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NEUTRON PRODUCTION IN $p + \text{Pb/U}$ AT 2 GeV

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Рождение нейтронов в $p + \text{Pb/U}$ при 2 ГэВ

Массивная свинцовая мишень, окруженная урановым blanketом установки «Энергия плюс трансмутация», облучалась пучком протонов нуклотрона ЛФВЭ ОИЯИ с энергией 2 ГэВ. Нейтроны, образующиеся в свинце и размножающиеся в уране, регистрируются с помощью различных активационных детекторов. Экспериментальные результаты сравниваются с расчетами по методу Монте-Карло, выполненными по программе MCNPX. Разыгрывались пространственное распределение и энергетические спектры нейтронов и протонов, образующиеся в установке, и сечения (n, xn) - и (p, pxn) -реакций. Моделирование качественно хорошо описывает продольное распределение выхода продуктов, однако предсказывает более резкий спад их выхода с ростом радиуса, чем экспериментальные результаты.

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Neutron Production in $p + \text{Pb/U}$ at 2 GeV

The «Energy plus Transmutation» setup consisting of a thick lead target surrounded by a uranium blanket was irradiated with 2 GeV protons. The produced neutrons were measured by means of activation detectors. The experimental results were compared with Monte Carlo simulations performed with the MCNPX code. The simulated quantities are spatial distributions and energy spectra of neutrons and protons produced in the setup and cross-sections of (n, xn) and (p, pxn) reactions. Simulations describe qualitatively well longitudinal distributions of activation yields, while they predict much steeper decrease of the yields with growing radial distance than it was measured.

The investigation has been performed at the Veksler and Baldin Laboratory of High Energy Physics, JINR.

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1. INTRODUCTION

Transmutations of long-lived actinides and fission products from nuclear waste, plutonium from nuclear weapons, or thorium (as an energy source) have been investigated with the increasing interest in the last two decades. Different concepts of transmutation also involve the Accelerator Driven Systems (ADS) [1] based on a subcritical nuclear reactor driven by an external spallation neutron source.

The spallation reaction is a process in which a light projectile (p, n , light nucleus) with $E \sim 0.1 \div 1$ GeV interacts with a heavy nucleus (e.g., W, Pb) and causes emission of a large number of hadrons. Spallation has two stages: intranuclear cascade (INC) (including preequilibrium emission) and deexcitation (evaporation or fission). In the case of a thick target, high-energy particles (mainly neutrons) emitted from the nucleus in the course of intranuclear cascade can induce further spallation reactions and generate internuclear cascade. For some target materials, spallation neutrons with $E_n \sim 10$ MeV can enlarge neutron production by (n, xn) reactions. Globally, the incident particle induces the production of a large amount of neutrons with wide energy spectra, which can be used for transmutation of relevant nuclei.

«Energy plus Transmutation» (E+T) is an international project [2] for the study of spallation reactions, neutron production and transport, and the transmutation of fission products and higher actinides by spallation neutrons. The E+T setup consists of a thick lead target with a subcritical, natural uranium blanket surrounded by polyethylene shielding. Six irradiations of the E+T setup have been performed until now, on the proton beam with the kinetic energies of 0.7, 1.0 [3], 1.5 [4–6], and 2.0 (this paper) GeV and deuteron beam with energies of 0.8 AGeV [7] and 1.26 AGeV [8].

This paper describes the measurement of the produced neutron field in the experiment performed on the proton beam with a kinetic energy of 2 GeV and the comparison of experimental results with Monte Carlo simulations performed with MCNPX.

2. EXPERIMENT

The E+T setup consists of three main parts: a cylindrical Pb target (diameter of 84 mm), a deep-subcritical ($k_{\text{eff}} = 0.202$ [9]) $^{\text{nat}}\text{U}$ blanket (of a hexagonal cross-section with a side length of 130 mm), and $(\text{CH}_2)_n$ shielding with inner Cd layer (Fig. 1). The target/blanket part has a length of 480 mm, which is divided into four sections of 114 mm in length separated by 8 mm gaps (Fig. 2). It is placed in a polyethylene shielding of approximately cubic size ($\approx 1 \text{ m}^3$). The inner walls of the shielding are coated with a Cd layer (thickness of 1 mm). The front and the back ends of the setup are not shielded. The whole assembly mass is 950 kg, thereof 28.66 kg weighs the Pb target and 206.4 kg weighs the U blanket. For detailed setup description and discussion about the influence of individual setup components on the produced neutron field, see [9].

The E+T setup was irradiated with the 2 GeV proton beam (left part of Fig. 3) from the Nuclotron accelerator at the Veksler and Baldin Laboratory of High

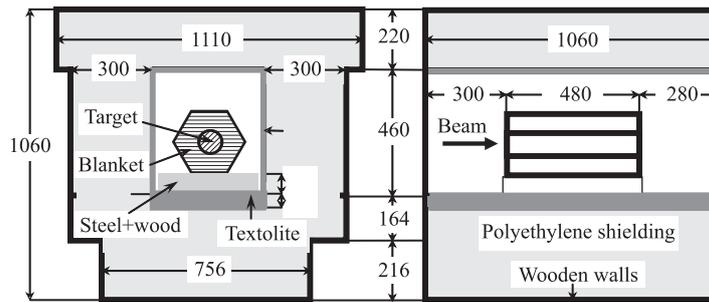


Fig. 1. Front view (left) and cross-sectional side view (right) of the «Energy plus Transmutation» setup. Dimensions are in millimeters

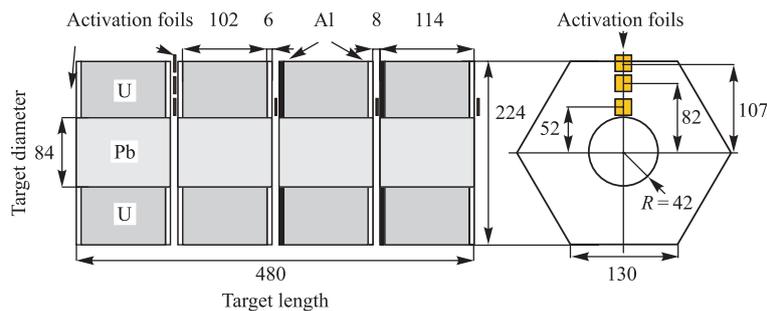


Fig. 2. The placement of activation foils: side view (left), cross-sectional view in the first gap (right). Dimensions are in millimeters

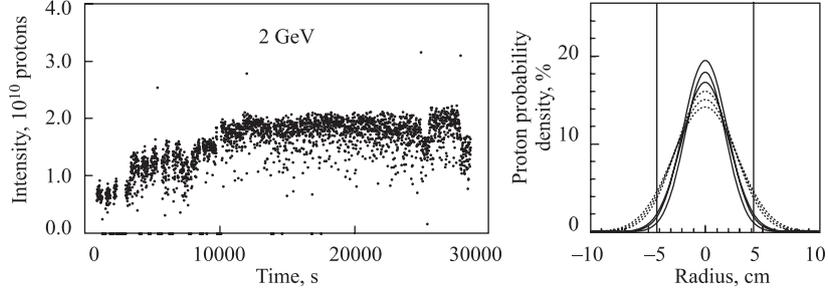


Fig. 3. The course of irradiation. Each point represents one pulse of protons measured by a proportional chamber (left). Profile of the 2 GeV proton beam is measured by SSNT detectors (in vertical direction) and fitted with a Gaussian curve. Solid curves represent fit and its errors in the position in front of the target. Dotted curves represent fit and its errors in the position in the first gap between target sections (right)

Energies at JINR, Dubna. The beam energy was determined with an accuracy of 0.5%. The total beam flux was measured by proportional chamber, Al and Cu activation foils. The beam geometry properties (shape, location, direction) were determined by lead solid-state nuclear track (SSNT) detectors and a set of Cu activation foils (details of these methods can be found in [10,11]). The beam had approximately elliptical shape and was parallel with the target axis. The central part of the beam profile was fitted with the Gaussian distribution (Table 1, right part of Fig. 3).

The produced neutron field was probed by nonthreshold (n, γ) reaction and threshold (n, α) , $(n, x/n)$ reactions in ^{27}Al , ^{197}Au , and ^{209}Bi foils. The foils had square size of 20×20 mm (Al, Au) and 25×25 mm (Bi) with thickness of 0.4 mm (Al), 0.04 mm (Au), and 1 mm (Bi).

Two sets of activation foils were placed in the gaps between target/blanket sections, first to measure longitudinal distribution and second to measure radial distribution of the produced neutron field (Fig. 2). The first set was placed at the radial distance $R = 5.2$ cm from the target axis at four *longitudinal* distances $X = 11.8, 24.0, 36.2, 48.4$ cm from the target front (i.e., in the gaps between blanket sections and behind it). The second set was placed in the first gap

Table 1. The parameters of 2 GeV proton beam

Irradiation time	Beam integral	Vertical FWHM	Horizontal FWHM	Vertical position	Horizontal position	Range (from [12])
7 h 43 min	$1.25(6) \times 10^{13}$	5.4(3)	3.8(3)	0.3(2) cm	-1.4(2) cm	132 cm

between the first and second blanket sections (i.e., at the longitudinal distance $X = 11.8$ cm from the target front) at three *radial* distances $R = 5.2, 8.2, 10.7$ cm from the target axis.

The activities of the activated foils were measured off-line by HPGe γ spectrometer at the Dzheleпов Laboratory of Nuclear Problems of JINR, Dubna, and at the Department of Nuclear Spectroscopy at the Nuclear Physics Institute of the Academy of Sciences of the Czech Republic in Řež. The measured γ spectra, covering a region approximately from 50 up to 3000 keV, were processed by the DEIMOS32 code [13] that provides a Gaussian fit of γ peaks. The fitted peak areas were corrected for standard spectroscopical corrections. The final obtained value for each produced isotope is the yield, i.e., the number of activated nuclei per one gram of activated material and per one incident proton.

Yields are shown in the semi-logarithmic scale in Fig. 4 and Tables 2, 3. The delineated errors are of statistical origin given by the error of the Gaussian fit of γ peaks. Experimental errors, mainly the inaccuracies of the beam and activation foils displacements, beam intensity, and γ -spectrometer efficiency determinations contribute another 30% [9] as a systematic error, which mainly change the absolute values and less the shape of spatial distribution.

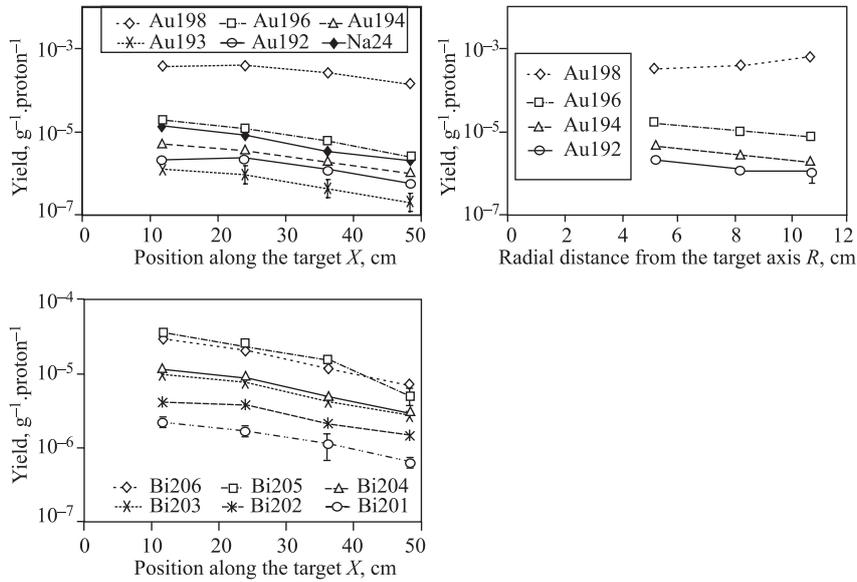


Fig. 4. Longitudinal (left) and radial (right) distributions of the experimental yields of nuclei produced in Al, Au, and Bi foils. The lines connecting experimental points are delineated to guide readers' eyes

Table 2. The experimental yields of nuclei produced in Al and Au foils

Foil	²⁷ Al	¹⁹⁷ Au				
Reaction	(<i>n</i> , α)	(<i>n</i> , γ)	(<i>n</i> , 2 <i>n</i>)	(<i>n</i> , 4 <i>n</i>)	(<i>n</i> , 5 <i>n</i>)	(<i>n</i> , 6 <i>n</i>)
Product	²⁴ Na	¹⁹⁸ Au	¹⁹⁶ Au	¹⁹⁴ Au	¹⁹³ Au	¹⁹² Au
E_{thresh} , MeV	5.5	—	8.1	23	30	39
$T_{1/2}$, h	15	65	148	38	18	5
X , cm	Longitudinal yields, $10^{-6} \cdot \text{g}^{-1} \cdot \text{proton}^{-1}$; $R = 5.2$ cm					
11.8	14.15(38)	381(5)	18.9(8)	5.14(24)	1.26(22)	2.10(29)
24.0	8.46(23)	395(5)	12.2(6)	3.54(28)	0.93(37)	2.28(32)
36.2	3.35(13)	271.6(33)	6.21(36)	1.86(17)	0.43(15)	1.24(23)
48.4	1.98(9)	145.0(17)	2.52(20)	0.97(10)	0.20(7)	0.58(6)
R , cm	Radial yields, $10^{-6} \cdot \text{g}^{-1} \cdot \text{proton}^{-1}$; $X = 11.8$ cm					
5.2		333(4)	16.4(5)	4.71(27)		2.08(28)
8.2		400(5)	10.0(6)	2.67(20)		1.22(39)
10.7		654(7)	7.58(34)	1.80(11)		1.15(27)

Table 3. The experimental yields of nuclei produced in Bi foils

Foil	²⁰⁹ Bi					
Reaction	(<i>n</i> , 4 <i>n</i>)	(<i>n</i> , 5 <i>n</i>)	(<i>n</i> , 6 <i>n</i>)	(<i>n</i> , 7 <i>n</i>)	(<i>n</i> , 8 <i>n</i>)	(<i>n</i> , 9 <i>n</i>)
Product	²⁰⁶ Bi	²⁰⁵ Bi	²⁰⁴ Bi	²⁰³ Bi	²⁰² Bi	²⁰¹ Bi
E_{thresh} , MeV	22	30	38	45	53	61
$T_{1/2}$, h	150	367	11	12	2	2
X , cm	Longitudinal yields, $10^{-6} \cdot \text{g}^{-1} \cdot \text{proton}^{-1}$; $R = 5.2$ cm					
11.8	29.1(6)	36(6)	11.33(14)	10.08(22)	4.19(7)	2.23(31)
24.0	21.95(38)	23.1(26)	8.61(10)	7.94(19)	3.91(9)	1.69(23)
36.2	12.13(33)	15.6(21)	4.82(9)	4.28(14)	2.16(6)	1.13(18)
48.4	6.84(24)	5.0(12)	2.97(5)	2.80(12)	1.49(5)	0.65(13)

The spatial distributions of yields have similar shapes for all threshold reactions. The longitudinal distributions of yields change for one order of magnitude and have maximum in the first gap between target/blanket sections. The radial distributions of yields decrease with increasing distance from the target (beam) axis.

In the contrary, the yields of nonthreshold reaction (¹⁹⁷Au(*n*, γ))¹⁹⁸Au) only slightly change. The moderation and scattering of neutrons in the polyethylene shielding created an intensive, homogeneous field of neutrons with $E < 1$ keV. These low-energy neutrons give major contribution to (*n*, γ) reaction in all Au foils in the setup. Therefore, the radial distributions of yields of ¹⁹⁸Au are almost flat. In the case of the longitudinal distribution, the contribution of low-energy

neutrons from moderator is decreased behind the target, because the target/blanket is not shielded from its ends, see Fig. 1. Therefore, the yield of ^{198}Au is lower in this position.

3. SIMULATIONS

The Monte Carlo simulations of neutron production in the E+T setup were performed with the MCNPX code version 2.6.C [14]. The influence of possible inaccuracies in the description of the E+T setup geometry on the produced neutron field is negligible [9].

All combinations of INC (Bertini, Isabel, Liège INCL4, CEM) and evaporation models (Dresner, ABLA) available in MCNPX 2.6.C were used. Bertini and Isabel INC models include the Multistage Preequilibrium Exciton Model for

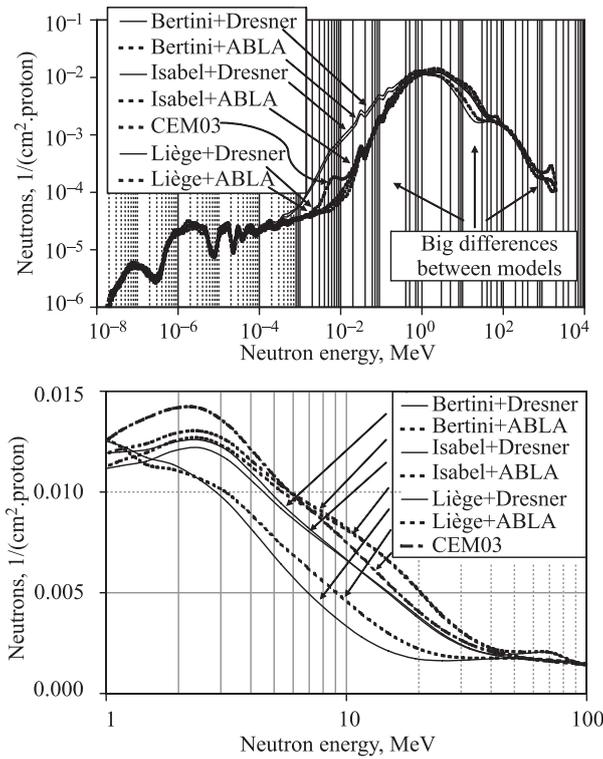


Fig. 5. Simulated neutron spectra (produced in the first target section) in log-log scale. In the lower figure, focus presentation for the domain 1–100 MeV is in semi-log scale

description of preequilibrium emission of particles (only nucleons, photons, and charged pions were taken into account in the presented simulations) and it can be switched on/off, see Fig. 6, right. The CEM03 model uses its own preequilibrium model (the improved Modified Exciton Model) without user-possibility to adjust it. The Liège INC model does not include any preequilibrium model.

The LA150 data library was used as the source of evaluated cross-sections. To reach high enough statistics, from $1 \cdot 10^6$ to $1 \cdot 10^7$ events were simulated in each case.

The simulated produced neutron spectra $\Phi_n(E)$ can be seen in Fig. 5. The largest differences between various INC+evaporation models were found in three regions: between $\sim 10^{-4}$ and $\sim 10^{-1}$ MeV, between 5 and 30 MeV, and above 700 MeV (near to the beam energy).

We concentrated on the domain of tens of MeV (Fig. 5, bottom) as the observed threshold reactions have threshold energies there. Two separated regions can be found in this domain: in the first region the simulations differ significantly when different evaporation models are used, in the second region when different INC models are used, see Fig. 6, left. This can be interpreted as the evidence of the border between intranuclear cascade and evaporation phase of spallation

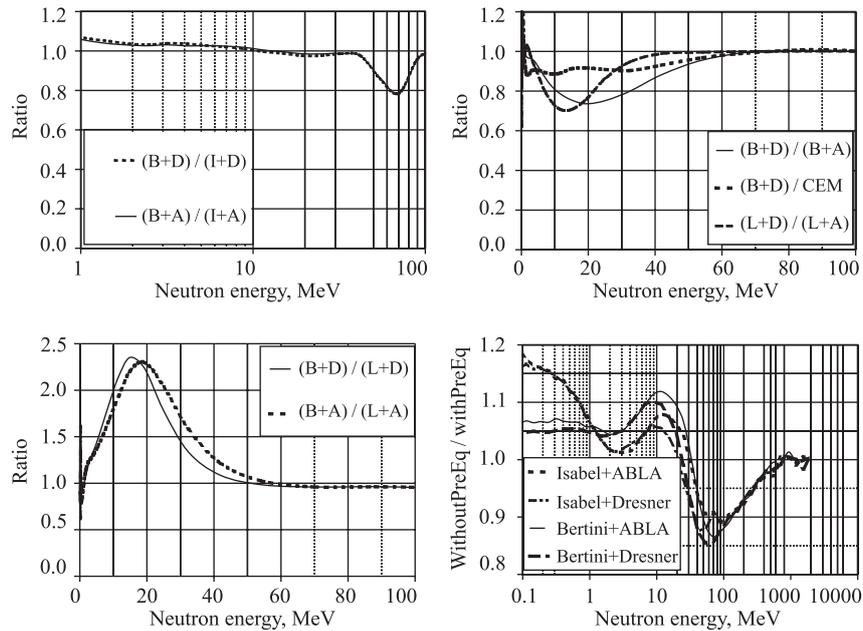


Fig. 6. Top and left bottom: Ratios of simulated neutron spectra from Fig. 5. Right bottom: Ratios of simulated neutron spectra when the preequilibrium models were not and were used. Differences do not exceed 15%

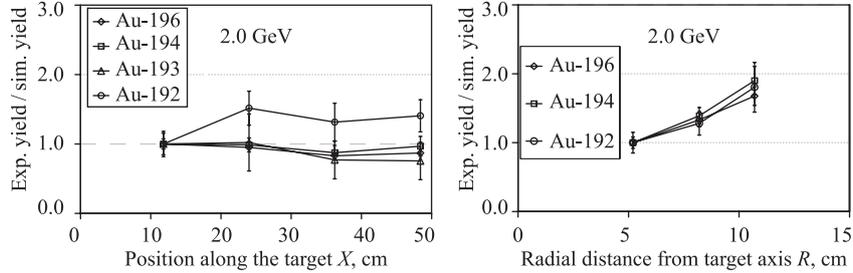


Fig. 7. Relative comparison of experimental and simulated (spectra simulated with Liège INC+ABLA and convoluted with TALYS+MCNPX cross-sections) Au yields in longitudinal (left) and radial (right) directions (normalized to the first foil in each set)

reaction as they are described in the used models. This point appears to be around 40 MeV, which corresponds to the nuclear potential well depth (separation energy plus Fermi energy).

The simulated yields were obtained by means of folding of neutron and proton spectra with relevant cross-sections. The cross-sections of several observed reactions are known with high enough accuracy at least in the energy region up to 40 MeV. The knowledge of most cross-sections is insufficient or they have not been studied at all.

To enable calculations of yields of other observed isotopes and improve existing cross-sections, the TALYS code version 0.79 [15] (up to $E_n = 150$ MeV) and MCNPX (for $E_n > 150$ MeV) were used for calculation of corresponding (n, xn) and (p, pxn) reactions cross-sections (as the isotopes observed in activation samples can be produced in indispensable amount by protons as well [9]).

Comparison with experimental yields shows a reasonable agreement in longitudinal direction, see Fig. 7, left. In the contrary, the ratios between experimental and simulated yields considerably increase with increasing radial distance from the target axis, see Fig.7, right. Similar trend was observed at the 1.5 GeV proton experiment [5], whereas the radial distribution is described well by MCNPX for 0.7 and 1.0 GeV proton experiments [3].

The reason could be in the evaluated cross-section libraries or intranuclear cascade+evaporation models. It looks that they do not describe correctly the angular distribution of the produced high-energy neutrons for proton beam energies bigger than some value between 1.0 and 1.5 GeV. Similar observations have been done on both thick and thin targets [16], when it was found that simulations underestimate the neutron production for backward angles.

Interesting is that the experimental results of the 1.26 AGeV deuteron experiment agree well with the MCNPX simulations. This indicates that the discrepancy in radial direction starts closer to the 1.5 GeV proton beam energy.

CONCLUSION

In the framework of the «Energy plus Transmutation» project, we studied neutron production in the spallation reactions of 2.0 GeV protons directed to the thick lead target with the uranium blanket surrounded by the polyethylene moderator.

The produced neutron field was measured by means of activation detectors. We observed isotopes produced in (n, xn) reactions with threshold energy up to $E_{\text{thresh}} \approx 60$ MeV. The maximum intensity of the high-energy neutron field produced in the spallation target is located in the region between the first and second target/blanket sections.

The experimental results were compared with Monte-Carlo simulations performed using MCNPX 2.6.C. MCNPX describes well the shape of the longitudinal distributions of the yields of threshold reactions. The simulations predict much steeper decrease of the yields with growing radial distance than it was measured. Similar trend was observed at the 1.5 GeV proton experiment. The reason could be in the LA150 evaluated cross-section libraries or intranuclear cascade+evaporation models included in MCNPX.

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REFERENCES

1. Nifenecker H. et al. *Hybrid Nuclear Reactors*, Progress in Particle and Nuclear Physics 43 (1999) 683–827 [<http://lpsc.in2p3.fr/gpr/PPNPport/node1.html>].
2. Krivopustov M. I. et al. *First Experiments with a Large Uranium Blanket within the Installation «Energy plus Transmutation» Exposed to 1.5 GeV Protons*, Kerntechnik 68 (2003) 48–54; JINR Preprint P1-2000-168. Dubna, 2000.
3. Krása A. et al. *Neutron Emission in the Spallation Reactions of 1 GeV Protons on a Thick, Lead Target Surrounded by a Uranium Blanket*, JINR Preprint E15-2007-81. Dubna, 2007.
4. Krivopustov M. I. et al. *Investigation of Neutron Spectra and Transmutation of ^{129}I , ^{237}Np and Other Nuclides with 1.5 GeV Protons from the Dubna Nuclotron Using the Electronuclear Installation «Energy plus Transmutation»*, JINR Preprint E1-2004-79. Dubna, 2004.
5. Křížek F. et al. *The Study of Spallation Reactions, Neutron Production and Transport in a Thick Lead Target and a Uranium Blanket During 1.5 GeV Proton Irradiation*, Czechoslovak Journal of Physics 56 (2006) 243–252.

6. Krása A. et al. *Neutron Production in Spallation Reactions of 0.9- and 1.5-GeV Protons on a Thick Lead Target — Comparison of Experimental Data and MCNPX Simulations*, International Conference on Nuclear Data for Science and Technology 2004, Santa Fe, New Mexico, 26 September – 1 October 2004. AIP Conference Proceedings, Volume 769 (2005) 1555-9. Dubna, 2008.
7. Krivopustov M. I. et al. JINR Preprint E1-2008-54. Dubna, 2008.
8. Krivopustov M. I. et al. *About the First Experimental Investigation of Neutron Production and Transmutation of ^{129}I , ^{237}Np , ^{238}Pu , ^{239}Pu on Pb Target with U Blanket «Energy plus Transmutation» Setup Irradiated by 2.52 GeV Deuterons*, JINR Preprint E1-2007-07. Dubna, 2007.
9. Majerle M. et al. *Monte Carlo Studies of the Energy plus Transmutation System*, JINR Preprint E15-2007-82. Dubna, 2007.
10. Zhuk I. V. et al. *Investigation of Energy-Space Distribution of Neutrons in the Lead Target and Uranium Blanket within the Installation «Energy plus Transmutation» Exposed to 1.5 GeV Protons*, JINR Preprint P1-2002-184. Dubna, 2002 (in Russian).
11. Krása A. et al. *Neutron Production in Spallation Reactions of 0.9 and 1.5 GeV Protons on a Thick Lead Target: Comparison between Experimental Data and Monte-Carlo Simulations*, JINR Preprint E1-2005-46. Dubna, 2005.
12. Berger M. J. et al. *Stopping-Power and Range Tables for Electrons, Protons, and Helium Ions* (2005) [<http://physics.nist.gov/PhysRefData/Star/Text/contents.html>].
13. Frána J. *Program DEIMOS32 for Gamma-Ray Spectra Evaluation*, Journal of Radio-analytical and Nuclear Chemistry 257 (2003) 583–587.
14. Pelowitz D. B. et al. *MCNPX Users's Manual. Version 2.5.0*, LANL report LA-CP-05-0369 (2005).
15. Koning A. J., Hilaire S., Duijvestijn M. C. *TALYS: Comprehensive Nuclear Reaction Modeling*, International Conference on Nuclear Data for Science and Technology 2004, Santa Fe, New Mexico, 26 September – 1 October 2004. AIP Conference Proceedings, Volume 769 (2005) 1154-9.
16. Meigo S. et al. *Measurements of Neutron Spectra Produced from a Thick Lead Target Bombarded with 0.5- and 1.5-GeV protons*, Nuclear Instruments and Methods in Physics Research A 431 (1999) 521–530.

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