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ACCELERATOR DRIVEN SYSTEM BASED ON A COMBINED PB/U-BLANKET SPALLATION SOURCE

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ABSTRACT
Spallation reactions of a relativistic proton beam in a solid target are able to produce intense neutron fluxes. Such sub-critical Accelerator Driven System can be used for transmutation or incinerations of long-lived radioactive waste by neutron capture or neutron induced fission. A combined Pb/U target irradiated as the spallation source by relativistic proton beams from 0.7 up to 2 GeV. Slow, intermediate and fast neutron detection was performed by several types of passive detectors i.e. SSNTDs (as particle and fission detectors) and activation detectors (^{238}U, ^{nat}Au, ^{nat}Cd). Neutron spatial distribution along the U-blanket is presented in order to estimate slow and fast neutron multiplicity that is essential to study the transmutation and incineration efficiency of the specific spallation source. To evaluate the tolerance of any structural material in source design and construction, except the fast neutron component, the secondary proton distribution was measured using ^{nat}Cd activation technique. Moreover, the effectiveness of a polyethylene shielding to reduce the elevated escaping rate of the secondary neutrons and shift them to energies useful for transmutation reactions is also discussed.

KEY WORDS
Spallation sources, Neutron and proton detection, Transmutation

1. Introduction

To transmute long-lived isotopes into stable or short-lived elements neutron capture or neutron induced fission reactions seems to be the more efficient way. Although both processes occur also in a reactor neutron_spectrum the necessity for a harder neutron spectrum has led to Accelerator Driven Systems [1-6]. At those systems high-energy light particles, such as a proton beam in the GeV range, hitting a heavy target produces a large number of secondary protons and neutrons. Such sources, called spallation sources, coupled with moderators are used in order to obtain high neutron fluxes with a wide energy spectrum from thermal up to (theoretically) the proton beam energy. The first neutron generation is coming from the target nucleus (intra-nuclear cascade), which after interaction with the light particles is left in an excited state and de-excitate by emitting mainly protons and neutrons (evaporation stage). Some of the nucleons ejected during the first reaction step have still sufficient energy to induce further spallation reactions (inter-nuclear cascade), leading to a multiplication of the emitted particles. To favor the neutron multiplicity a lead target can be used as neutrons in the lead have ~~a~~ small average lethargy $\xi \approx 0.01$, a high and nearly energy-independent elastic scattering cross section and it is almost transparent to energies below 1 keV. The energy cost of neutron production is around to 20-25 MeV/n [2]. Fast neutrons produced by spallation, after moderation into the target by (n,xn) and (n,n') reactions reach the intermediate energy range and finally a fraction of the intermediate-fast neutrons enter in the resonance energy range by small iso-lethargic steps. When additional material, such as U, cover the Pb target results to further neutron multiplication due to its high cross section in fast neutron induced spallation and/or fission reactions. On this basis, a subcritical Pb/U-blanket electronuclear set-up has been build at the Laboratory of High Energies, JINR, Dubna by the motivation to perform experiments on increasing safety nuclear power engineering and transmutation of radioactive waste ("Energy plus Transmutation" project) [7].

A detailed engineering design of an ADSystem requires an optimization of its performance in terms of neutron production, and an assessment of problems occurring in such systems. 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. Especially the neutron spectrum is a decisive factor for the source efficiency for transmutation or incineration experiments. Thus, 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. During the spallation process, in addition to neutron production, a large amount of secondary charged hadrons, mainly protons, and many gamma rays are also emitted from the target. In order to cope with the intense radiation field around a spallation source, an effective shielding to energetic hadrons and photons is required. Moreover, the structural materials of the source have to be chosen so as to tolerate not only fast secondary neutrons but also the secondary protons as well as protons generated by (n,xp) or (p,xp) reactions and slowed down inside the material [8-11].

During the last decades, several spallation sources for solid-state and material physics have been constructed and studied worldwide. Many experimental results as well as Monte Carlo simulations about spallation physics and neutron multiplicity have been presented [12-18]. In addition, various spallation sources for transmutation or incineration experiments have already been tested and they need continued consideration [2-7]. Among these is the GAMMA-2 set up, a Pb target surrounded by a 6 cm thick paraffin moderator, that has already been studied and the recent set-up “Energy plus Transmutation” with a Pb target surrounded by a ~7 cm thick U-blanket. The ^{239}Pu(n,f), ^{238}Pu(n,f), ^{238}U(n,γ), ^{237}Np(n,γ), ^{129}I(n,γ) transmutation effectiveness has been studied in irradiations using relativistic proton beams at the above setups [19-24]. Our contribution to these experiments was to determine the secondary neutron and proton fluence by using supplementary passive methods in order to cover the wide energy emitted hadron spectrum.

In the present work, the secondary neutron and proton production from the combined Pb/U-blanket target is discussed. Slow, intermediate and fast neutron detection was performed by several types of passive detectors i.e. SSNTDs (as particle and fission detectors) and activation detectors (^{238}U, ^{nat}Au, ^{nat}Cd). Their spatial distribution along the U-blanket is presented, which is then used to estimate the slow and fast neutron multiplicity that is essential to verify the transmutation and incineration efficiency of the specific spallation source. In order to evaluate the tolerance of any structural material in source design-construction, except the fast neutron component, the secondary proton distribution was measured using ^{nat}Cd activation technique. Moreover, the effectiveness of a polyethylene shielding to reduce the elevated escaping rate of the secondary neutrons and shift their energies to lower range, useful for transmutation reactions, is also discussed.

2. Instrumentation

The “Energy plus Transmutation” set-up is a spallation source where the combined Pb/U-blanket target consists of a cylindrical Pb core of 8.4 cm in diameter surrounded by natural U rods of 3.6 cm in diameter and 10.4 cm in length (U blanket) which form a hexagon around the target. The set-up is constructed using four similar sections of the combined target with about 0.8 cm gap between them and total length 50 cm (Fig. 1). The “E+T” set-up is surrounded by an external shielding for radiation protection reasons as shown in Figure 2. The shielding consists of a wood container filled with granulated polyethylene. The walls from all sides are plated with sheets of cadmium. High-energy neutrons are slowed down within the polyethylene. The 1 mm thick Cd-absorbers located at the inner walls of the container reduce significantly the backscattering of thermal neutrons into the target and detectors volume [7].

Proton beams with energy 0.7, 1, 1.5 and 2 GeV delivered from Nuclotron accelerator at Laboratory of High Energies, JINR Dubna. The integrated intensity of the beam was 10^{11} and/or 10^{13} protons depending on the type of the detectors used for neutron determination. The accurate position monitoring of the proton beam together with the determination of the integral proton fluence was carried out during the entire run using standard aluminum and copper foils as activation detectors. The Al-monitor reaction ^{27}Al(p,3pn)^{24}Na as well as the Cu-monitor (p,x) reactions were used to determine the proton fluence [25-26].

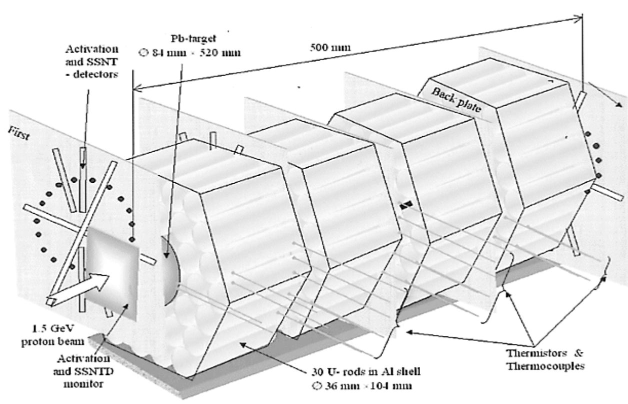

Figure 1. The “Energy plus Transmutation” set-up.

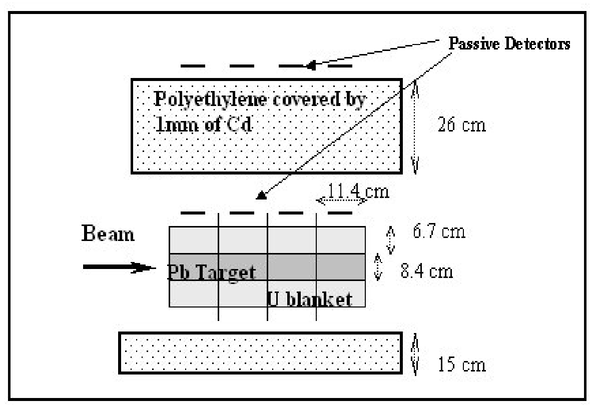

Figure 2. A cross-section of the Pb/U-blanket assembly.

Neutron fluence measurements were performed on the surface of U blanket as well as on top of the polyethylene shielding, using various passive detectors as indicated in Figure 2. On the surface of each section of the U-blanket a set of SSNTDs plus a set of activation detectors (^{238}U, ^{nat}Au, ^{nat}Cd) were positioned parallel to the target axis. In addition, on the top of the shielding at three different positions along the target axis, five sets of SSNT Detectors (totally 15 sets) were placed on each position vertically to the target axis. These detectors were used to measure neutrons escaping from the setup. The SSNTDs were employed as particle detectors with appropriate moderators and also as fission detectors with fissile targets. The advantage of SSNTDs is that they are insensitive to gamma rays, which are significant around spallation sources.

Each set of SSNTDs including three different detection areas, consisted of a CR-39 foil on polyethylene, half-covered with $^{6}Li_2B_4O_7$ converter material. The area with the $^{6}Li_2B_4O_7$ converter was partially covered on both sides with 1 mm of Cd. Such detection system can simultaneously detect thermal and intermediate-fast neutrons [27]. The track density difference between the Cd-covered and uncovered CR-39 foil plus $^{6}Li_2B_4O_7$ converter corresponds to the thermal neutron fluence due to (n,α)-processes induced by neutrons in $^{10}B(n,\alpha)^{7}Li$ and $^{6}Li(n,\alpha)^{3}H$ reactions. The track density on the completely uncovered CR-39 foil originating by recoil proton gives indirect information about intermediate-fast neutrons in the energy range of $0.3 < E_n < 3$ MeV, which is the range with about constant response of CR-39 to protons [28]. Another SSNTD set up was applied to detect fission reactions induced by neutrons using for this purpose Lexan as SSNTDetector. Fissile targets such as ^{235}U for thermal neutron (<1 eV) and ^{232}Th for fast neutron (> 2 MeV) detection were used having mass of around 100 and 300 μg respectively [29]. The applied SSNTDs as particle detectors irradiated with 10^{11} protons while the fission detectors irradiated with 10^{13} protons in order to achive a well-measured track density. After their irradiation, the etching process and track-scanning under an optical microscope were carried out.

The secondary neutron distribution along the target was determined using also activation detectors. Depleted U ($^{235}U/^{238}U$ = 0.18 ±0.01%) and natural Au samples (mass ~ 3 up to 6 mgr) irradiated over the U-blanket to measure the slow neutron fluencies at the resonance region (epithermal neutrons) [30]. In accessory, natural Cd foils of 1mm thickness (mass ~ 2 gr, purity 99.9%) were used for determining both the secondary epithermal neutrons as well as the secondary proton fluence [31]. The natural Cd foil effectively captures neutrons below 1 eV, mainly because of the very high cross section of ^{113}Cd for thermal neutron capture, but at the same time it can be used as an epithermal neutron detector via the $^{114}Cd(n,\gamma)^{115}Cd$ reaction. Moreover, natural Cd has a significant cross section for the $^{nat}Cd(p,x)^{111}In$ reaction for proton in the energy range 1 MeV ≤ E_p ≤100 MeV, which covers well the secondary proton spectrum around a spallation target [16, 32]. The activation detectors irradiated with 10^{13} protons regarding to measure both secondary neutron and proton with uncertanty less than 5%.

The experimental data, i.e. the counting track density from the SSNTDs and gamma spectrum from the activation detectors, need to be converted to neutron fluence. In both cases the use of a cross section of the reaction with which the detector is responding must be applied. Especially, when the neutron spectrum extends over a wide energy range-as does a spallation neutron spectrum, it is important to specify the energy range in which each reaction is effective. In this way it is possible to estimate the effective cross section, σ_{eff} which dominates in the spectrum [30]. For these calculations the secondary neutron and proton energy spectrum was estimated theoretically by using the high-energy transport code DCM-DEM [33]. Typical hadron spectra over each section of the U-blanket are presented at Figure 3. The ascendancy of the intermediate-fast neutron is evident in an energy range from 10 keV up to 20 MeV, peaking at 0.6 MeV. The secondary proton fluence is three orders of magnitude less than the neutron one, with energy higher than 10 MeV (peak around 60 MeV).

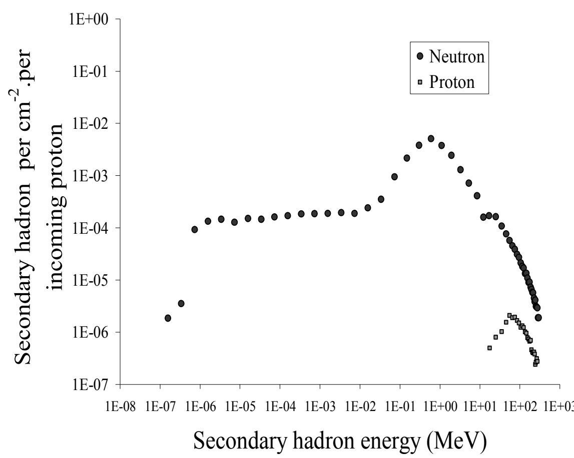

Figure 3. Monte-Carlo simulations of the energy dependent secondary hadron fluence using the Dubna Cascade Model for the Pb (4-section U-blanket) target irradiated by a relativistic proton beam.

3. Results and Discussion

Typical secondary hadrons' distributions determined over the U-blanket for various energy ranges are presented in Figure 4. According to the experimental data the neutron production at the intermediate-fast region (0.3-3 MeV) is 10^{-2} n.cm^{-2}.p^{-1}, one order of magnitude higher than the slow (<10 keV) neutron fluence. The slow neutron production is about half of the fast neutron (>2 MeV): 10^{-3} n.cm^{-2}.p^{-1}. From the energy range of slow neutrons (thermal to 10 keV) the most important contribution comes from epithermal area (1eV up to 10 keV) as it is concluded from SSNTDs measurements. No significant difference in track densities between the Cd-covered and uncovered CR-39 foil plus $^{6}Li_2B_4O_7$ converter was measured. Thus, the secondary neutron spectrum contains only a negligible amount of thermal neutron. Consequently, the measured track density from $^{235}U(n,f)$ reaction occurs from neutron with energy higher than 1 eV. Taken into account the effective cross section of $^{232}Th(n,f)$ reaction data, a neutron production around

5×10^{-3} $n.cm^{-2}.p^{-1}$ is determined. The secondary proton production is about three orders of magnitude lower than the secondary neutron. The experimental data seems to be in fairly good agreement with the Monte-Carlo simulations using the DCM-DEM code (see Fig.3).

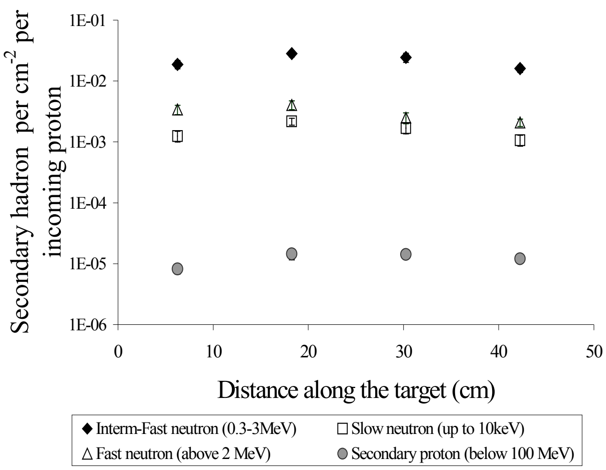

Figure 4. Hadrons production yield over the Pb(U-blanket) target irradiated by a relativistic proton beam with energy 1 GeV.

The total secondary hadron spatial distribution over the U-blanket is presented in Figure 5. Both neutron and proton production increases with a maximum value at around 18 cm from the beam entrance in the target, over the second section of the U-blanket. After that, a decrease towards the target end is observed.

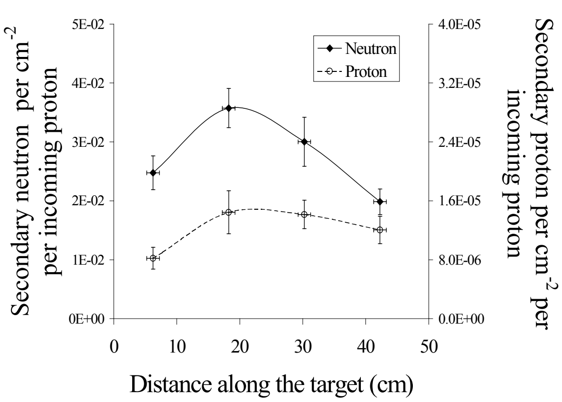

Figure 5. **S**econdary hadron spatial distribution measured over the Pb(U-blanket) target irradiated by a relativistic proton beam with energy 2 GeV.

The experimental spatial distribution of hadrons can be explained by taking into account the interactions of high-energy protons in a thick target, where two competitive effects have to be considered: an exponential increase of secondary particle production in the beginning of the target (build-up effect) and an exponential decrease of the intensity of the primary proton beam along the target (attenuation effect) [16]. The neutron production drops more evident than the secondary proton production that seems to be quite constant from second up to fourth section of U-blanket, within uncertainty to the value obtained for the proton distribution. The neutron production at the fourth section appears to be about 40% less than at the second section. Thus, the second section of the U-blanket is the more effective area over the target for transmutation and/or incineration purpose.

In order to determine the neutron and proton multiplicity as a function of proton beam energy, the determined hadron fluences were integrated over the entire surface of each particular section of the U-blanket. The total multiplicities over the entire target for various energy ranges and for all incoming proton beams are presented at Table 1. The secondary neutron production from this electronuclear set-up is about the double compared with the neutron production around a similar Pb target without the U-blanket, whereon ~30 secondary neutron produced by primary proton for a proton beam energy 1.5 GeV [15].

Table 1. Neutron and proton multiplicity determined in irradiations of the Pb(U-blanket) target with relativistic proton beams from 0.7 up to 2 GeV.

	Secondary hadron per primary proton			
Energy range	0.7 GeV	1.0 GeV	1.5 GeV	2.0 GeV
Slow neutron (< 10 keV)	3 ±0.2	5 ±0.4	6 ±0.4	8 ±0.5
Intermediate-Fast Neutron (0.3-3MeV)	30 ±2	38 ±5	64 ±5	77 ±5
Fast neutron (>2 MeV)	7 ±0.3	9 ±0.4	12 ±1	15 ±1
Total neutron	**40 ±2**	**51 ±5**	**82 ±5**	**100 ±5**
Total proton (<100 MeV)	**0.023 ±0.002**	**0.036 ±0.003**	**0.051 ±0.004**	**0.064 ±0.005**

To optimize the performance of a system with respect to neutron production the energy cost to produce the proton beam has to be taken into account. Thus, the neutron cost defined as the number of produced neutrons per incoming proton normalized to the proton beam energy was calculated for “Energy plus Transmutation” set-ups. According to the experimental results (Fig. 6) the total neutron cost presented to be constant for all studied proton beam energies. These values are quite different with those reported from other studies on extended Pb target without a surrounding moderator where the best neutron cost appeared at 1 GeV incoming proton beam [1,2,15]. Although the total neutron cost remains stable the neutron cost in the appropriate energy ranges for transmutation (i.e. slow neutron) and/or incineration (i.e. fast neutron) seems to increase for low energy incoming proton.

Using this neutron source in an hypothetical Accelerator Driven System with 10 mA proton beam, the high production rate of fast neutrons of about 5×10^{17} neutrons s^{-1} will result in a substantial incineration rate through (n,f) reaction to long-lived actinides produced during the fuel process. Moreover, the rate of the secondary slow neutrons about 2 x 10^{17} neutrons·s^{-1} could be used for the transmutation of ^{239}Pu via (n,f) reactions and long-lived fission products through (n,γ) reactions. The following transmutation/incineration rates (% per month) have been determined by Adam et al. 2004 over the second section of the U-blanket for an hypothetical Accelerator Driven System working with 10 mA proton beam (incoming proton beam energy 2GeV): ^{239}Pu(n,f) = 4.28, ^{238}Pu(n,f) = 1.14, ^{237}Np(n,γ) = 2.80 and ^{129}I(n,γ) = 0.15 % per month.

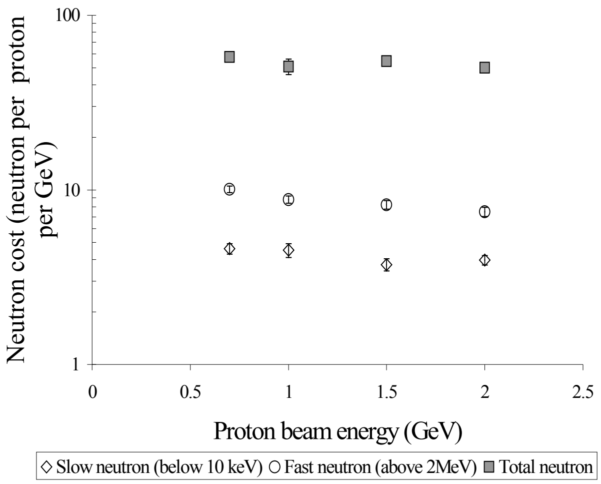

Figure 6. The "neutron cost" of secondary neutron production using relativistic proton beams.

The operation of the ADS require a proper shielding in order to reduce the elevated secondary neutron production, about 2 x 10^{18} neutrons s^{-1}, and shift them, particularly those emitted in intermediate-fast energy range (between 0.3 MeV and 3 MeV), to energies useful for transmutation reactions. After 16 cm in thickness polyethylene shielding covered by 1 mm of Cd, the measured intermediate-fast neutron fluence is about one order of magnitude less than the fluence produced from the target. The slow neutron (<10 keV) fluence after the shielding presented to be similar to the intermediate-fast neutron fluence, on the contrary to the combined Pb/U-blanket target (Table 2.)

Table 2. Secondary neutron fluence measured over the 2nd section of the U-blanket and after 16 cm in thickness polyethylene plus 1mm of Cd shielding (irradiation with 1.5 GeV proton beam).

	Neutron fluence (x10^{-3} n.cm^{-2}.p^{-1})	
	Over U-blanket	After shielding
Slow neutron (< 10 keV)	1.9 ±0.4	0.7 ±0.1
Intermediate-Fast neutron (0.3-3MeV)	30 ±3	0.9 ±0.2

Moreover, the released secondary fast neutron rate (~5 x 10^{17} neutrons.s^{-1}) as well as secondary proton rate (~2 x 10^{15} protons.s^{-1}) has to be considered regarding to choose the appropriate construction materials in order to tolerate the He and H production, respectively. For a Fe-based structural material with 0.15 b cross section in fast n+Fe produced He reaction [10], which after one year of the specific ADS operation it irradiated by ~ 10^{22} neutron.cm^{-2}. The accumulation of such amount of fast neutron causes ~2x10^{-3} deposited He atoms in the material per year. Assuming that the total amount of produced protons slows down inside the constructing materials they should tolerate maximum 0.01 moles of H per year. Especially, the combined target could be affected by one order of magnitude more hydrogen moles per year, noticing that the evaporating secondary protons are trapped in the target. So, the use of a liquid target could solve the problem since is easier to remove the produced hydrogen and to avoid any adverse effects like bubbles or even cracks in the solid matter, as has been proposed by G.S. Bauer [16].

4. Conclusion

The "Energy plus Transmutation" electronuclear set-up is a spallation source with a combined Pb/U target ~50 cm in length consisted of a cylindrical Pb core surrounded by a ~7 cm thick U-blanket. Irradiations with relativistic proton beams (from 0.7 up to 2 GeV) produce neutron mostly at the intermediate-fast region (0.3-3 MeV) that appears to be 10^{-2} n.cm^{-2}.p^{-1}, one order of magnitude higher than the slow neutron (<0.01 MeV). The slow neutron production was measured to be the half of the fast neutron one. The secondary proton production is about tree order of magnitude lower than the secondary neutron. For all studied proton energies, both neutron and proton production increases after ~ 18 cm from the beam entrance in the target with a maximum value over the second section of the U-blanket. After that a decrease towards the target end is observed. The neutron production drops more evident than the secondary proton production that seems to be quite constant from second up to fourth section of U-blanket.

The secondary neutron production from this electronuclear set-up is about the double compared with the neutron production around a similar Pb target without the U-blanket [15]. Considering the cost effectiveness of neutron production, the neutron cost appears to remains constant for all studies proton beam energies. That is quite different compared with the data reported from other studies on extended Pb target without a surrounding moderator where the best neutron cost appeared at 1 GeV incoming proton beam [1,2,15].

Using this spallation target in an hypothetical Accelerator Driven System with 10 mA proton beam, the production rate of fast neutrons at about 5 x 10^{17} neutrons s^{-1} would cause substantial incineration of long-lived actinides through (n,f) reactions. Moreover, the rate of the secondary slow neutrons about 2 x 10^{17} neutrons·s^{-1} could be used for the transmutation of ^{239}Pu via (n,f) reactions and long-lived fission products through (n,γ) reactions.

The ADS requires a proper shielding in order to reduce the elevated secondary neutron production, about 2 x 10^{18} neutrons s^{-1}. After 16 cm in thickness polyethylene shielding covered by 1 mm of Cd, the measured intermediate-fast neutron was about one order of magnitude less than the neutron produced from the target. Taking into account the secondary fast neutron rate (~5 x 10^{17} neutrons.s^{-1}) as well as secondary proton rate (~2 x 10^{15} protons.s^{-1}) the construction materials should be able to withstand up to $2x10^{-3}$ deposited He atoms and 0.01 moles of H per year, assuming that the total amount of produced protons slows down inside the constructing materials.

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