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Spatial distribution of moderated neutrons along a Pb target irradiated by high-energy protons

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Abstract

High-energy protons in the range of 0.5–7.4 GeV have irradiated an extended Pb target covered with a paraffin moderator. The moderator was used in order to shift the hard Pb spallation neutron spectrum to lower energies and to increase the transmutation efficiency via (n,γ) reactions. Neutron distributions along and inside the paraffin moderator were measured. An analysis of the experimental results was performed based on particle production by high-energy interactions with heavy targets and neutron spectrum shifting by the paraffin. Conclusions about the spallation neutron production in the target and moderation through the paraffin are presented. The study of the total neutron fluence on the moderator surface as a function of the proton beam energy shows that neutron cost is improved up to 1 GeV. For higher proton beam energies it remains constant with a tendency to decline.

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Keywords: Spallation reactions; Neutron sources

1. Introduction

Intense neutron production is possible by spallation sources in high-energy proton accelerators. Such sources can be used for energy production by accelerator driven systems (ADS) and transmutation experiments. The design of a spallation setup depends on the required neutron spectrum, for (n,f) or (n,γ) reactions. By irradiating heavy targets with high-energy protons, a hard spectrum is obtained which contains only a small number of neutrons in the (n,γ) resonance range. Over the last decade many experiments have been performed by proton irradiation on thick heavy targets in order to study the behavior of the neutron production at proton energies of hundreds of MeV to about 10 GeV [1–6]. The neutron cost defined as the number of produced neutrons per incident proton and GeV is found to be advantageous around 1 GeV proton beam for thick Pb target [5,7,8]. Regarding the increase of the transmutation efficiency via (n,γ) reactions, a paraffin moderator was applied around the Pb target in order to shift the spallation neutron spectrum to lower energies. Repeated irradiations were performed with proton beams in the energy range of 0.5–7.4 GeV [9–11].

The objective of the current research was to explore the influence of the paraffin moderator upon the spallation neutron produced by a thick heavy solid target. The moderated neutron spatial distribution was measured along the paraffin surface parallel to the beam axis. In addition, the neutron behavior was examined inside the paraffin moderator perpendicular to the beam direction and downstream. An analysis of the experimental results

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based on particle production via high-energy interactions on heavy targets and neutron spectrum shifting by the paraffin is presented. In addition, the cost of producing a neutron as a function of the proton beam energy was determined by observing the neutron behavior along the paraffin moderator surface.

2. Experimental

Synchrophasotron/Nuclotron accelerators, at the Laboratory of High Energy (LHE) JINR, Dubna were provided the proton beams used by the current study. Most of the experiments were performed with protons in the energy range of 0.5–7.4 GeV with integrated beams in the order of 10^{11} protons [9–11]. The experimental setup consisted of a Pb target surrounded by a 6 cm thick paraffin moderator (Fig. 1). The Pb target was constituted by a stack of 8 cm in diameter disks with an overall length of 20 cm.

Solid State Nuclear Track Detectors (SSNTDs) were used for neutron detection. CR39 detectors were placed parallel to the target axis on the paraffin surface, every 3 cm. One part of the detector was in contact with a neutron converter, (Kodak LR115 type 2B, containing $Li_2B_4O_7$). This converter provides information about neutron fluence, detecting the alpha particles' tracks on CR39 emitted via $^{10}B(n,\alpha)^7Li$ and $^6Li(n,\alpha)^3H$ reactions. Another part of CR39 was in contact with the converter and was covered on both sides with 1 mm Cd foils. The slow neutron component was calculated by subtracting the measured track density of the Cd-covered from the Cd-uncovered region of the CR39 detector. The fast neutrons were determined by proton recoil tracks on the detector itself (neutron elastic scattering on H of the detector) [12]. The neutron energy region detected by proton recoils is between 0.3 and 3 MeV due to limitations in the proton registration efficiency [13].

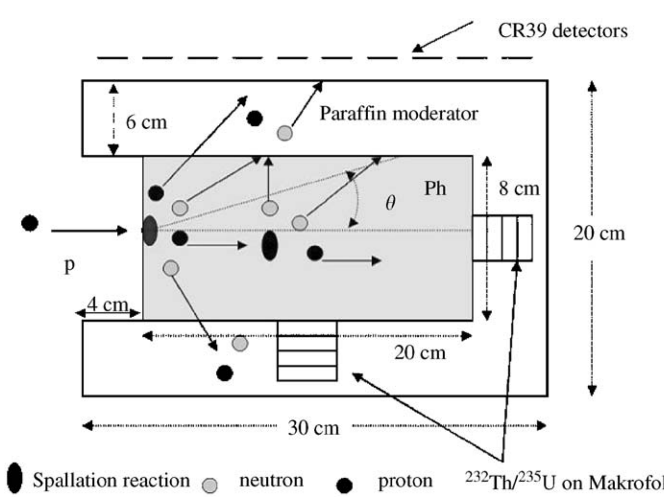

Fig. 1. A cross-section of the experimental setup with a sketch of particle multiplication along the target.

SSNTDs were also used as fission detectors. The fissioning target of approximately 100 μg cm^{-2} was evaporated on Makrofol detector. Targets of ^{235}U and ^{232}Th were used in order to study the slow and fast neutron distributions inside the moderator [11]. A separate experiment with 10 cm thickness paraffin was performed with proton beam energy 1 GeV. Two vessels were used to monitor fission rates within the paraffin. One vessel was placed at the center of the moderator at 90° relative to the beam direction. The other vessel was placed downstream, on the beam axis at 0° (Fig. 1). After irradiation the target was removed and fissions were counted on the detector (after appropriate chemical development).

3. Results and discussion

The neutron distributions inside the paraffin moderator are presented in Figs. 2(a) and (b) corresponding to the two directions: perpendicular to the beam at the center of the target and downstream, on the beam axis at 0°, as a function of the paraffin thickness measured from the Pb target surface. Slow neutrons dominate in both directions relative to the beam axis with almost equal fluences while downstream there are more fast neutrons than in the

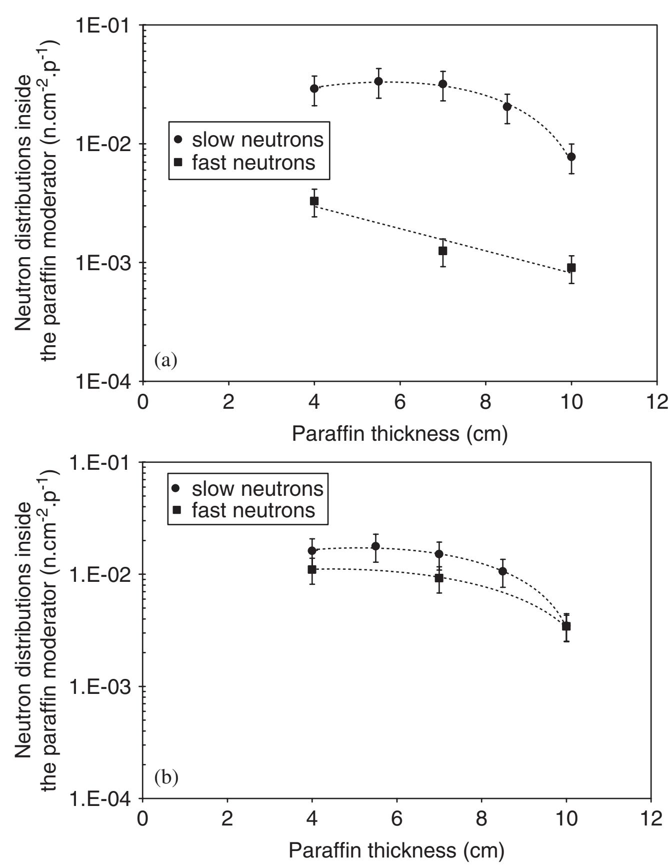

Fig. 2. (a) The neutron distribution measured inside the paraffin moderator, perpendicular to the beam axis, as a function of the paraffin thickness (the lines are draw to guide the eye). (b) The neutron distribution measured inside the paraffin moderator, downstream, as a function of the paraffin thickness (the lines are draw to guide the eye).

perpendicular direction. The fast neutrons' decrease, while crossing the paraffin moderator, exhibits a more intense thermalization in the vertical direction than downstream. However at 0° their drop-off is less sharp than at 90° due to the difference of the incoming neutron spectrum from the target. As it is known from high-energy kinematics high energy secondary neutrons follow the beam direction while at 90° their energy is considerably lower.

The neutron distributions along the moderator surface were found to be similar for all the proton beam energies in the range from 0.5 up to 7.4 GeV. A representative neutron distribution corresponding to a proton energy of 1 GeV is presented in Fig. 3. Slow and fast neutrons show a similar behavior along the paraffin surface. The total number of neutrons, n, was taken as the sum of slow and fast neutrons. A common observation for all the proton energies was that the main part of the neutron production takes place at about 15 cm along the paraffin moderator corresponding to about 10 cm into the Pb target (see Fig. 1). After the maximum neutron production is reached the distribution decreases towards the end of the paraffin. These results were similar to those found in previous experiment [11] and agree with MC calculations describing neutron distributions along and inside the moderator [14].

To study the neutron multiplication and to estimate the neutron production cost a fitting process was applied to the experimental results in order to calculate the total number, n_{tot}, of produced neutrons from the entire cylindrical surface of the paraffin. The best fitting was achieved using a gaussian function superposed to a quadratic background. Taken into account the fitting results of each proton energy, the mean centroid of the gauss function was found to be at 14.8 ± 1.2 cm on the moderator corresponding to 10.8 ± 1.2 cm in the Pb target.

Using the results of the fitting procedure the total number of measured neutrons at each position, n, along the moderator was normalized to the total number of neutrons, n_{tot}, escaping by the entire cylindrical surface of the target. The representative normalized total neutron distribution, n/n_{tot}, was determined using SSNTDs, and corresponded to proton energy of 1 GeV as presented in Fig. 4. The measured total neutron distributions along the outside paraffin surface were related to the spallation reactions taking place in the Pb target and the moderation of secondary neutrons inside the paraffin. Therefore, a two-steps procedure was applied in order to analyze the experimental results.

In the first step of the data analysis particle production were calculated according to high-energy interactions with heavy targets, as by Sullivan [15]. In front of the target, secondary particles are produced by direct interactions of the proton beam with the Pb, mainly by spallation reactions. Going deeper into the Pb target, primary and secondary reactions led to further particle multiplication. Some of the secondary particles have enough energy to give higher order production but the main part of the particles arises from reactions induced by primary and secondary interactions. Thus, the following contributions were taken into account: (1) spallation neutrons and protons produced from proton beam interactions with the Pb target (secondary particles). Neutron production by secondary protons was also taken into account, according to their average energy. For 1 GeV and below, the proton beam produces particles having an average energy of about 120 MeV while above 1 GeV the average particle energy was found to be around 200 MeV. At those energies spallation reactions have about the same cross section (1.8 b) as at 1 GeV (1.6 b) [16]. (2) An exponential proton beam attenuation along the target was included in the calculation, with an interaction length (λ) of 18 cm for Pb. An exponential attenuation of secondary protons with the same λ was assumed because of its non-significant variation with energy. (3) Secondary particle angular distributions: in the calculation only proton and neutron distributions were taken into account and only neutrons crossing the moderator were accounted for. This means that the main particle emission is at angles from about 10° up to 90°, (Fig. 1). Particle contribution at larger angles was assumed negligible. (4) Neutrons and protons coming from previous interactions in the target were also calculated at each position. Proton interactions from distances 10 cm, before

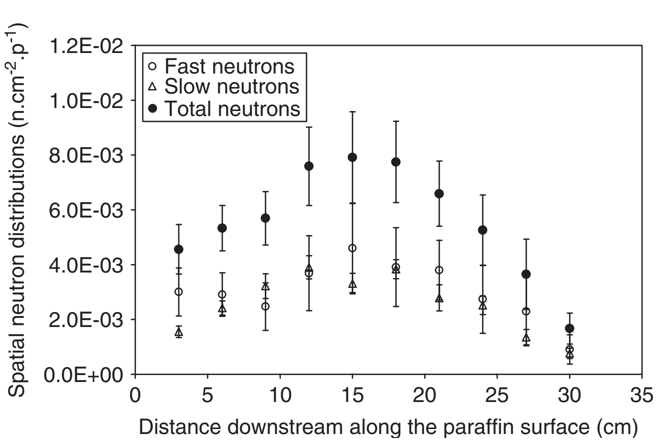

Fig. 3. The neutron distribution measured along the paraffin moderator surface corresponding to 1 GeV proton beam.

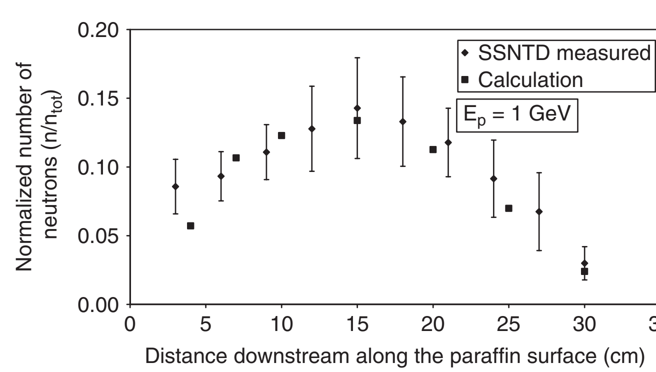

Fig. 4. A comparison between the normalized neutron distributions along the paraffin moderator surface measured by SSNTDs and calculated for 1 GeV proton beam.

the considered point, were also added because of the protons mean free path in Pb, about 10 cm. For 3.7 and 7.4 GeV a correction for fission probabilities by secondary particles (mainly for protons) was applied.

In the second step, the moderation of the neutrons passing through the paraffin moderator was taken. The results obtained using a relationship related to dose calculations behind shielding of high-energy proton accelerators. The reason of using such relationships is that they include spectral parameters in the dose calculation, due to the energy dependency of neutron quality factor. The number of neutrons produced in the Pb target was used as the number of incident neutrons in the paraffin moderator. The neutron fluence after the paraffin is given by the following relationship [17]:

$$\Phi(x,\theta) = \frac{\Phi_0(\theta)}{r^2}\exp\left[-\frac{x}{g(\theta)\lambda}\right]. \quad (1)$$

The $\Phi_0(\theta)$ describes the number of neutrons crossing at 90° the paraffin surface because registration efficiency on the CR39 detector has its maximum value at 90° angle. The r corresponds to the distance of the primary proton interaction in the target, x is the paraffin thickness (6 cm) and $g(\theta)$ is taken equal to unity, because it is defined as the sin 90°. The λ is the interaction length which depends on the energy of neutrons arriving to the paraffin from the Pb target. For each neutron energy (from thermal up to 1 GeV) the interaction length has been calculated using the formula, which relates the inelastic cross section of neutrons inside the paraffin and their interaction length. A fitting process was applied to the calculated data using an equation similar to the respective one of the secondary particle attenuation as presented by Sullivan [15]. Therefore, the neutrons' interaction length in cm was estimated as follows:

$$\lambda = \lambda_0\left[1-(0.97\pm0.01)\,e^{-(8.6\pm0.7)E}\right], \quad (2)$$

where λ_0 is 84 cm for 1 GeV neutrons crossing the paraffin and E is the neutron energy in GeV.

The theoretical calculation of the normalized total neutron distribution, n/n_{tot}, along the paraffin surface was in agreement with the experimental results (Fig. 4). Although, relationships describing the high-energy interactions, as presented by Sullivan [15], are referred to Fe and Cu targets the agreement of the present study calculation with experimental results demonstrate that the same relationships can be applied also to a Pb target. This can be attributed to almost the same attenuation mean free path for the three targets.

Considering the previously mentioned high-energy protons' interactions with Pb targets there are two competitive effects, which take place inside the Pb target. The exponential decrease of the proton beam intensity along the target (attenuation effect) and the exponential increase of secondary particle production at the head of the target due to the internuclear cascade (buildup effect) [18]. The buildup zone extents mainly up to the proton mean free path in the Pb (about 10 cm for few GeVs proton energy) and becomes deeper at higher energies. According to the specific geometry of the Pb target (20 cm in length) the reason of the gaussian function appeared during the fitting process applied to the neutron distribution along the moderator might be the competition between these effects.

In order to study the spallation neutron distributions along the paraffin surface as a function of the distance x from the beginning of the paraffin a fitting process was applied to the surface density of the total neutrons measured per incident proton (ρ_n, $n\,\mathrm{cm}^{-2}\,\mathrm{proton}^{-1}$). The same procedure was repeated for all proton energies studied. In the fitting process, the experimental data of the first and the last positions, corresponding only to paraffin material have been omitted. The following relation was used:

$$\rho_n = C\,(1-a e^{-bx})\,e^{-dx}, \quad (3)$$

where C is a parameter ranged from 0.3 up to $3.2\,n\,\mathrm{cm}^{-2}\,\mathrm{proton}^{-1}$. The first part of the Eq. (3) was setting to describe a buildup function with α being the buildup parameter and b the buildup coefficient (cm^{-1}). The second part represents the beam attenuation along the target as it was observed at the moderator surface, whereat d is the attenuation coefficient (cm^{-1}). The fitting results of the buildup and attenuation functions are presented in Figs. 5 and 6, respectively. The buildup parameter remains constant with beam energy and it was found to be closed to unity (0.98 ± 0.05). The buildup coefficient increases slightly with proton beam energy (Fig. 5) following an exponential function: $b = (0.026\pm0.003)\cdot\left[1-e^{-(0.21\pm0.04)E_p}\right]$. The attenuation coefficient (Fig. 6) has a linear decreasing behavior when proton beam energy increases: $d = (0.104\pm0.001)-(0.0050\pm0.0003)E_p$.

A higher energy-incoming proton beam induces more secondary particles increasing the spallation neutron multiplicity at the Pb target. This fact can explain the increase of the buildup coefficient. Similar conclusion was also demonstrated by the n_x/n_{tot} plot as function of the distance x along the paraffin surface (Fig. 7). The n_x was

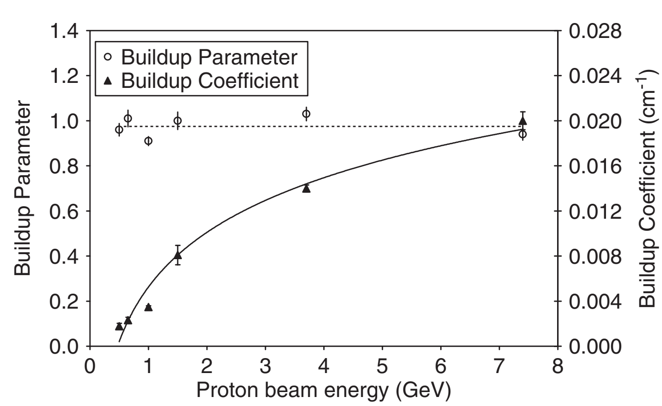

Fig. 5. The buildup parameter and coefficient of the neutron fluence as a function of the proton beam energy. The results were deduced from neutron distributions on the outer paraffin moderator.

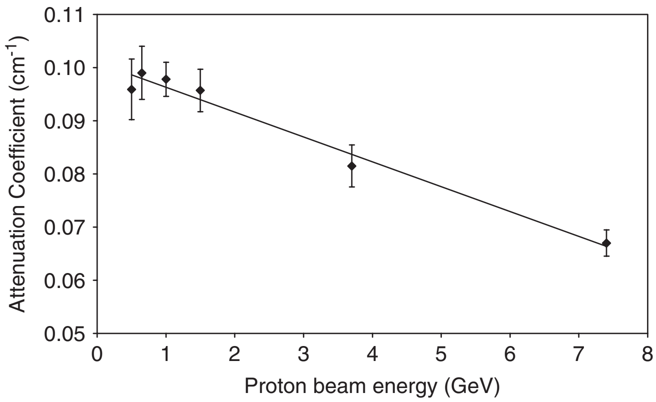

Fig. 6. The attenuation coefficient of neutron fluence as a function of the proton beam energy. The results were deduced from neutron distributions on the outer paraffin moderator.

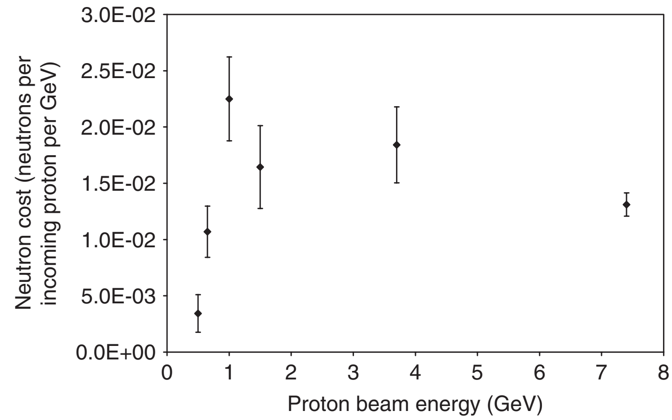

Fig. 8. The neutron production cost corresponding to a Pb target surrounded by 6 cm paraffin moderator as a function of the proton beam energy.

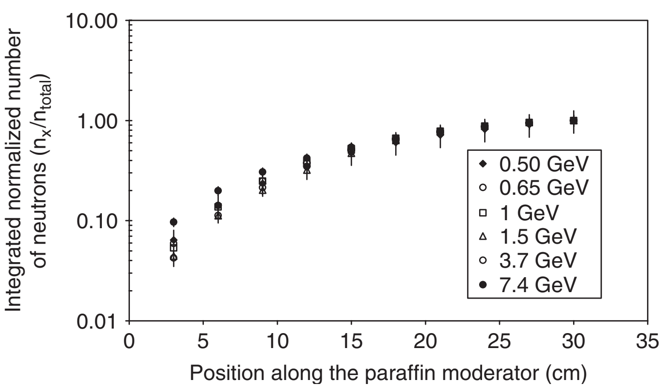

Fig. 7. The integrated number of neutrons measured up to a distance x along the paraffin surface normalized to the total neutrons escaping from the entire paraffin surface.

defined as the integrated number of neutrons measured up to a distance x along the paraffin surface and n_{tot} as the total number of neutron escaping the entire cylindrical surface of the paraffin. For 7.4 GeV protons an increase of spallation neutron multiplicity was found at the target entrance related to a more intense buildup effect occurred at that proton energy than at lower energies. According to the conceptual basis of the calculation, an almost constant neutron multiplication per unit length is expected for the integrated neutron production along the Pb target [2,3]. In order to study the behavior of neutron multiplication, as was presented at the paraffin surface; a fitting process was applied to the integrated neutron distributions (n_x/n_{tot}) as a function of the distance along the paraffin surface (x, cm), using the following relation:

$$n_x/n_{\text{tot}} = A\,x^B. \tag{4}$$

The parameter B was found to be equal to unity (0.99 ± 0.06) and remains almost constant with beam energy. So the behavior of n_x/n_{tot} versus moderator length is proved to be linear with a slope of $0.035 \pm 0.011\,\text{cm}^{-1}$ (mean value) supporting the constant neutron multiplication per unit length. This behavior is very close to that observed in Pb targets without moderator [2,3].

The neutron production cost is defined as the number of produced neutrons per incident proton normalized to the proton beam energy [7,8]. The total neutrons escaping the moderator surface as a function of the proton beam energy can be shown in Fig. 8. According to the experimental results was not clear whether or not the best neutron cost appears at 1 GeV proton beam energy, as it has been shown from other studies [5,7,8] on extended Pb targets without a surrounding moderator. In these experiments the neutron production increases up to 1 GeV indicating better neutron cost. For higher proton beam energies the neutron cost could be considered to be constant or decreasing function of proton beam energy due to the large experimental errors. Another reason could be the complication of the observation of the neutron production in the Pb target through measurements on the paraffin moderator.

4. Conclusions

Intensive research programs were run on the investigation of spallation targets regarding the development of full-scale ADS in the future. Only few studies were performed on the use of the spallation targets with moderator. Such a set up can be proved useful by the fact that the fission chain process relies mainly on reactions induced by thermal neutrons, which are not present in a neutron spectrum produced by a heavy target. Some of the results concerning neutron distributions along the moderator as well as nuclear waste transmutation obtained during experiments performed at the Dubna Synchrophasotron and Nuclotron accelerators over the last years are already reported. This work was a study of the overall results giving an overview of this particular set up. The general conclusion was that the neutron behavior along the moderator surface was very close to that observed in previous studies by others in the target, without moderator.

In addition a calculation based on principles of high-energy physics was applied in order to fit the experimental results. The agreement between calculation and experimental data shows that the moderator acts partially as shielding material around the target.

The study of slow and fast neutron behavior inside the moderator provide evidence that slow neutrons dominate in both directions relative to the beam axis with almost equal fluences while downstream there are more fast neutrons than in the perpendicular direction. Fast neutrons decreased while traveling through the paraffin moderator, indicating that a part of them was thermalized. In the vertical direction more fast neutrons were thermalized than downstream. However, downstream (at 0°) the drop-off was less sharp than in vertical direction (at 90°) due to the hardness of the incoming neutron spectrum from the target at small angles.

The main part of the neutron production derives from proton reactions, which take place inside the first proton mean free path in the Pb target. The effect was observed by the increasing neutron fluence at the beginning of the moderator. However, the secondary particles production competes the beam attenuation. Due to this effect the neutron distribution diminishes over the rest of the target length. The neutron distribution behavior was similar for all proton beam energies. The first-increasing part of the neutron distributions corresponding to a buildup effect was found to be an increasing function of proton energy, while the second-decreasing part of the neutron distributions corresponding to an attenuation effect was found to be a decreasing function of proton beam energy.

The neutron cost for the specific target appeared to be an increasing function of proton beam energy up to 1 GeV and for higher energies can be considered as a constant or decreasing function of energy. These results can only be compared qualitatively with the respective ones corresponding to extend Pb targets without a surrounding moderator.

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