the output as shown in Table 2. It can be seen from Table 2 that out of 17 reactions only 7 reactions contribute to neutron production via C^{12}(L^7, n), and C^{13}(Li^7, n) reactions.

4. Simulation results

This section is subdivided into two subsections. The first subsection discusses, neutron generation leading to the neutron spectrum. The second subsection discusses the pulse height distribution due to the neutron spectrum and comparing stilbene, EJ309 and BC501.

4.1. Neutron spectrum determination

The C(Li^7, n) reaction has seven probable reactions for neutron generation. To determine at what energy Lithium-ion, react with carbon atoms, reaction threshold energy is determined as shown in Figure 1.

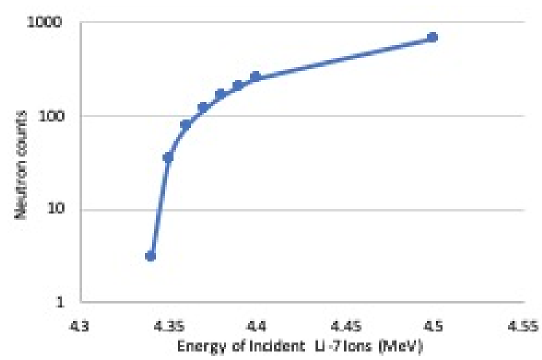

Figure 1. Threshold Energy of C(Li^7,n) reaction

For minimizing computational time, 10^6 Li^7 ions is scattered on a 300-micron Teflon sheet. It is seen that till energies of 4.3 MeV incident ions, no neutrons are produced. It is observed that at 4.34 MeV, three neutrons are generated per 10^6 incident lithium ions. It also can be observed that the neutron count increases drastically with increasing energy. Thus the threshold energy for C(Li^7, n) reaction is 4.34 MeV. Additionally, it can be seen that to obtain sufficiently high flux of neutrons, determination of the thickness of the Teflon target is done and further a tally is made for total neutrons created with its mean energy as shown in Figure. 2.

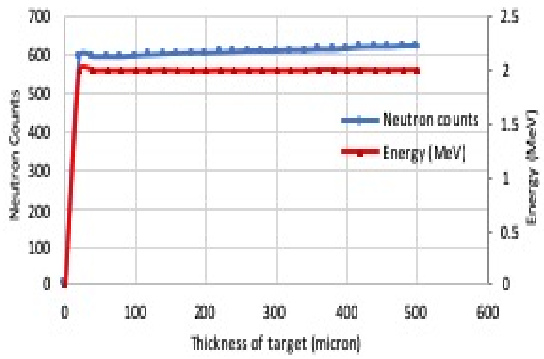

Figure 2. Teflon target thickness versus the total number of neutrons and its mean energy

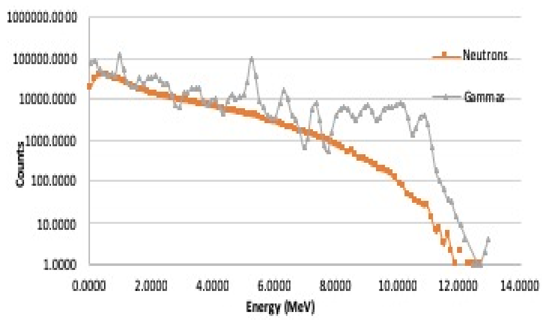

Figure 3. Neutron and gamma spectrum

Table 3. By-products of the interaction of Li^7 ion beam with C^{12} and C^{13}

Particle generated	Counts	Mean Energy	Energy range
C^{12}	106574	0.019705 eV	0.019705 eV --> 0.019705 eV
C^{13}	1139	0.016407 eV	0.016407 eV --> 0.016407 eV
F^{18}	14419	1.8383 MeV	304.56 keV --> 3.8071 MeV
F^{19}	87	1.7784 MeV	387.97 keV --> 3.3938 MeV
N^{14}	49262	2.1062 MeV	719.06 eV --> 5.3123 MeV
N^{15}	32908	2.9548 MeV	1.0321 keV --> 8.0613 MeV
N^{16}	300	2.5697 MeV	16.972 keV --> 6.6928 MeV
O^{16}	100	1.7449 MeV	288.44 keV --> 3.2467 MeV
O^{17}	443	1.8505 MeV	326.66 keV --> 3.8913 MeV
O^{18}	1893	1.7727 MeV	453 keV --> 3.2402 MeV
O^{19}	28	1.5365 MeV	496.9 keV --> 3.1469 MeV
Alpha	82470	4.7214 MeV	490.19 keV --> 14.706 MeV
Deuteron	442	2.3259 MeV	623.61 keV --> 5.1923 MeV
Gamma	143074	2.9785 MeV	156.02 eV --> 13.155 MeV
Neutron	64373	2.047 MeV	46.301 eV --> 12.478 MeV
Proton	1915	3.576 MeV	1.2579 MeV --> 9.212 MeV
Triton	107	2.1788 MeV	637.72 keV --> 5.4415 MeV

After determining the required thickness of the Teflon target and most achievable energy of the Li^7 beam after acceleration, the energy spectrum of neutrons and gammas are simulated. From Table 2, it is seen that there are 7 reactions out of 18 reactions contribute to neutron production and are inelastic in nature. Since the reaction is exothermic in nature, the Q- value of the reactions would not play a significant role and only the acceleration energy of the lithium-ion will lead to the reaction taking place.

The simulation leads to a single neutron spectrum accommodating all 7 reactions. The total number of neutrons generated from combined reactions is 646139 neutrons and 1435942 gammas for a 10^9 Li-7 beam strength. The combined spectrum of neutrons and gammas is shown in Figure 3.

The energy range of the combined neutron spectrum is 65 keV to 12.6 MeV with a mean of 2.015 MeV. Additionally, gamma rays are also generated having major peaks at 975 keV and 5.27 MeV. Other particles are also generated as a by-product of Li^7 interaction with C^{12} and C^{13} atoms of Teflon sheet along with its counts, mean energy, and their energy range are shown in Table 3.

To verify that the total number of particles incident are conserved, the number of primary particles interacting with Teflon which survived are scored. Additionally, particles other than neutrons generated from Li^7 beam reaction are also scored.

4.2. Pulse height distribution determination

This section describes pulse height distribution simulated for neutron spectrum using EJ309, BC501, and the stilbene organic scintillation detector. The detectors are modelled using GEANT4 as shown in Fig. 5. Studies have shown to achieve resolved events for neutrons using EJ309, BC501 and stilbene a typical threshold of 200 keV of neutron energy is measured [13]. Hence, we deduce that all the organic scintillators respond to the spectrum as shown in Figure 4.

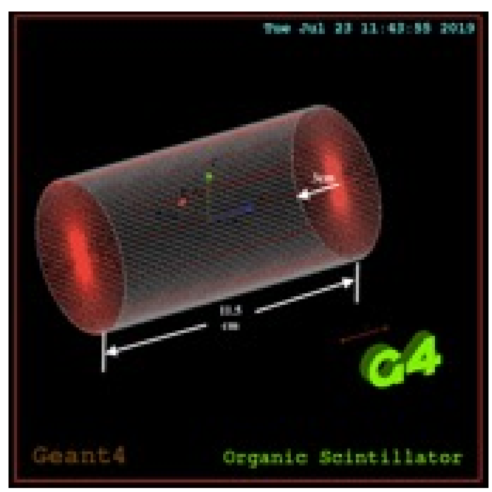

Figure 4. Detector geometry

The input neutron spectrum is normalized to its total count and 643730 neutrons are used to minimize the variance. Pulse height distribution of all the three organic scintillators for the spectrum is shown in Figure 5.

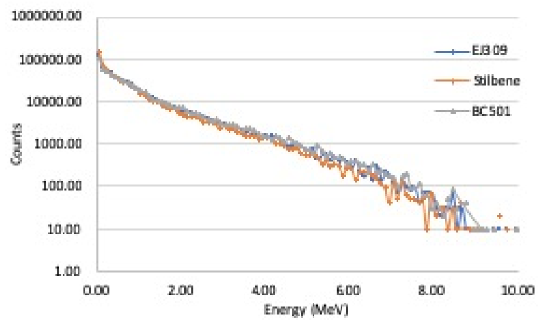

Figure 5. Pulse height distribution of EJ309, BC501 and stilbene detector

It is observed that BC501 shows 11% lower energy responses and stilbene shows 38% lower responses than EJ309 for entire neutron energy spectrum. This phenomenon can be explained from the number density of hydrogen in both stilbene and BC501. The number density of hydrogen for the same mass of EJ309 (C_8H_{10}) is 4.95×10^{21}. In BC501 (C_9H_{12}), the number density of hydrogen is 4.38×10^{21} and for the same in stilbene crystal ($C_{14}H_{12}$) it is 3.044×10^{21}. This means the number density of BC501 for the same mass of EJ309 is 11% lower and for stilbene is 38% lower which corresponds to the simulated energy response of detectors for neutrons.

5. Conclusion

In this paper, we have explored a way to generate neutrons up-to 12.5 MeV by Li^7(C,n) reaction with 1.7 MV IIT-K Tandetron, simulated using a general-purpose Monte Carlo simulation kit, GEANT4. Using Tandetron specifications, the maximum energy achieved by accelerating Li^7 ions is 4.5 MeV. Simulations are performed to determine the threshold energy of the reaction. After the determination of threshold energy, the target thickness is determined and 300 microns were selected to achieve sufficient neutron counts. Next, using the Li^7(C,n) reactions, it is found that out of 18 probable reactions, 7 reactions produce neutrons whose energy ranges from 65 eV to 12.5 MeV peaking at 455 keV as seen in Fig. 4. Furthermore, the pulse height distribution for neutron spectrum is analysed for organic scintillator detectors EJ309, BC501 and stilbene crystal. Since these scintillation detectors have a threshold energy of approximately 200 keV as found in the literature, detectors will only respond to the neutron spectrum above 200 keV. Thus any such detectors will respond to essentially the entire spectrum generated by the Li^6(C,n) reaction. It is observed that BC501 exhibits 11% less energy response in comparison to EJ309 and stilbene shows 38% less energy response which is in accordance with the number density of hydrogen atoms in the respective scintillation detector. Hence, we conclude that the energy response of BC501 and stilbene detector is comparable for neutron energies up-to 12.5 MeV in comparison to EJ309 making both these detectors a good alternative for neutron detection keeping in mind the toxicity of EJ309. In future, simulations, as discussed above, will be validated with measurements by using the 1.7 MV Tandetron at IIT-Kanpur and will determine the light output response and calculate the efficiency of all three detectors.

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