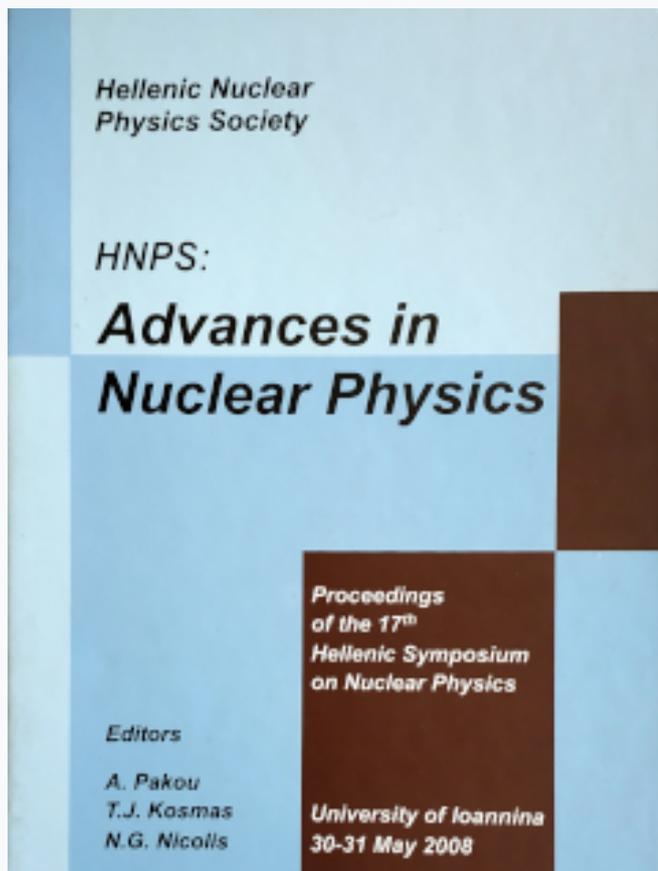


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DOSE MEASUREMENTS AROUND SPALLATION NEUTRON SOURCES

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Abstract

Neutron dose measurements and calculations around spallation sources are of importance for an appropriate shielding study. Two spallation sources, consisted of Pb target, have been irradiated by high-energy proton beams, delivered by the Nuclotron accelerator (JINR), Dubna. Dose measurements of the neutrons produced by the two spallation sources were performed using Solid State Nuclear Track Detectors (SSNTDs). In addition, the neutron dose after polyethylene and concrete was calculated using phenomenological model based on empirical relations applied in high energy Physics. Analytical and experimental neutron benchmark analysis has been performed using the transmission factor. A comparison of experimental results with calculations is given.

1. Introduction

Spallation is an efficient reaction for releasing neutrons from nuclei. In order to sustain spallation reactions an energetic beam of light particles have to be supplied into a heavy target. However during the spallation process neutrons but also protons, photons and other light particles are emitted from the target nucleus. Therefore a spallation facility is quite demanded for transmutation studies because of the high levels of radiation generated by the target. The construction of an appropriate shielding to surround the source is necessary for radiation protection purposes. Several experiments were

performed in order to study the neutron shielding around nuclear reactors^(1,2,3), as well as around high energy accelerators^(4,5). Radiation effects in a spallation environment are different from that commonly encountered in a reactor or accelerator since spallation sources can generate higher neutron densities and harder spectra than nuclear reactors⁽⁶⁾. Hence calculation and measurements of the neutron dose around spallation sources are important.

So far, the neutron spectrum produced by a spallation source has been thoroughly investigated, during the last decades, especially the low energy region $E_n < 5\text{MeV}$ ^(7,8). Such experiments have also been performed in Dubna using a large cylindrical Pb target surrounded by a paraffin moderator or a U-blanket^(9, 10), but dose measurements after shielding are rarely presented. The cost for the radiation shielding contributes to a considerable part of the total financial cost of the spallation source, since massive shields for high energy neutrons, having strong penetrability, are required. The most common materials used as shielding materials are: concrete, iron, polyethylene, paraffin and graphite. The present work studies only polyethylene and concrete as shielding materials. In radiation shielding research, un-charged particles, such as photons and neutrons, are the main radiation to be considered. Calculations were performed in order to design an optimal shielding, taken into account mainly the neutron contribution. The criterion for the appropriate shielding is the dose rate after it, which must be less than $1\mu\text{Sv/h}$ ^(11,12,13,14,15) and the construction materials should be readily available and not expensive. During this study, calculations were performed using phenomenological model based on empirical relations coming from high energy Physics. In addition the neutron doses after shielding materials have been measured for the two different spallation sources. Analytical and experimental neutron benchmark analysis has been performed with the transmission factor.

2. Experimental

This work deals with the neutron dose produced by two different spallation neutron sources consisted of cylindrical Pb target. In the first spallation source, Gamma-2 set-up,

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the Pb target was covered by paraffin moderator and irradiated by 0.65 and 1 GeV protons. The Lead target was cylindrical with 8 cm diameter and 20 cm length and the paraffin moderator that surrounded the target was also cylindrical with thickness of 6 cm. The paraffin was opened from the beam side, (Figure 1a). The specific spallation source intended to moderate the hard neutron spectrum produced by the Pb target. In the next set-up, “Energy plus Transmutation” (E+T), the Pb target was covered by four-sections of natural Uranium blanket and was irradiated by protons with energy from 0.7 up to 2 GeV, (Figure 1b). The construction of this spallation source was made in order to achieve higher multiplication factor than with paraffin moderator and to obtain harder neutron spectrum. For radiation protection reasons, 26 cm of polyethylene surrounded the (E+T) spallation source. In addition a Cd foil, of 1 mm in thickness, was placed around polyethylene from the source side, in order to prevent U-blanket irradiation by low energy back scattered neutrons. Both spallation sources have a simple geometry in order to be used for further benchmark analysis.

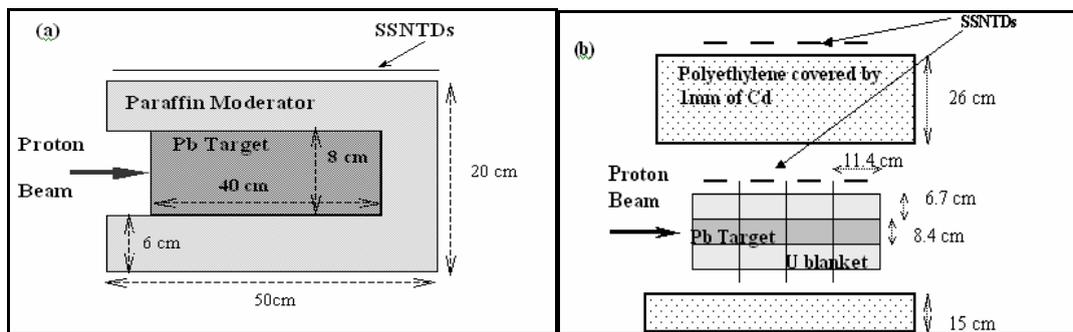


Figure 1. The spallation sources studied in the present work: (a) “Gamma-2” and (b) “Energy plus Transmutation” assembly.

In both experiments, the experimental hall was shielded by 1 m concrete. The spallation sources were positioned in the middle of the experimental hall, about 3 meters from the concrete. The neutron spallation sources were irradiated in Nuclotron accelerator, at the Laboratory of High Energy of Joint Institute for Nuclear Research (JINR), Dubna.

The neutron fluence produced by both spallation sources, as well as the neutron fluence escaping the shielding materials was measured. The measurements were

performed using Solid State Nuclear Track Detectors (SSNTDs). Each set of SSNTDs contains PADC foils (Perschore Mouldings standard grade, PM355) acting as particle detector, (Figure 2). The foils, 250 μ m in thickness, were placed parallel to the target axis after the shielding materials. One part of the detector was in contact with a neutron converter, (Kodak LR115 type 2B, containing $\text{Li}_2\text{B}_4\text{O}_7$). This part of detector provides information about total neutron fluence, detecting the alpha particles' emitted via $^{10}\text{B}(n,\alpha)^7\text{Li}$ and $^6\text{Li}(n,\alpha)^3\text{H}$ reactions. Another part of the detector was in contact with the converter and was covered on both sides with 1 mm Cd foils detecting likewise resonance up to fast neutrons. The thermal-epithermal neutron component (up to about 1 eV) was calculated by subtracting the measured track density of the Cd-covered from the Cd-uncovered region of the detector. Fast neutrons were determined also by proton recoil tracks on the detector itself (neutron elastic scattering on H of the detector)⁽¹⁶⁾. The neutron energy region detected by proton recoils is between 0.3-3 MeV due to limitations in the proton registration efficiency⁽¹⁷⁾. The dosimeters were calibrated in the frame of EURADOS actions for neutron dosimetry^(16,18). The calibration of track number to neutron ambient dose equivalent was performed with monoenergetic neutrons from 144 KeV to 15.3 MeV. Linearity, energy and angular response have been studied. Therefore from the track number using the appropriate conversion coefficient, for the specific energy region, the neutron ambient dose equivalent was estimated^(16,19).

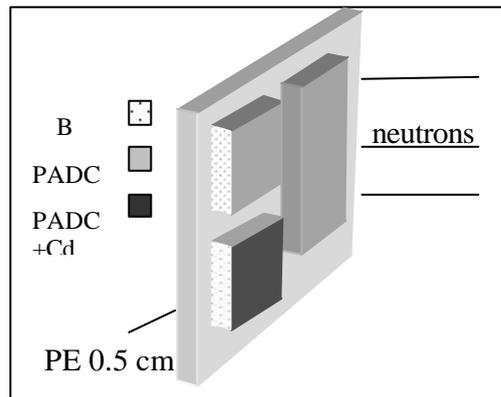


Figure 2. A simple diagram that illustrates the SSNTD geometry.

3. Results and discussion

In order to study neutron dose rates after the shielding the neutron spectrum coming from the spallation sources was converted to the spectrum after shielding using Moyer model. The Moyer model is a point kernel Method which is based on exponential attenuation of the neutron dose equivalent in a thick shield, when neutrons reach the equilibrium state ⁽²⁰⁾. A comparison of calculated doses with measurements was performed.

3.1 Experimental Results

The goal of an efficient shielding design is to attenuate the high radiation intensities, produced by the spallation source, to levels that are in acceptable dose rates after the shielding. In order to select an appropriate shielding to surround a spallation source, the neutron dose produced by the source must be determined. Therefore, SSNTDs were placed along both of the spallation sources, parallel to the target axis above the paraffin and the U-blanket. Thermal-epithermal and intermediate-fast neutrons were measured ^(21,22). The neutron spatial distribution along the target axis for both spallation sources was found to be similar, for each spallation source, for all proton beam energies. For the conversion of neutron fluence to ambient dose equivalent, the corresponding conversion coefficient neutrons to H*(10), to each neutron energy bin, was used ^(16,23). The experimental results are summarized in Tables 1a and 1b. For both spallation sources, the neutron ambient dose equivalent increases with the beam energy, while the main part of this dose is due to the fast neutrons. In table 1 uncertainties indicated for each result comes mainly from track measurements and the conversion coefficient experimentally determined. The same uncertainties were also applying to the experimental results presented in following tables. The total neutron ambient dose equivalent measured at the U-blanket surface of “E+T” set up is higher than the corresponding at the paraffin surface of “Gamma-2” set up. For radiation protection purpose a polyethylene shielding surrounded the “E+T” source. SSNTDs were placed above the polyethylene moderator that covers the U-blanket ⁽²⁴⁾ in direction parallel to the

target axis, fig 1b. The polyethylene moderator reduces the neutron ambient dose equivalent measured at the U blanket surface 60 times for 0.7 GeV and about 30 times for 2 GeV.

Table 1a. The neutron ambient dose equivalent (Sv) measurements on the “Gamma-2” surface during the total irradiation

| Proton Energy (GeV) | Thermal- Epithermal (Sv) | Intermediate- Fast (Sv) |
|------------------------|--------------------------------|----------------------------|
| 0.65 | 0.22 ± 0.02 | 5 ± 1.5 |
| 1 | 0.61 ± 0.03 | 8 ± 3 |

Table 1b. The neutron ambient dose equivalent (Sv) measurements on U-blanket surface during the total irradiation

| Proton Energy (GeV) | Thermal- Epithermal (Sv) | Intermediate- Fast (Sv) |
|---------------------------|-----------------------------|----------------------------|
| 0.7 | 0.11 ± 0.01 | 12 ± 1.5 |
| 1 | 0.16 ± 0.03 | 17 ± 4 |
| 1.5 | 0.25 ± 0.04 | 28 ± 3 |
| 2 | 0.32 ± 0.09 | 38 ± 4 |

The neutron spectrum produced by “E+T” set-up was calculated using MCNPX and DCM/DEM codes ^(25,26). According to the Monte Carlo calculation of neutron spectrum, in which all neutrons were taken in account (including neutrons above 3 MeV), the moderator diminishes the neutron dose about 100 times for 1 GeV proton beam. The main part of the total dose after polyethylene comes from fast neutrons, (Table 2). The comparison of neutron doses produced by the two sources show that Gamma 2 set up gives lower doses than E+T set up. However, after polyethylene moderator E+T set up produced smaller doses than Gamma-2 set-up. The most important for radiation protection purposes is the neutron dose after experimental hall shielding which is composed by concrete iron enriched (heavy concrete). The neutron dose measured after the concrete for “E+T” source is found to be less than the lower detection limit of the detector (10^5 tracks per cm^2) that corresponds to 1.5 μSv for thermal neutrons and about 30 μSv for fast neutrons. These results are summarized in Table 3. As it is presented in the table, the neutron ambient dose equivalent after the concrete, for “Gamma-2” set-up,

is higher than the respective neutron ambient dose equivalent for “E+T” set-up. The main part of dose for “Gamma-2” set-up is due to the thermal-epithermal neutrons. For E+T set-up the main part of the neutron dose derives from intermediate-fast neutrons and have the tendency to meet radiation protection standards. The total neutron ambient dose equivalent for the case of Gamma-2 set up remains still higher from radiation protection standards. In case of E+T set up the level of neutron ambient dose equivalent can not be compared to radiation protection standards because of limitation of detector measurements.

Table 2. Neutron ambient dose equivalent measurements (mSv), during the total irradiation, after 26 cm polyethylene and 1mm Cd of “E+T” spallation source

| Proton Energy (GeV) | Thermal-Epithermal (mSv) | Intermediate-Fast (mSv) |
|---------------------|--------------------------|-------------------------|
| 0.7 | 2.86 ± 0.26 | 204 ± 25 |
| 1 | 4.16 ± 0.78 | 289 ± 68 |
| 1.5 | 21.5 ± 3.4 | 812 ± 87 |
| 2 | 41.6 ± 11.7 | 1370 ± 144 |

Table 3. Neutron ambient dose equivalent measurements, during the total irradiation, after the concrete for Gamma 2 and E+T set ups.

| Proton Energy (GeV) | Thermal-Epithermal (μ Sv) | Intermediate-Fast (μ Sv) |
|------------------------------------|--------------------------------|-------------------------------|
| <i>“Gamma-2” Spallation source</i> | | |
| 0.65 | 298 ± 46.5 | < 30 |
| 1 | 411 ± 114 | < 30 |
| <i>“?+?” Spallation source</i> | | |
| 1.5 GeV | < 1.5 | < 30 |

3.2 Calculations

An analytical calculation of the neutron ambient dose equivalent was made for both spallation sources. Two assumptions are often made in shielding calculations for thin target sources. The first assumption is that the source can be approximated by a point source. For this assumption, the source must be localised in a geometrical volume small

in size compared with the other dimensions of the shielding. The second assumption is that the dose D , as a function of the source position, can be described in terms of the relative coordinates of the point source with the point of interest and that there is no contribution from any other secondary sources. The above assumption represents a pure point source/ line-of-sight model. Such a model is directly applicable to the shielding of low energy proton accelerator and has been extended to proton energies in GeV range by Moyer. In the current work, the Moyer model was applied in low energy neutrons, in order to be used for dose calculations after the shielding surrounded spallation sources with thick targets. The point kernel method, named as Moyer model, is based on exponential attenuation of neutron dose equivalent for neutrons, when they reach the equilibrium state after thick shield ⁽²⁰⁾, using a single built-up factor and an attenuation length. According to this model the dose equivalent at the point of interest can be estimated using the following phenomenological equation ⁽²⁷⁾:

$$H(x, \mathbf{q}) = \frac{H_0(\mathbf{q})}{r^2} \exp\left[-\frac{x}{g(\mathbf{q})l}\right] \quad (1)$$

Where $H_0(?)$ is taken as $H_0(90^\circ)$, that represents the dose equivalent from the number of neutrons crossing at 90° the source surface. The calculation was made only for 90° because the maximum of detector's efficiency is at 90° while in the intermediate angles between 90° and 0° it drops according the law of $1/\cos^2?$, as it happens for every flat detector. The r corresponds to the distance between the source and the point of interest, x is the depth inside the shielding, $g(?)$ is defined as $\sin?$ for lateral shielding and $?$ is the interaction length. However, for lower energies, the interaction length depends on the neutron energy and the simple Moyer model is no longer applicable. In order to use Moyer model for low energy neutrons, the interaction length of neutrons has to be estimated for a shielding material, in each neutron energy range. During this study, the interaction length of neutrons for each energy bin has been calculated using the relationship between the interaction length and the inelastic cross section ⁽²⁷⁾. Using the same relationship the mean free path of neutrons can also be estimated. After the estimations of interaction length and mean free path of neutrons, for each neutron energy

range, the neutron spectrum after the shielding can be calculated, taken into account the lethargy of neutrons in a shielding material, using the equation 1. In order to calculate the neutron spectrum after the shielding material the neutron spectrum produced by the spallation sources was taken from the calculations made using the Monte Carlo DCM/DEM code⁽²⁵⁾. The calculated neutron spectrum corresponds to a proton beam up to 10^{13} protons. In all calculations the statistical errors range between 3-6 % ⁽²⁶⁾. The neutron ambient dose equivalent was estimated taken into account the dose equivalent factor $H^*(10)$ ⁽²³⁾ for each energy point of the calculation. Shielding (or moderator) materials, such as polyethylene and concrete, were studied and the obtained results are presented in Table 4.

Table 4. Calculated neutron ambient dose equivalent after the shielding surrounded both spallation sources

| Proton Energy (GeV) | Thermal-Epithermal | Intermediate-Fast |
|---|--------------------|-------------------|
| <i>After Concrete, "Gamma-2" source</i> | | |
| 1 GeV | 375 μ Sv | 20.1 μ Sv |
| <i>After Concrete, "E+T" source</i> | | |
| 1.5 GeV | 1.26 μ Sv | 26.1 μ Sv |
| <i>After Polyethylene, "E+T" source</i> | | |
| 1.5 GeV | 34,3 mSv | 880 mSv |

3.3 Comparison between measurements and calculations

Regarding the results presented in Tables 3 and 4 the calculation can satisfactorily describe the experimental results. For the comparison of the above calculations with the experimental results, the transmission factor of neutrons after the shielding was estimated. The transmission factor defined as the ratio of the neutron ambient dose equivalent values with and without shield. In Table 5, the transmission factor of neutrons after polyethylene, obtained by calculation and experimental results for both thermal-epithermal and intermediate-fast neutron component is presented. According to Table 5, the calculations converge to the experimental results. These analytical calculations based on Moyer model indicate that this model can be applied for the estimation of the neutron dose after a shielding. However, the deviations observed between experiment and

calculation can be attributed to the assumptions made for the application of the model i.e. the spallation sources can not be considered as a point source because they have significant dimensions.

Table 5. Transmission factor of neutrons (%)

| After Polyethylene the "E+T" source | | |
|-------------------------------------|--------------------|--------------------|
| <i>Neutron Energy range</i> | <i>Calculation</i> | <i>Measurement</i> |
| <i>Thermal-Epithermal</i> | 8.4 | 8.6 ± 0.3 |
| <i>Intermediate-Fast</i> | 2.6 | 2.9 ± 0.8 |
| After Concrete, "Gamma-2" source | | |
| <i>Neutron Energy range</i> | <i>Calculation</i> | <i>Measurement</i> |
| <i>Thermal-Epithermal</i> | 0.25 | 0.18 ± 0.03 |

Conclusion

The main objective of the present work was to determine experimentally and by calculation the ambient dose equivalent induced by neutrons that are produced by two different spallation sources, consisted of Pb target. From tables 1a and 1b, it is deduced that E+T set up gives higher neutron dose comparing to the Gamma-2 set up for the same proton beam energies. This effect is due to the higher fast neutron production from E+T set up compared to the spectrum corresponding to Gamma-2 set up. Gamma-2 spectrum has in addition thermal-epithermal neutrons but the total dose remain higher in the case of E+T set up, in which the thermal neutron contribution (thermal-1eV) is negligible. As it was experimentally confirmed, in U-blanket surface some thermal neutrons come from neutron back scattering in the polyethylene shielding and the fluence was measured to be of the order of 10^{-5} cm^{-2} per proton incident on the target. The polyethylene shielding diminishes the total neutron ambient dose equivalent of E+T set up about 100 times. However a part of those neutrons are shifted to the thermal – epithermal neutron range. Their contribution to the total neutron ambient dose equivalent after polyethylene is about 100 times lower than fast neutron dose. From these neutrons the ratio intermediate-fast/thermal-epithermal is about 70 for 0.7 GeV and about 30 for 2 GeV. The

polyethylene shielding can finally lower the dose of E+T set up below the corresponding to the Gamma-2 set up.

For both set ups, after concrete fast neutron component is below the detection limit of the PADC, which is 10^5 tracks per cm^2 for fast neutrons. The dose coming from thermal –epithermal neutrons is still higher for Gamma-2 set up than in E+T set up. Concerning the comparison of experimental results with calculations for the neutron ambient dose equivalent after polyethylene shielding (table 5), a satisfactory agreement is concluded from the transmission factor for both thermal-epithermal and intermediate-fast neutrons. For fast neutrons the same comparison after concrete is not feasible because their number is below detection limit.

The agreement of experimental results with analytical calculations based on Moyer model, demonstrates that the application of the model can be employed for low energy neutrons without significant deviations. The results also indicate that the model applied for thin targets could be used for thick targets with large dimensions comparing to a point source with a deviation of 3-30%. The large deviation is due to the small track number measured, which induce large experimental uncertainties.

In order to compare the ambient dose equivalent with radiation protection standards after concrete, the ambient dose equivalent was converted to dose rate. For that reason the duration of the irradiations (to complete about 10^{13} beam protons) was considered in order to estimate the neutron dose rates. The total neutron ambient equivalent dose rate produced by both spallation sources was calculated by applying appropriate conversion factors to the data obtained using MCNPX and DCM/DEM codes. According to the last radiation protection recommendations of 2007, the effective dose limits in planned occupational exposure must be less than 20 mSv/year, average over defined periods of 5 years⁽¹⁴⁾. The commission has concluded that the existing dose limit recommended by ICRP60 continues to provide an appropriate level of protection⁽¹¹⁾. For this work, the ambient dose equivalent has been used instead of effective dose, taken into account that for the above recommended effective dose limit the total tissue weighting factor is 1 ($\sum w_t = 1$). The tissue weighting factor for a uniform irradiation of a body can be taken equal to 1⁽¹¹⁾. An additional constrain of $1\mu\text{Sv/h}$ neutron ambient dose equivalent for workers is an optimum lower limit. The total neutron ambient equivalent

dose rate after concrete in the described experiments was found to be higher than the maximal allowed effective dose in personal dosimetry determined by ICRP^(11,12,13,14). For Gamma-2 set up a dose rate is 37 $\mu\text{Sv/h}$ (34 $\mu\text{Sv/h}$ from thermal-epithermal and 2.5 $\mu\text{Sv/h}$ from fast neutrons) for 1 GeV protons. For E+T set up, 11 $\mu\text{Sv/h}$ were measured (about 0.5 $\mu\text{Sv/h}$ from thermal-epithermal and 10 $\mu\text{Sv/h}$ from fast neutrons) for 1.5 GeV protons. These results demonstrate that an additional shielding has to be calculated for spallation sources. The most practical and cost effective solution is to add iron (of about 40 cm) from the experimental hall side taken into account the neutron's calculations, coming from Monte Carlo results.

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