Experimental Studies of the Spatial Distribution of Neutron Production Around a Thick Lead Target Irradiated by 0.9 GeV Protons

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Abstract: This study is part of a complex research of Accelerator Driven Transmutation Technologies (ADTT) carried out by a collaboration of the NPI ASCR in Řež with the JINR in Dubna. The aim of the experiment was to check the validity of the model descriptions and the cross-section libraries used in the corresponding Monte-Carlo simulations of spallation reactions, and the propagation of the produced high-energy neutrons passed through a thick target. The experiment was carried out at the synchrophasotron of the Laboratory for High Energies at the JINR. Relativistic protons with an energy of 885 MeV interacting with a massive cylindrical lead target produced the spallation neutrons. The spatial and energetic distributions of the produced neutron field were measured by the activation of Al, Cu and Au foils placed on the surface of and next to the target. The HPGe detectors then measured the activity of the foils. The resulting \(\gamma\)-spectra of the activated foils were analysed, the yields of the corresponding radioactive nuclei were determined, and compared with Monte-Carlo based simulations performed both with the LAHET+MCNP code and the MCNPX code.

1. INTRODUCTION

1.1. Motivation and aim

Spallation reactions can be used to produce high neutron fluxes by bombarding a thick, heavy target with a high-intensity relativistic proton beam. Recently, possible applications, such as accelerator-driven transmutation of nuclear waste\textsuperscript{2}, have increased the interest in spallation reactions and in the transport of the produced neutrons. The idea of this work consists in the experimental study of the spatial and the energetic distributions of the neutron field (we concentrated our attention mainly on high-energy neutrons) produced in the spallation reactions of high-energy protons on a thick lead target. The main aim is a so called benchmark test – comparison between experimental data and values obtained from the corresponding simulation codes, which must be able to describe the course of the spallation, interactions of secondary particles, and neutron transport through the target material.

1.2. Simulation codes

There are several simulation codes and combinations of these codes describing spallation reactions and the following transport of neutrons. We used a combination of two simulation codes – LAHET and MCNP. They are based on the Monte-Carlo method, and they use various physical models of spallation reactions and cross-section libraries of neutron induced reactions with nuclei.

LAHET\textsuperscript{3} \{Los Alamos High Energy Transport Code\} can model spallation reactions, transport of particles and high-energy neutrons (\(E > 20\) MeV). LAHET generates cross sections for individual processes.

MCNP\textsuperscript{4} \{Monte Carlo N-Particle Transport Code\} is able to model the transport of neutrons in an energy range 10\textsuperscript{-11} MeV < \(E < 20\) MeV. It uses libraries of evaluated data as a source of the cross sections.
One of the newest simulation codes is MCNPX\textsuperscript{3}, which links the advantages of both LAHET and of MCNP, and exploits libraries of evaluated cross sections up to 150 MeV.

2. **EXPERIMENT**

2.1. Spallation target and experimental set-up

We carried out systematic measurements of the neutron field (and optionally of iodine\textsuperscript{6} transmutation) in different set-ups and at different beam energies at the synchrophasotron in Dubna. This paper reports on an experiment, with a beam of 885 MeV protons hitting a multi-section massive cylindrical lead target (diameter $d = 9.6$ cm and total length $l = 50$ cm) surrounded by a box of expanded polystyrene ($17.6 \times 17.1 \times 52.6$ cm\textsuperscript{3}) that worked as a thermal isolation to allow the measurement of the heat production. A box of granulated polyethylene ($100 \times 100 \times 100$ cm\textsuperscript{3}) surrounded this all. The polyethylene moderated the high-energy neutrons; hence they did not come back after scattering on surrounding material, and thus scattered neutrons did not induce threshold reactions.

The primary protons with an energy of 885(5) MeV were stopped after passing about 30 cm in the target. Further part of the target was influenced only by the shower of secondary particles, which consists mainly of neutrons. The irradiation continued for about two hours. The number of beam protons was $3.6 \times 10^{13}$ as determined by the activation method (see chapter 2.3.).

2.2. Activation analysis method

The spatial distribution of the produced neutron field was measured by the activation of Al, Cu and Au foils placed on the surface of and next to the target. We study neutron-induced reactions both with a threshold in neutron energy and without it, see Table 1.

<table>
<thead>
<tr>
<th>reaction</th>
<th>$^{19}$Au (n,2n) $^{19}$Au</th>
<th>$^{19}$Au (n,4n) $^{19}$Au</th>
<th>$^{19}$Au (n, $\gamma$) $^{19}$Au</th>
<th>$^{27}$Al (n,$\alpha$) $^{24}$Na</th>
<th>$^{63}$Cu (n, $\gamma$) $^{64}$Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td>threshold energy</td>
<td>8.5 MeV</td>
<td>24.5 MeV</td>
<td>without threshold</td>
<td>5.5 MeV</td>
<td>without threshold</td>
</tr>
</tbody>
</table>

The advantage of the activation-analysis method is that detectors can be simple and can have arbitrary format (we used foils $2 \times 2$ cm\textsuperscript{2} with 50 $\mu$m thickness), and it is possible to place them at any position of the set-up. The disadvantage is that we measure the amount of produced radioactive nuclei, from which it is not always straightforward to determine the incident neutron field.

The foils were located closely above the target (exactly 5 cm above the target axis), closely above the polystyrene box (9.3 cm above the target axis) and on the right side of the polystyrene box (9.1 cm to the right from the target axis). The foils were located all along the target in order to determine the spatial distribution of the neutron field, see Fig.1.

The foils were then measured by the High-Purity Germanium (HPGe) $\gamma$-spectrometers. These measurements were accumulated in histograms with 8192 channels, which were processed by the DEIMOS32\textsuperscript{7} code. The acquired areas were corrected for coincidences and other standard decay effects.
2.3. Determination of beam intensity and beam position

We also studied the beam geometry with the use of high-energy proton reactions on Cu and Au (production of \( ^{48}\text{V} \), \(^{52}\text{Mn} \), \(^{58}\text{Co} \), \(^{44}\text{Sc} \), \(^{47}\text{Sc} \), \(^{191}\text{Pt} \), \(^{74}\text{As} \)). We placed a group of five Cu and Au foils closely in front of the target and compared the yields in different foils. Based on the trace of the beam in a Polaroid foil, we used three assumptions:

− the central foil is fully covered,
− the proton distribution is homogenous,
− the beam has circular cross section.

We found out that the beam was shifted \((0.8 \pm 0.3) \) cm down and \((0.8 \pm 0.3) \) cm right from the target axis and that the beam radius was \((3.5 \pm 0.3) \) cm.

The total beam intensity was determined by activation of beam monitors (composed of \(10\times10 \) cm\(^2\) Cu foils with a thickness of 25 \(\mu\)m and Al foils with a thickness of 100 \(\mu\)m) placed 30 cm ahead of the target. In particular, we measured the yields of the reactions \(^{27}\text{Al}(p,3pn)\text{P}^{24}\text{Na} \), \(^{27}\text{Al}(p,X)\text{Be} \) and \(^{\text{nat}}\text{Cu}(p,X)\text{P}^{24}\text{Na} \). The total proton flux determined by the activation was \((3.6 \pm 0.3)\times10^{13}\) (the error includes statistic and systematic errors of the applied cross sections, the calibration and the thickness of monitors). This value seems to be more reliable than the value \(4.3\times10^{13}\) determined by the current integrator, which suffered from a significant systematic error.

2.4. Production of \(^{196}\text{Au} \), \(^{194}\text{Au} \), \(^{24}\text{Na} \) in foils along the target

The yields (number of activated nuclei per gram of activated material and per incident proton) of threshold reactions in gold and aluminium foils are presented as a function of the position along the target in Fig.2. Neutrons produce the main part of radioactive nuclei; most of them are emitted isotropically in spallation reactions. The shape of the yield distribution reflects the interplay of two main processes. First, the spallation cross section of protons decreases along the target in relation with the decrease in the primary proton energy due to the ionisation looses. Second, the intensity of the primary proton beam decreases too, as part of the protons is scattered out. Consequently, the maximum intensity of the fast neutron field is shifted from the centre to the target’s front – to the region between 7 – 11 cm from the target forehead.
3. SIMULATIONS

The processing of the experimental data was accompanied by simulations of the neutron production in spallation reactions. Simulations were performed by LAHET 2.7. (Bertini INC model with preequilibrium phase) and MCNP4B. We made calculations in 2 steps:

– calculation of neutron (proton) energy spectra (Fig.3),
– calculation of the yields of produced nuclei by convolution of these spectra with the corresponding cross-sections.

We made not only simulations with the used experimental set-up, but also simulations with simplified geometries, to thoroughly study the influence of simplification and variation in the set-up geometry (foil positions; the gaps between parts of the segmented target; trajectory, profile and intensity of the proton beam). Since our main focus was on high-energy neutrons, the rest of the article is dedicated to the threshold reactions.

Fig. 3. Spectrum of neutrons and protons, which passed through the foil located closely above the target, 45 cm from the target beginning (simple simulation)

Fig. 4. Influence of proton interactions in the foils – ratio of the amount of nuclei produced by protons to the total amount of produced nuclei, i.e., in proton- and neutron-induced reactions together (LAHET+MCNP simulation)
3.1. Influence of proton interactions in the foils

The studied isotopes $^{24}$Na, $^{196}$Au, $^{194}$Au can be produced not only in neutron-induced reactions, but also in proton-induced reactions. There are two types of protons at the positions of the activation detectors: scattered primary protons and secondary protons from spallation reactions. The share of proton-induced reactions reaches up to several tens of percent at larger distances from the target forehead, see Fig.4. Therefore, it was necessary to include secondary protons in our simulations, too.

3.2. Influence of the beam geometry

We found out by activation analysis that the beam was shifted 0.8 cm down and 0.8 cm right from the target axis, and that the beam radius was app. 3.5 cm. The amount of activated nuclei is sensitive to the beam position with respect to the target axis, see Fig.5. On the vertical axis is the ratio between the number of nuclei produced in the case of a centred beam and the number of nuclei produced in the case of a shifted beam. Of course, the ratios in the foils above the target are greater than unity, because the beam was shifted right and down. The difference between the descriptions with centred and shifted beam is significant, therefore, the influence of the beam geometry is indispensable and it is necessary to take it into account.

3.3. Influence of polystyrene and polyethylene on threshold reactions

Now we will look at the influence of other components of the experimental set-up on the yields of threshold reactions. We compared simple simulations (which take into account only the target) with full simulations (which take into account all components, e.g., polystyrene and polyethylene). The simple and the full simulations in the foils closely above the target differ only at the end of the target by about 10% (Fig.6). At larger perpendicular distance, there is a decrease of only about 2–5% along the target. It is comparable to the experimental errors and hence simple simulations are applicable. In this way, by application of the simple simulations, we can obtain better statistics.
4. **COMPARISON BETWEEN SIMULATIONS AND EXPERIMENT**

The main point of our study is the comparison between the experimental results and the simulations. In the case of threshold reactions we compared the experimental values and the values from simple simulations. The simple simulations describe the shape of the spatial distribution very well until the distance of 40 cm from the beginning of the target (Fig.7). The maximum difference in absolute values is about 20%. Beyond 40 cm, the simulation underestimates the experiment and the ratio of experimental values to the simulated ones reaches two.

Neither the usage of the simple simulation (see Fig.6) nor the influence of the beam geometry (see Fig.5) can explain this discrepancy. We increased the beam energy by the given inaccuracy of 0.5 %, then we increased the gaps between the segments of the target and/or changed the shift of the beam position with respect to the target’s axis. None of these changes had significant impact on the simulated yields. Moreover, they do not go in the direction needed.

The range of 885 MeV protons in lead is about 30 cm, hence the primary proton beam should cease out behind that point. The same holds for secondary protons produced from primary spallation reactions, as their energy is not greater than the energy of corresponding primary proton. Therefore, we concluded that the simulations underestimate the development of the shower produced by high-energy secondary neutrons and their interactions within the target.

4.1. **LAHET+MCNP vs. MCNPX**

We made all the above-mentioned simulations by a combination of the LAHET and the MCNP codes. As the last step, we tried to change the simulation code and we made simulations by the MCNPX code with several INC models (Bertini, Isabel, CEM). The final result is that, in our case, the results of these codes do not differ significantly, therefore, new code is also not able to explain the discrepancy between the simulation and the experiment at the end of the target.

5. **CONCLUSIONS**

We studied the neutron production in the reaction of relativistic protons in a thick lead target. The shape and the intensity of the neutron field was measured by the activation analysis.

We found out that the beam geometry and the secondary protons have important influence on the activation yields (~ 10 %). The secondary protons and the non-central beam trajectory must be included into the simulations.

We also found out that we can neglect the influence of polyethylene and polystyrene on the production of high-energy neutrons. Their influence is smaller than the experimental errors – we can include into the simulations only the spallation target and compare our data with other simple spallation-target simulations.

We reached good agreement between experiment and simulations for high-energy neutron production. The two times higher experimental neutron intensity near the end of the target indicates a difference between the development of the secondary-particle shower in the real experiment and in the
model used in the simulations. The difference between the values from LAHET+MCNP and MCNPX is not significant in our case.

A further detailed analysis of the sources of the differences between experiment and simulation are in progress. We plan also to carry out a comparison with experiments with different proton energies and set-ups.

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