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Neutron yields from massive lead and uranium targets irradiated with relativistic protons

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Abstract

Long-lived isotopes can be transmuted into stable or short-lived elements either by neutron captures or neutron induced fission. The need of a large excess of neutrons has led to the use of accelerator driven sources (ADS). A series of experiments were carried out at the Synchrophasotron/Nuclotron of the Joint Institute for Nuclear Research (JINR) Dubna, using protons of 1.0 GeV. Solid Lead and Uranium targets surrounded by paraffin moderator were irradiated. On the outer surface of the moderator a number of Solid State Track Detectors were placed to monitor neutron spatial distribution. The results showed that the maximum neutron production was reached within the range of one to two proton mean free paths in the target. Then decreasing neutron production follows the proton beam attenuation along the target. Moreover, the results showed both targets neutron production evolution along the target, to be the same. However, neutron flux per incident proton is depended on the target mass, which was found to be higher for the heavier target.

Keywords: Spallation sources; Accelerator driven systems; Neutron detectors

1. Introduction

To transmute long-lived isotopes into stable or short-lived elements neutron capture or neutron induced fission seems to be the more efficient way (GSI-NACHRICHTEN, 1999). Although both processes occur in a reactor type neutron flux the need for a large excess of neutrons has led to accelerator driven systems (ADS). At those systems high-energy light particles, in the GeV range, hitting a heavy target produces a large number of protons and neutrons. Such sources, called spallation sources, are used now in order to obtain high neutron fluxes with a wide energy spectrum from thermal to (theoretically) the proton beam energy. The first neutron generation is coming from the target nucleus, which after interaction with the light particles is left in an excited state and decays by emitting mainly protons and neutrons. Some of the nucleons ejected during the first reaction step have still sufficient energy to induce further spallation reactions, leading to a multiplication of the emitted particles. The actual understanding of such a source needs systematic

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knowledge of their operation. In particular, the optimization of the target—moderator assembly requires detailed information on (a) the number of spallation neutrons produced per incident particle for various target materials and geometry and (b) the energy spectrum and angular distribution of spallation neutrons, which are necessary for the design of the source. Especially the neutron spectrum is a decisive factor for the source efficiency for transmutation or incineration experiments. This means that the neutron number at the resonance energy range for (n, γ) as well as fast neutron range for (n, f) and (n, xn) reactions must be determined.

Last decades a number of experiments have been performed concerning the physics involved on spallation sources. Proton beams at the GeVs carried out most of those experiments. The beam energy, target material and neutron spectrum were examined in order to investigate the source performance and optimization (Pienkowski et al., 1997; Hilscher et al., 1998; Letourneau et al., 2000). Some of those experiments have run in Synchrophasotron/Nuclotron accelerators of the High Energy Laboratory (JINR) Dubna by our group. Solid Pb and composite Pb–U targets have been irradiated by protons in the energy interval of 0.5–7.4 GeV (Wan et al., 1998; Adam et al., 2002; Zamani et al., 2003). At those experiments moderator, for neutron spectrum softening surrounded the target. The target length was 20, 40 and 50 cm.

Solid State Nuclear Track Detectors were used in all experiments for neutron flux measurements and to study spatial distribution around each specific source. This work deals with neutron production distributions on the moderator surface for various target-moderator systems used as spallation sources. The results are referred to target irradiation with protons of 1 GeV.

2. Experimental arrangement

Proton beam delivered from Synchrophasotron/Nuclotron accelerators has an intensity of 10^{11} or 10^{13} depending on the set-up used for neutron detection. The beam was focused axially to the cylindrical target. The Pb target consists from Pb disks of 8 cm in diameter giving a final cylindrical form of 20 and 40 cm in length. The target was surrounded by a cylindrical paraffin moderator 6 cm in thickness, which was open from the beam side, Fig. 1. The simplest set up is shown in Fig. 1a. A complex target is presented in Fig. 1b consisting of a U core covered by Pb target. The whole system was enveloped in the paraffin moderator. A cylindrical Pb core was also used in the case of Fig. 1c. Additional natural U rods of 3.6 cm in diameter and 10.6 cm in length compose the target (U blanket). The set up was constructed by four similar sections with a final length of about 50 cm (Zamani et al., 2003).

This study is a comparison of the neutrons escaping each target-moderator system. Neutron distributions along the moderator surface were obtained by CR-39 Detectors positioned parallel to the axis of the cylindrical target. Two types of detector systems were used for the neutron detection. The one CR-39 detector is in contact with $Li_2B_4O_7$ converter (from Kodak LR 115 type 2B) CR-39 detects alpha particles from $^{10}B(n, \alpha)^7Li$ reaction. Half detector surface was covered by 1 mm Cd. This system is able to give information about thermal–epithermal and intermediate–fast neutron fluxes. An additional CR-39 foil is used to detect fast neutrons by proton elastic scattering from the detector itself. Proton recoils from fast neutrons in the range of 0.3–3.0 MeV can be detected (due to limitations of the detector registration properties). The responses used for the conversion tracks to neutrons were taken by Stoulos et al. (2004). Along paraffin moderator the detectors were positioned every 1.5 cm along the axis of the cylinder. In the case of the U-blanket surface, one detector system is placed in the middle of each section. A second SSNTD set up was applied to detect fission reactions induced by neutrons. For this purpose Mackrofol detector was used as detector. The fissile targets were ^{235}U and ^{232}Th. These targets detected thermal, < 1 eV, and fast neutrons, above 2 MeV respectively. The conversion of fission number to neutron flux was obtained by calculating an effective cross-section for each special experimental arrangement (Stoulos et al., 2004). Target masses were of the order of 100–300 $\mu g/cm^2$. For neutron flux estimation other methods were also used, mainly activation detectors. In Fig. 1a,b for example ^{139}La sensors can be seen. The results are given in Adam et al. (2002) and are in good agreement with results obtained by SSNTDs.

3. Results and discussion

Considering symmetry around the cylindrical set up neutron distributions are given for the upper surface of the cylindrical set-ups in Fig. 2a,b, for targets of 50 cm in length. In Fig. 2a thermal–epithermal neutrons are presented while in 2b intermediate–fast neutrons for the same positions. The shapes of the distributions are quite similar and can be described by an increase at the beginning of the target. A maximum corresponding to about a mean free path of proton interaction with Pb target is observed. This part of the neutron distributions reflects the number of the neutrons emitted in the primary spallation reactions. A small fraction of the primary spallation products (mainly protons and neutrons) can induce secondary reactions. Then, deeper into the target, beam attenuation and secondary production give as final result a decreasing behaviour as it is seen in Fig. 2a,b. However in this image we must take into account the neutron thermalization travelling the moderator. Nevertheless, the shape of neutron distributions along the moderator surface is similar to those observed in the target (Carpenter et al., 1999). The characteristic of those distributions is a build up effect upstream to a maximum and a drop when going to downstream, for all the targets have been investigated. The results of all targets studied the total neutron production is

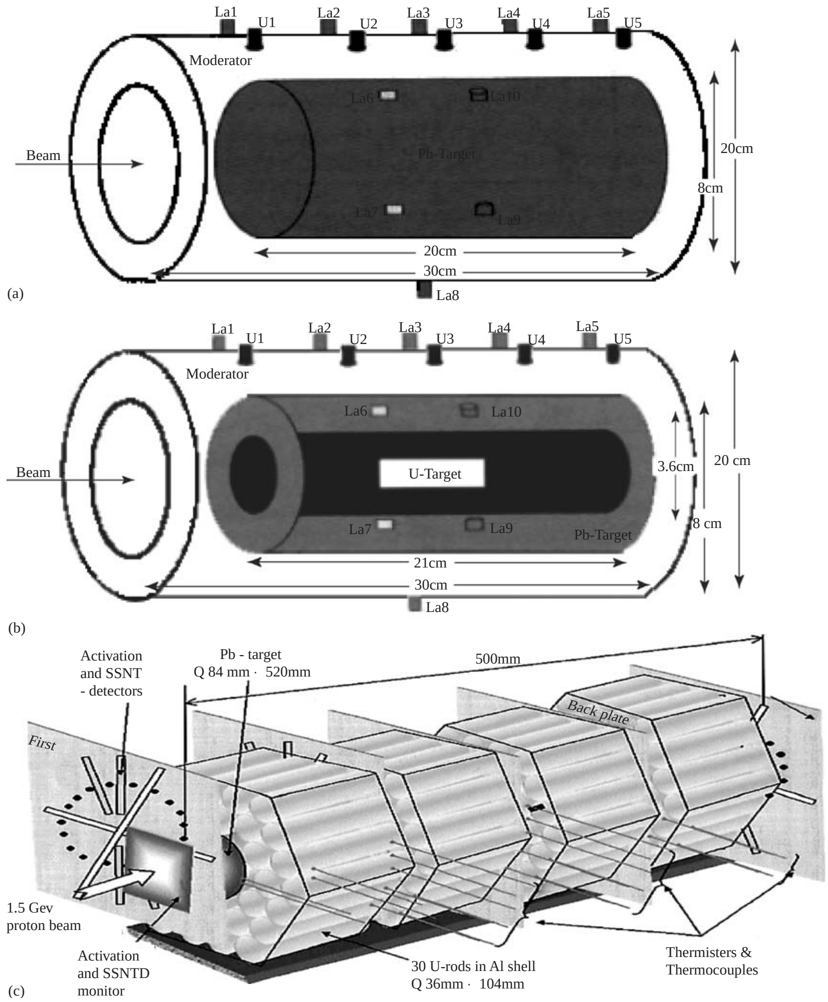

Fig. 1. Different set-ups studied as spallation sources: (a) Pb target with paraffin moderator, (b) Composite natural U and Pb target with paraffin moderator, (c) Pb target enveloped by natural U rods (U blanket).

presented in Table 1 for the comparison. Each neutron component is given also in Table 1. The data were taken by integration of the fitted neutron distribution along the length of each moderator surface. The results are presented as the neutron number per cm along the upper moderator surface and per incident proton. The indicated length is the moderator length.

For the same target and moderator the longer target produces more neutrons (Fragopoulou et al., 2005). From columns 1 and 2 of Table 1 is concluded that duplication in target length result to about three times more neutrons. It is explained by the lower neutron production in the smaller target account taken that the target length is smaller than proton range in the target. The fraction of backscattered neutrons enhances also the neutron number at the moderator surface in the case of the longer target. In fact the neutrons of the thermal–epithermal region are only be doubled but those in intermediate-fast region are tripled giving higher contribution to the total neutrons produced at the moderator surface. This observation indicates that longer targets need thicker moderators in order to obtain more neutrons in the thermal–epithermal energy range.

The use of Uranium blanket results also to increasing contribution of intermediate–fast neutrons while thermal–epithermal remains almost constant. A separate study with ^{235}U fissions was performed in order to estimate thermal neutrons. Their spatial distribution is presented in Fig. 2a. From Table 1 is concluded that their contribution is very low so this set-up gives mainly epithermal component. Fast component ($E_n > 2\,\mathrm{MeV}$) in U blanket case was also studied via ^{232}Th fissions, Fig. 2b. Their contribution is relatively low comparing to those estimated in

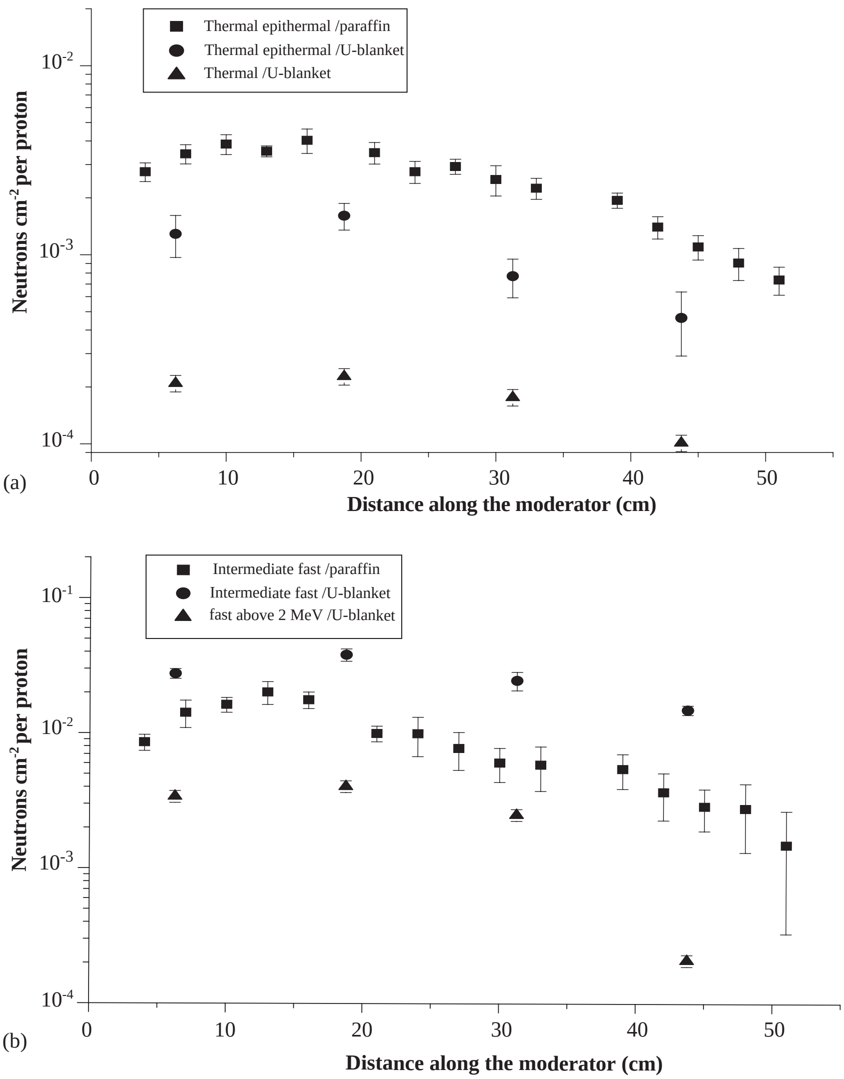

Fig. 2. Spatial neutron distribution along the moderator surface: (a) thermal–epithermal and (b) intermediate–fast neutrons for Pb target and Pb target plus U blanket (50 cm). In (a) thermal neutrons from ^{235}U fissions and (b) fast neutrons from ^{232}Th fissions are also seen.

Table 1
Neutrons/proton production from the moderator surface for various spallation set-ups with Pb target

Neutron energy range	Pb target + paraffin moderator (30 cm) (%)	Pb target + paraffin moderator (50 cm) (%)	U(Pb) target + paraffin moderator (30 cm) (%)	Pb target + U blanket (50 cm) (%)
Thermal (^{235}U fission)				0.6 ± 21[a]
Thermal–epithermal	5.0 ± 24	8.2 ± 18	6.3 ± 30	7.5 ± 19
Intermediate–fast	11.3 ± 17	29.5 ± 18	53.4 ± 30	82.9 ± 20
Total	15.7 ± 17	38.3 ± 18	59.0 ± 30	91.1 ± 19
Fast (^{232}Th fission)				8.8 ± 19[b]

[a]Correspond to thermal neutrons up to 1 eV.
[b]Correspond to fast neutrons above 2 MeV.

0.3–3.0 MeV interval, which is found to be 10 times more (Table 1). Comparing to the total neutron production of the Pb target, of the same length, 50 cm, it is concluded that by U blanket the total neutron number increases more than two times. Concerning the neutron spectrum it is concluded that contains more intermediate–fast neutrons than in case of paraffin moderator. One can obtain similar results by the use of the composite U–Pb target (Fig. 1b). The total neutrons produced from this target, measured at the U-blanket surface, are more than three times higher relative to those produced by the same Pb target length and paraffin dimensions. Paraffin moderator should be thicker in order to equilibrate fast neutron production originating mainly from the heavier target. Intermediate–fast neutrons are about four times more than in a single Pb target of the same dimensions while thermal–epithermal neutron contribution is approximately the same.

4. Conclusions

Spallation reactions taking place in accelerator driven systems may offer a solution to the nuclear waste problem. The methods proposed are: (a) incineration (nuclear fission following neutron capture) and (b) transmutation (by transformation of fission products after neutron capture). Both reactions can be performed under a reaction rate $R \sim \sigma.\Phi$, where σ is the cross-section of the radionuclide to a neutron energy interval and Φ the neutron flux at the same energy range. Most neutron capture reactions have high capture cross-sections at the resonance energy region while some fission reactions need thermal (< 1 eV) or fast neutrons (> 1 MeV). From this study is concluded that increasing the length of a heavy target rather fast neutrons are increased. Further increase in length result to limited neutron flux enhancement. On the other hand by using heavier target materials actually more fast neutrons are produced favoring (n, f) and (n, xn) reactions. In agreement with previous works, a heavier than Pb target of about 40 cm in combination with a light moderator of appropriate thickness can increase also thermal–epithermal neutrons for (n, γ) reactions. So these results could be a pathway for future spallation source constructions.

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