Comparison between experimental data and Monte-Carlo simulations of neutron production in spallation reactions of 0.7-1.5 GeV protons on a thick, lead target

A Krása¹, M Majerle¹, F Křížek¹, V Wagner¹, A Kugler¹, O Svoboda¹, V Henzl¹, D Henzlsová¹, J Adam¹², P Čaloun¹², V G Kalinnikov², M I Krivopustov², V I Stegailov², V M Tsoupko-Sitnikov²

¹ Nuclear Physics Institute of the Academy of Sciences of the Czech Republic, Řež near Prague, 25068, Czech Republic
² Joint Institute for Nuclear Research, Dubna, Russia

E-mail: krasa@ujf.cas.cz

Abstract. Relativistic protons with energies 0.7-1.5 GeV interacting with a thick, cylindrical, lead target, surrounded by a uranium blanket and a polyethylene moderator, produced spallation neutrons. The spatial and energetic distributions of the produced neutron field were measured by the Activation Analysis Method using Al, Au, Bi, and Co radio-chemical sensors. The experimental yields of isotopes induced in the sensors were compared with Monte-Carlo calculations performed with the MCNPX 2.4.0 code.

1. Introduction
Within the frame of a complex research of ADS [1], plenty of experiments were performed by the “Energy plus Transmutation” collaboration [2-4] using the superconducting, strong focusing synchrotron named Nuclotron in JINR Dubna, Russia. Protons in a GeV range bombarding a thick heavy metal target cause spallation reactions. Intensive neutron fluxes created in these reactions are of special interest as they can be used to transmute long-lived radioactive isotopes. In this study, three experiments with the same setup, but different proton energies are discussed. The interest is focused on the production of high-energy neutron field, which was measured by the means of the Activation Analysis Method. The aim of performed experiments is to check the validity of the model descriptions and of the cross-section libraries used in the corresponding Monte-Carlo simulations of spallation reactions and of the propagation of the produced high-energy neutrons through thick target.

2. Experimental apparatus
The “Energy plus transmutation” installation (figure 1) was irradiated with proton beams of energies of 0.7, 1.0, and 1.5 GeV, see table 1. This assembly was divided into four sections of 114 mm in length separated by 8 mm empty intervals, totally 480 mm (figure 4). Each section
Figure 1. “Energy plus Transmutation” setup (dimensions are in millimeters).

| Parameter                                | 0.7   | 1.0   | 1.5   |
|------------------------------------------|-------|-------|-------|
| Proton energy [GeV]                      | 0.7   | 1.0   | 1.5   |
| Irradiation time [h]                     | 9     | 6     | 12    |
| Beam integral $[10^{13}]$                | 1.47(5)| 3.40(15)| 1.14(6)|
| Beam integral on Pb target $[10^{13}]$   | 1.04(8)| 3.24(15)| 1.10(5)|
| FWHM vertical [cm]                       | 5.9(2) | 4.1(3) | 3.7(5) |
| FWHM horizontal [cm]                     | 5.9(2) | 2.5(3) | 2.4(5) |
| Position vertical [cm]                   | -0.4(9)| 0.2(2) | 0.1(2) |
| Position horizontal [cm]                 | 0.2(2) | 0.0(2) | 0.3(2) |

was composed of a cylindrical lead target (of 84 mm diameter) and of a natural uranium blanket with a hexagonal cross-section (side length of 130 mm). Each of the blanket sections contained 30 uranium rods of 36 mm diameter and 104 mm length. The lead target and the uranium rods were sealed in an aluminium cover of 2 mm and 1.27 mm thick, respectively.

This U/Pb-assembly was fixed on a wooden-metal rack ($362 \times 505 \times 72$ mm$^3$) and surrounded by a biological shielding consisted of a cubic container ($\approx 1$ m side length) filled with granulated polyethylene ($\text{CH}_2)_n$ with an admixture of boron. The inner walls of the container were coated with a 1 mm thick Cd layer (used for absorption of thermal neutrons). The front and the back ends of the setup were without any shielding. The polyethylene moderated high-energy neutrons outgoing from the setup and then partly scattered them back. Herewith, the moderator created a homogeneous field of neutrons with energies $1 \text{ eV} < E < 0.1 \text{ MeV}$ inside the container.
(see figure 2) and, thus, disallowed the study of spatial distribution of low-energy neutrons produced in the target. The influence of the shielding on high-energy neutron component \((E_n > 1 \text{ MeV})\) was negligible (see figure 3).

**Figure 2.** Influence of the polyethylene container and the Cd-layers on neutron spectra at \(X = 11.8 \text{ cm}, R = 3 \text{ cm}\) (MCNPX simulation of 1.5 GeV experiment).

**Figure 3.** Ratio of neutron fluxes (from high-energy region of figure 2) between the whole setup (Pb+U+Cd+(CH₂)n) and the setup only with Pb+U.

The spatial distribution of the produced neutron field was measured by the Activation Analysis Method using radio-chemical sensors placed in the gaps between sections of the U/Pb-assembly (figure 4). Used sensors were packed into sandwiches compound of \(^{27}\text{Al}, ^{197}\text{Au}, ^{209}\text{Bi}, \text{and} ^{59}\text{Co}\) samples. Aluminium and gold sensors had the \(20 \times 20 \text{ mm}^2\) square shape (of \(\approx 0.5 \text{ g}\) and \(0.3 \text{ g}\) weight, resp.), bismuth sensors had the \(25 \times 25 \text{ mm}^2\) square shape (of \(\approx 6 \text{ g}\) weight), and cobalt sensors had the circular shape with the diameter of \(10 \text{ mm}\) (of \(\approx 3 \text{ g}\) weight). The first set of sandwiches was placed at a radial distance \(R = 3 \text{ cm}\) from the target axis at five longitudinal distances \(X = 0.0, 11.8, 24.0, 36.2, 48.4 \text{ cm}\) from the target front. The second set was placed at a longitudinal distance \(X = 11.8 \text{ cm}\) from the target front at four radial distances \(R = 3.0, 6.0, 8.5, 13.5 \text{ cm}\) from the target axis, see figure 4. There is only one exception, in cases of 0.7 and 1.0 GeV experiments, the last sandwich of the radial set was placed not at radial

**Figure 4.** A scheme of placement of activation sensors (dimensions are in millimeters).
distance $R = 13.5$ cm, but at $R = 10.7$ cm.

In the process of irradiation, the stable isotopes of sensor materials were transmuted by $(n,\gamma)$, $(n,\alpha)$, $(n,xnyp)$-reactions into radioactive ones. Studied neutron-induced reactions, both with a threshold in neutron energy and without it, are shown in table 2.

### Table 2. Threshold and non-threshold reactions studied by the Activation Analysis Method.

| Reaction                  | Threshold energy [MeV] | Half-life of product | Reaction                  | Threshold energy [MeV] | Half-life of product |
|---------------------------|------------------------|----------------------|---------------------------|------------------------|----------------------|
| $^{27}$Al$(n,\alpha)^{24}$Na | 5.5                    | 14.959 h             | $^{197}$Au$(n,\gamma)^{198}$Au | -                      | 2.69517 d           |
| $^{209}$Bi$(n,4n)^{206}$Bi | 22.6                   | 6.243 d              | $^{197}$Au$(n,2n)^{196}$Au | 8.1                    | 6.183 d              |
| $^{209}$Bi$(n,5n)^{205}$Bi | 29.6                   | 15.31 d              | $^{197}$Au$(n,3n)^{195}$Au | 14.8                   | 186.10 d             |
| $^{209}$Bi$(n,6n)^{204}$Bi | 38.1                   | 11.22 h              | $^{197}$Au$(n,4n)^{194}$Au | 23.2                   | 1.584 d              |
| $^{209}$Bi$(n,7n)^{203}$Bi | 45.2                   | 11.76 h              | $^{197}$Au$(n,5n)^{193}$Au | 30.2                   | 17.65 h              |
| $^{209}$Bi$(n,8n)^{202}$Bi | 54.0                   | 1.72 h               | $^{197}$Au$(n,6n)^{192}$Au | 38.9                   | 4.94 h               |
| $^{209}$Bi$(n,9n)^{201}$Bi | 61.4                   | 1.8 h                | $^{197}$Au$(n,7n)^{191}$Au | 45.7                   | 3.18 h               |
| $^{59}$Co$(n,\gamma)^{60}$Co | -                      | 5.271 y              | $^{59}$Co$(n,4n)^{56}$Co | 30.9                   | 77.27 d              |
| $^{59}$Co$(n,2n)^{58}$Co | 10.6                   | 70.82 d              | $^{59}$Co$(n,5n)^{55}$Co | 41.2                   | 17.53 h              |
| $^{59}$Co$(n,3n)^{57}$Co | 19.4                   | 271.79 d             |                           |                        |                      |

The activity of the irradiated sensors was measured by HPGe $\gamma$-spectrometers in close geometries because of a low activity of the samples. Each sample was measured few times in order to identify isotopes with different half-lives. The measured $\gamma$-spectra, covering a region approximately from 50 up to 3500 keV, were processed by the DEIMOS32 code [8] that provides a Gaussian fit of $\gamma$-peaks taking into account the background fitted with a parabola. The acquired areas were corrected for coincidence summing effects, for a peak efficiency of the HPGe detector, for beam instabilities during the irradiation, and for decay during irradiation, cooling, and measurement. 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.

### 3. Experimental results

The yields of radioactive isotopes produced in Al, Au, Bi, and Co sensors in units of $[g^{-1} \text{ proton}^{-1}]$ are presented on example of the 1.5 GeV experiment in figures 5-10 (the lines linking experimental points in these figures are delineated to guide readers’ eyes). The trends are identical for all three experiments. Dependencies of yields on the position along the target are given on the left side, dependencies of yields on the radial distance from the target axis are given on the right side of the figures. The delineated errors are only of statistical character. Systematic errors, such as the inaccuracy of the positioning of sensors, contribute about next 5%.

The longitudinal distributions of yields of all isotopes produced in threshold reactions have maximum around 10 cm from the target forehead. However, the neutron field around the setup is a complicated mixture of spallation, fission, moderated and back-scattered neutrons, the isotopes generated in threshold reactions are produced mainly by neutrons isotropically evaporated in spallation reactions. Thus, one could expect the maximum should be in the centre of the target.
But because of ionization losses and out-scattering of protons, the primary proton energy along the target and the intensity of the primary proton beam decrease. Therefore, the maximum intensity of the fast neutron field is shifted from the centre to the target’s front.
The radial distributions of yields of all isotopes produced in threshold reactions decrease, as the intensity of spallation part of neutron spectra falls down with growing distance from the spallation target.

The production maximum of isotopes $^{198}$Au and $^{60}$Co produced in $(n,\gamma)$-reactions is evidently not so sharp in comparison with those produced in threshold reactions (in longitudinal as well as in radial distributions). The reason can be the different course of cross-sections of neutron capture contrary to cross-sections of threshold reactions. The polyethylene shielding partly moderated high-energy neutrons outgoing from the setup and could scattered low-energy neutrons back. Herewith, a homogeneous field of low-energy neutrons was created and this field gives constant contribution to the production of $^{198}$Au and $^{60}$Co in the whole setup, what explains a very flat shape of their distributions.

4. Calculations
Monte-Carlo simulations of the neutron production in the setup and of the activation reactions in the sensors were performed by MCNPX 2.4.0 code [9]. The intranuclear cascade of spallation reactions was simulated using Bertini INC model, the Multistage Pre-equilibrium Exciton Model was used for pre-equilibrium emission of particles (only nucleons and charged pions were taken into account). Simulations were done using SSW (Surface Source Write) card, neutron spectra $\Phi_n(E)$ were counted with HTAPE3X program, see figure 11. The calculated yields of produced nuclei were determined (using F4 tally with the FM multiplier card) by the convolution of the simulated neutron spectra $\Phi_n(E,r,z)$ with the corresponding cross-sections $\sigma_n(E)$ (inbuilt in MCNPX from ENDF/B-VI library [7]):

$$N_{\text{yield}}(r,z) = \frac{1}{A_r m_u I(p)_{\text{sim}}} \int_0^{\infty} \Phi_n(E,r,z)\sigma_n(E)dE,$$

(1)

where $A_r$ is specific atomic mass of a chemical element from which the sensor is made, $m_u$ is unified atomic mass unit, and $I(p)_{\text{sim}}$ is the number of simulated incident protons ($3 \times 10^6$ for all cases). Apparently, the energetic spectrum is harder at the end of the target when compared to its beginning, see figure 12.

The simulations describe the shape of longitudinal distribution of yields very well, see examples of $^{196}$Au, $^{194}$Au in figures 13, 15. A quantitative agreement between experimental and simulated values is worse, the differences reach about 60 %, but it is still quite nice (compare with [10]). Too high ratios (experimental yields divided by simulated ones) in front of the target in the case of 0.7 GeV experiment are the only exception. This was probably caused by worse focusing of the proton beam (see table 1) that hit directly sensors in front of the target.
and caused on the $^{197}$Au-sensor parasitic reactions (e.g., $(p,\text{np})^{196}\text{Au}$, $(p,d)^{196}\text{Au}$, $(p,p3n)^{194}\text{Au}$, $(p,d2n)^{194}\text{Au}$, $(p,\text{tn})^{194}\text{Au}$). Unfortunately, MCNPX does not contain cross-section libraries for these reactions. Therefore, it will be necessary to find these cross-sections $\sigma_p(E)$ in other libraries and to convolute them with simulated proton spectra $\Phi_p(E, r, z)$:

$$N_{\text{yield}}(r, z) = \frac{1}{A_r m_u I_p}\int_0^\infty [\Phi_n(E, r, z)\sigma_n(E) + \Phi_p(E, r, z)\sigma_p(E)]dE.$$ (2)

Regarding radial distributions, the trends of experimental data and simulations are in perfect agreement for the 0.7 and 1.0 GeV experiments, see examples of $^{196}\text{Au}$, $^{194}\text{Au}$ in figures 14, 16. Discrepancies in absolute values do not exceed 50%. Absolutely different situation is for the experiment with the highest beam energy 1.5 GeV, the discrepancy between the experimental and simulated values increases quickly with growing perpendicular distance from the target axis up to more than two times. It is planned to test other INC models (as Isabel or CEM) included in MCNPX, how they influence calculated neutron spectra [11] and if they are able to describe better neutron production in the used setup.

**Figure 13.** Comparison of experimental and simulated yields of $^{194}\text{Au}$ in longitudinal direction.

**Figure 14.** Comparison of experimental and simulated yields of $^{194}\text{Au}$ in radial direction.

**Figure 15.** Comparison of experimental and simulated yields of $^{196}\text{Au}$ in longitudinal direction.

**Figure 16.** Comparison of experimental and simulated yields of $^{196}\text{Au}$ in radial direction.
5. Plans for future

Goals for near future are to carry out experiments with

- higher beam energies (2.0-3.0 GeV),
- different beam particles (deuterons),
- new setups (a usage of graphite as a moderator - GAMMA MD installation, extensions of Pb-target and U-blanket, a usage of Pb+Bi eutectics as a target), and
- another activation sensors (In, Ta).

Moreover, the possibility of using helium-jet transport [12] of fission-product radioisotopes is being investigated. Helium-jet transport systems are able to quickly transfer short-lived isotopes, with help of a helium flux, from their source to detector.

The intensive 660 MeV proton beam \((10^{13} \text{protons/s})\) from the Dubna Phasotron is planned to be used for irradiation of a thick Pb target. The U-foils (with a thickness of 6 mg cm\(^{-2}\)) in a cell on the target surface will be irradiated by secondary neutrons. Fission products (beside Xe, Kr) will be caught up by chemical filter on the exit of the cell. The flux of helium will transport Xe and Kr from the cell into a Cu-pipe cooled in liquid nitrogen \((T_{\text{boil}} = 77 \text{ K})\), where Xe \((T_{\text{boil}} = 165 \text{ K})\) and Kr \((T_{\text{boil}} = 120 \text{ K})\) will be frost. The aim is to measure the production of Xe and Kr isotopes, of their daughter’s products, and of fission products from the chemical filter.

6. Conclusion

The high energy neutron production in spallation reactions of protons with energies of 700, 1000, and 1500 MeV in a thick, lead target with the uranium blanket surrounded by the polyethylene moderator was studied. The shape and the intensity of the produced neutron field were measured by the Activation Analysis Method.

Due to the hard part of the neutron spectrum in the U/Pb-assembly, isotopes produced in \((n,xn)\)-reactions (the emission of up to \(x = 9\) neutrons) with high threshold energy (up to \(\sim 60 \text{ MeV}\)) were observed. The maximum intensity of the fast neutron field \((E_n > 1 \text{ MeV})\) produced in the spallation target is located in the region around 10 cm from the target forehead. The energetic spectrum becomes harder at the end of the target.

MCNPX describes qualitatively very well the shape of the spatial distribution of yields of produced isotopes, the maximum difference in absolute values reaches \(\sim 60\%\). The exception is the 1.5 GeV experiment, in this case the MCNPX simulations underestimate the isotope production with growing radial coordinate, especially behind the uranium blanket (more than two times). The including of proton interactions or usage of more precise intra-nuclear cascade models are expected to reduce observed discrepancies.

Acknowledgments

The authors would like to thank to the Veksler and Baldin Laboratory of High Energies at JINR for the possibility of using the Nuclotron. This work was carried out under support of the Grant Agency of the Czech Republic (grant No. 202/03/H043) and under support of Grant Agency of the Academy of Sciences of the Czech Republic (grant No. K2067107).

References

[1] Brandt R et al 2004 Kerntechnik \textbf{69} 37-50
[2] Henzl V et al 2001 J. Nucl. Sci. and Techn., Proc. Intern. Conf. on Nuclear Data for Science and Technology \textit{ND 2001} (Tsukuba) vol 2 pp 1248-51
[3] Krivopustov M I et al 1997 J. Radioanalytical and Nuclear Chemistry \textbf{222} 267-70
[4] Krivopustov M I et al 2004 Preprint JINR E1-2004-79
[5] Qtool: Calculation of Reaction Q-values and Thresholds 2005 [http://t2.lanl.gov/data/qtool.html]
[6] Audi G et al 1997 Nuclear Physics A \textbf{624} 1-124
[7] Evaluated Nuclear Reaction Data File (ENDF) 2005 [http://www.nndc.bnl.gov/endf]
[8] Frána J 2003 J. Radioanal. and Nuclear Chemistry 3 257 583-7
[9] Waters L S 2002 LANL report LA-CP-02-408 (Los Alamos)
[10] Krása A et al 2005 Proc. Intern. Conf. on Nuclear Data for Science and Technology ND 2004 (Santa Fe) vol 769 pp 1555-9
[11] Majerle M et al 2005 this proceedings
[12] Jungclas H et al 1976 Nucl. Instr. and Meth. 137 93-8