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A spallation neutron source based on Pb target surrounded by U blanket

M. Zamani^{a,*}, M. Fragopoulou^a, S. Stoulos^a, M.I. Krivopustov^b, A.N. Sosnin^b, R. Brandt^c, W. Westmeier^c, M. Manolopoulou^a

^aNuclear and Elementary Particle Physics Department, School of Physics, Aristotle University of Thessaloniki, Greece ^bHigh Energy Laboratory, JINR, Dubna 141980, Russia ^cPhilipps Universitat, Marburg 35032, Germany

Abstract

A new spallation source, efficient for transmutation experiments, was designed and constructed at the Dubna High Energy Laboratory (LHE). The spallation source has a cylindrical shape Pb target surrounded by ^{nat}U rods. Experiments with protons of 0.7–2 GeV were performed and neutron spatial distribution on the surface of U blanket was studied. Total neutron fluences and estimates of their energy distributions were determined using solid state nuclear track detectors. Slow and fast neutron components were studied as a function of the proton beam energy. The experimental results were fitted and compared with calculations derived from empirical relations based on physics near high-energy accelerators. The results show that neutron spatial distribution along the U blanket surface (parallel to the beam direction) has the same shape independent of the proton beam energy. The neutron fluence spatial distribution is characterized by an increase at the beginning of the target and after reaching a maximum drops as a function of the target thickness. The maximum is reached at about one mean free path of protons in the Pb target. The total number of neutrons produced, as was measured on U blanket surface, is an increasing function of the proton beam energy. © 2008 Elsevier Ltd. All rights reserved.

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1. Introduction

In the last decade, in order to reduce the radiotoxic inventory of nuclear waste, accelerator-driven systems (ADS) have been suggested to transmute the accumulated transuranic elements (Bowman et al., 1992). In a subcritical core it is possible to use large fractions of fuels, apart from plutonium also other materials consisting of the minor actinides americium and curium. Intensive research programs investigating physics and technology of proton accelerator spallation targets and subcritical cores are required for the development of full-scale ADS in the future.

With regard to the nuclear physics properties underlying these projects, proton-induced spallation reactions on various high-Z targets at incident energies $E_p > 500 \text{ MeV}$ have been extensively studied. Spatial distributions of total neutron fluence and the energy distributions at different locations around the spallation sources were investigated in order to design an

* Corresponding author. Tel./fax: +30 231 099 8176.

E-mail address: zamani@physics.auth.gr (M. Zamani).

efficient source concerning radionuclides of interest to be transmuted.

Parallel to the various projects two spallation setups have been developed at the Laboratory of High Energies in Dubna (Russia). The two facilities have the same basic construction consisting of a Pb target but enveloped by different materials. The first one is covered by a paraffin moderator while the second one by natural U for neutron multiplication. The resulting neutron spectrum is for the first case a fast neutron spectrum that contains a high thermal neutron component (Gamma-2 experiment) while the second setup gives a fast neutron spectrum (Energy + Transmutation experiment). The low energy spectrum is useful for fission chain process by reactions induced by thermal neutrons but also for transmutation reactions. Some summarized results collected during last decade experiments are presented in Table 1. It is obvious that for some nuclides the Gamma-2 neutron spectrum is more convenient and for other the fast neutron spectrum (including neutrons from xn reactions) gives higher transmutation efficiencies.

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Table 1 The most important radionuclides to be transmuted at the Dubna transmutation setups

Element	Isotope	Half life (years)	Experimental setup
Plutonium	²³⁸ Pu	88	U blanket
	²³⁹ Pu	2.4×10^{4}	Gamma-2
	²⁴⁰ Pu	6.5×10^{3}	U blanket
Minor actinides	²³⁷ Np	2.1×10^{6}	Gamma-2
	²⁴¹ Am	430	U blanket
	²⁴³ Am	7.5×10^{3}	U blanket
	²⁴³ Cm	8.5×10^3	Gamma-2
Fission products	⁹⁰ Sr	29	Gamma-2
	⁹³ Zr	1.5×10^{6}	Gamma-2
	⁹⁹ Tc	2.1×10^{5}	Gamma-2
	¹⁰⁷ Pd	6.5×10^{6}	Gamma-2
	¹²⁹ I	1.6×10^{7}	Gamma-2
	¹³⁵ Cs	2.3×10^{6}	Gamma-2
	¹³⁷ Cs	30	Gamma-2

The results concerning neutron yields and distributions of Gamma-2 experiments have been described in various publications (Adloff et al., 1999; Stoulos et al., 2004; Fragopoulou et al., 2005). Radiochemical results have also been published (Wan et al., 1998; Adam et al., 2002) and some of them will be presented in this conference. The present work is dedicated to the study of neutron yields and distribution of the Energy + Transmutation experiment. A theoretical approach of the results based on high energy physics concept will also be discussed. The shielding of the source is also studied and the results are given in this paper.

2. Experimental

The new transmutation assembly consists of a cylindrical Pb target of 48 cm in length and 8 cm in diameter. Around the Pb target, U rods are placed hexagonally. Each U rod is a cylinder, 10.4 cm in length and 3.6 cm in diameter. Four sections of this U blanket were needed to cover the target. The shielding consists of 1 mm Cd and 15–26 cm in thickness granulated polyethylene. Experiments were performed at the Nuclotron accelerator (High Energy Laboratory) in Dubna, using protons from 0.7–2 GeV.

Neutron fluence measurements in the surface of U blanket as well as on the top of the shielding were performed. On the surface of the U blanket, four sets of SSNTD were positioned in the parallel direction relative to the axis of the cylindrical Pb target. At the corresponding positions on top of the shielding, three sets of SSNTD were placed to measure the neutrons escaping the setup.

Each set of SSNTDs contains $P_{oly}A_{llyl}D_{iglycol}C_{arbonate}$ (Pershore Mouldings Standard Grade, PM355) foils that act as particle detectors, in contact with Kodak LR115 Type 2B partially covered by 1 mm Cd. In addition, a bare foil was also used in each set to detect proton recoils and give supplementary information for intermediate-fast neutrons above 0.3 MeV (Harvey et al., 1998). So each set was able to detect thermal–epithermal and intermediate–fast neutrons at the same

positions. Details on the operation of these systems and their response are given in our previous paper (Zamani et al., 1996).

Sets of SSNTDs as fission detectors were placed on the U blanket surface in the middle of each section for measuring neutron spatial distributions. They consist of about 1 mg/cm^2 of ^{235}U or ^{232}Th evaporated on Makrofol. Details of the method can be found in the paper by Remy et al., 1970.

3. Results and discussion

The large place to put samples for transmutation practically is the U-blanket surface. So, it is important to study neutron yield along the U-blanket surface parallel to the beam direction. The results taken by 235 U and 232 Th in each U-blanket section are given in Fig. 1. It is clearly seen that neutron yield depends on the position along the U blanket. The maximum of the distribution is found to be around 120 mm from the beginning of the target, at the beam entrance side. The result is connected to the proton mean free path in Pb (100 mm). The same result was obtained by the use of Boron converter as can be seen in Fig. 2, for various proton beam energies. Enhanced neutron



Fig. 1. Fast neutron spatial distribution along the surface of the U blanket derived by SSNTDs as fission detectors with appropriate converters for 2 GeV incoming proton beam.



Fig. 2. Total neutron spatial distribution along the surface of the U blanket for various relativistic proton beams obtained using SSNTDs as particle detectors.



Fig. 3. The experimental arrangement of ²³⁵U fission detectors plus 1 mm Cd foil placed along the U-blanket surface regarding study of the thermal neutron production.



Fig. 4. The measured fission tracks per incoming proton over the surface of the U blanket derived by 235 U plus 1 mm Cd foil experimental arrangement.

fluence was observed with the increasing of beam energy in all positions where neutrons were measured. The difference between covered and uncovered with Cd samples is not observed which means that there are no thermal neutrons (upto 1 eV) at the U-blanket surface. To answer the question if they produced some thermal neutrons by the U-blanket an additional experiment was performed. Makrofol detectors with ²³⁵U converter were placed along the U-blanket surface, parallel to the beam direction, every 3 cm. The arrangement can be seen in Fig. 3. Cd foils, 1 mm in thickness, from the shielding side, covered one sample (Series A'). The next sample was covered by the Cd foil from the U-blanket side (Series A). From the fissions counted in Makrofol detectors it shows that some thermal-epithermal neutrons come to the U-blanket surface from the shielding, as seen in Fig. 4; thus there are some thermal neutrons back scattered in the shielding polyethylene. Taking into account that the bottom of the polyethylene contains 2 mm Cd, the backscattered neutrons counted at the U-blanket surface are about 10% of the neutrons escaping the shielding. So, the measured fissions by ²³⁵U converter at the U-blanket surface correspond in the majority to fast and intermediate neutrons. This fact was

verified by the fission measurements of 232 Th converter, which were placed at the same position with 235 U samples. The small difference in neutron numbers counted by 235 U and 232 Th, seen in Fig. 1 can be attributed to the fact that fission of 232 Th starts from about 2 MeV while in 235 U fission reactions have a continuous cross section from thermal to fast neutrons.

The neutron spatial distribution along the U blanket was fitted according to the conception of high energy physics reactions (Sullivan, 1992). We assume that at the beam side of the target (upstream) primary reactions take place. The particles produced (mainly protons and neutrons) after the first reaction stage have enough energy to produce secondary reactions, etc. giving rise to multiplication of primary produced particles. In parallel, because of the beam attenuation along the target, the number of the beam protons diminishes. So, the competition between the two effects, of a built-up effect and beam attenuation deeper into the target can describe the observed behavior of the neutron spatial distribution at the U-blanket surface (N, neutron fluence per proton, cm⁻²). This behavior can be expressed by a relationship:

$$N = C(1 - ae^{-bx})e^{-dx} \tag{1}$$

The first part of the relation describes the built-up effect and the second corresponds to the beam attenuation; a and b are called built-up parameter and built-up coefficient (mm⁻¹), respectively, while d represents the beam attenuation coefficient (mm⁻¹). C is a constant neutron fluence per proton having value from 0.03 to 0.21 cm⁻² for proton energy ranges from 0.7 to 2 GeV. Fitting results are presented in Fig. 5. The behavior of the built-up coefficient versus beam energy can be described by a relationship of the form:

$$b = (1.21 \pm 0.37)(1 - e^{[(7.6 \pm 0.9).10^{-3}]E_p})$$
(2)

The built-up parameter is found to be independent of the proton beam energy. Its value is around unity. The beam attenuation coefficient is energy dependent as in all cases of radiation. A linear expression versus proton energy fits the data, for the



Fig. 5. The fitting process (solid line) applied to experimental data of neutron spatial distribution over the surface of the U blanket for the 0.7 GeV incoming proton beam.



Fig. 6. The build-up parameter and coefficient of the neutron fluence as a function of proton beam energy. The results were deduced from neutron spatial distribution over the surface of the U blanket.

energy range of the presented experiments:

$$d = (8.3 \pm 0.1) \cdot 10^{-3} - [(2.6 \pm 0.1) \cdot 10^{-3}]E_{\rm p}$$
(3)

The behavior of the built-up parameter and coefficient versus proton beam energy are given in Fig. 6. In Fig. 7 the attenuation coefficient versus proton beam energy is presented.

3.1. The U-blanket shielding

The knowledge of escaping neutrons from the shielding is an important factor for the realization of experiments with spallation sources. In all cases the number of escaping neutrons has to be a converter in neutron doses, for a given spectrum and these doses have to meet radiation protection standards. For that reason separate measurements were performed in the upper surface of the shielding by SSNTDs. The samples with Boron converter, covered and uncovered with Cd 1 mm, were positioned along the target direction. The neutron spatial distribution is found to be stable along the shielding surface. The intermediate-fast neutrons produced by the target diminish



Fig. 7. The attenuation coefficient of the neutron fluence as a function of proton beam energy. The results were deduced from neutron spatial distribution over the surface of the U blanket.

at about two orders of magnitude traversing the polyethylene. About half of the initial fast neutrons slow down to thermal energies. The corresponding dose remains high compared to permit doses, so additional shielding is needed in order to safely operate the spallation source.

4. Conclusion

A spallation source consisting of Pb target and U blanket with hard neutron spectrum operates in the Dubna High Energy Laboratory. Experiments with proton beams at 0.7–2 GeV show a neutron spatial distribution along the U-blanket surface peaked at about one mean free path of protons in the Pb target. The neutron energies are in the region of intermediate–fast neutrons. On the polyethylene shielding neutrons escaping are stable along the upper surface and parallel to the target. Their number is about two orders of magnitude less than in the U-blanket surface. Half of the escaping neutrons are thermalized (about $10^{-5} n/cm^2.p$) and the total radiation dose produced at the shielding surface is found to be greater than the permitted radiation limits.

References

- Adam, J., et al., 2002. Transmutation of ²³⁹Pu and other nuclides using spallation neutrons produced by relativistic protons reacting with massive U and Pb targets. Radiochim. Acta 90, 431–442.
- Adloff, J.C., Brandt, R., Debeauvais, M., Fernandez, F., Krivopustov, M.I., Kulakov, B.A., Sosnin, A.N., Zamani, M., 1999. Secondary neutron production from thick Pb target by light particle irradiation. Radiat. Meas. 31, 551–554.
- Bowman, C.D., et al., 1992. Nuclear energy generation and waste transmutation using an accelerator driven intense thermal neutron source. Nucl. Instrum. Meth. Phys. Res. A 320, 336–367.
- Fragopoulou, M., et al., 2005. Spatial distribution of moderated neutrons along a Pb target irradiated by high energy protons. Nucl. Instrum. Meth. Phys. Res. A 560, 571–576.
- Harvey, J.R., Tanner, R.J., Alberts, W.G., Bartlet, D.T., Piesch, E.K., Schraube, H., 1998. The contribution of Eurados and CENDOS to etched track neutron dosimetry. Radiat. Prot. Dos. 77, 267–304.
- Stoulos, S., et al., 2004. Neutron measurements by passive methods in the Dubna transmutation assemblies. Nucl. Instrum. Meth. Phys. Res. A 519, 651–658.

- Sullivan, A.H., 1992. A Guide to Radiation and Radioactivity Levels near High Energy Particle Accelerators. Nuclear Technology Publishing, Ashford, Kent, UK, pp. 7–49.
- Remy, G., Ralarosy, J., Stein, R., Debeauvais, M., Tripier, J., 1970. Heavy fragment emission in high energy reactions on heavy nuclei. J. de Phys. 31, 27–34.
- Wan, J.-S., et al., 1998. Transmutation of radioactive waste by means of relativistic heavy ions. Kerntechnik 63, 167–177.
- Zamani, M., Sampsonidis, D., Savvidis, E., 1996. An individual neutron dosemeter with (n, a) and (n, p) converter. Radiat. Meas. 26, 87–92.