EXPERIMENTAL STUDIES OF TRANSMUTATION OF $^{129}$I BY SPALLATION NEUTRONS USING JINR DUBNA PHASOTRON


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Abstract

An intensive beam of 660 MeV protons was directed towards a lead target (not surrounded by shielding or neutron reflectors) for 10 minutes. On the top of the target were placed detectors and iodine samples. The neutron field and the transmutation of $^{129}$I were studied by the neutron activation analysis method. First simulations were compared to the experimental data.

1 Description of the experiment

The collaboration ”Energy plus transmutation” works on benchmark experiments intended to test simulation programs, which shall be used in design of Accelerator Driven Systems. The work consist of a series of similar experiments, of which each is concentrated on a specific topic. Generally said, we study production of neutrons by spallation reactions on massive targets, their transport, and use for transmutation. Our experimental setups consist of heavy metal (Pb, W, Bi, U) targets of cylindrical shape to which a narrow beam of relativistic protons is directed. In the target protons cause spallations of heavy nuclei to nucleons, and smaller fractions of nucleus. Relativistic nucleons cause further spallations, and mostly neutrons of all energies - from thermal up to the energy of the proton beam - exit the target sideways. Such neutron production shall be used in the devices called ADS (Accelerator Driven Systems), which will be able to use a wider spectrum of nuclear fuel ($^{232}$Th, nuclear waste from thermal reactors), and their waste should be much less radioactive than the waste from thermal reactors. These systems cannot be built in smaller scale as a test model, and the accelerators to be added to the reactors will not be a cheap extension. That is why we need the precise descriptions of what will be going on in these systems. Our experiments are simplified setups of them, and are used as benchmark tests for the computer simulations that will be used to describe larger and more complicated systems.

One of the experiments was held on Dubna Phasotron in December, 2003, and was focused on the studies of short-lived iodine isotopes production. We needed to make our measurements as soon as possible after the irradiation, and use an intensive proton beam as possible. The proton beam was provided by Dubna Phasotron, which accelerated the intensive proton beam to the energy 660 MeV. The beam was directed to the bare lead
target, which was composed of four cylinders \((r = 4.8 \text{ cm}, d = 12 \text{ cm})\). Before the target were placed two metal (Al and Cu) monitor foils to determine the proton beam integral. Between the first and the second cylinder there was a 7 mm empty interval, where were placed 5 detectors to determine the dislocation of the beam from the central axis. Other detectors (Al, Au, and Bi foils of dimensions \(2 \times 2 \text{ cm}^2\)) were put on the top of the target along its entire length. At the 9th cm, we put two iodine samples, one of natural iodine \(^{127}\text{I}\), and the other of iodine from radioactive waste (mixture of 10% \(^{127}\text{I}\) and 90% \(^{129}\text{I}\)). Two identical samples were put at the 21st cm. The iodine was packed in Al cases for safety reasons. Similar samples were already used in previous experiments [1, 2], and the aluminum shielding was remodeled for this experiment: to provide reasonable safety at the minimum of Al used (Al is activated during the irradiation, and the radiation of its products adversely affects the measurements). After 10 minutes of irradiation with the intensive proton beam, the detectors and samples were collected from the setup, and the measurements with three HPGe detectors started. The \(\gamma\)-spectra of foils were measured at least twice, firstly soon after the irradiation for a short time, to detect short-lived isotopes, and secondly for a longer time, to accurately detect the amount of produced longer lived isotopes. The iodine samples were measured more times (10 times). Finally the spectra of calibration samples were measured to determine the efficiencies of the detectors.

With the analysis of these spectra we determined: the yields of produced isotopes in the monitor foils, in the neutron detectors, and in the iodine samples, and we further deduced the parameters of the proton beam, the neutron spectrum and distribution, and the production rates of transmutation of \(^{129}\text{I}\) to shorter lived isotopes.

2 Experimental data

2.1 Longitudinal neutron field

For calculations concerning neutron production, we needed to know the parameters of the proton beam: the integral of the proton flux (measured with Al and Cu foils of \(8 \times 8 \text{ cm}^2\) at the front of the target), and the displacement of the beam from the central axis (measured with five Al and Au foils of \(2 \times 2 \text{ cm}^2\) inserted after the first segment of the target).

During the irradiation, the beam intensity and size were measured by wire chamber: \(10^{13}\) protons/s, the horizontal and the vertical diameters of the beam were 1.6 cm - 1.9 cm. The beam profile had a Gaussian profile, so the word "diameter" does not define the exact beam borders but the limit, where we can find most of protons. Our beam was focused enough that all of the protons passed through \(8 \times 8 \text{ cm}\) foils before continuing to the target. In monitor foils, protons produced radioactive isotopes, which were later detected by the means of the \(\gamma\)-spectroscopy. The library of the experimental cross-sections for reactions with protons of energy 660 MeV is voluminous and precise, and the results calculated from different produced elements were in good concordance with each other, and with the expected number of protons. Through the monitor foils passed \(1.58 \times 10^{15}\) protons, the accuracy of this result is 6% (included are systematic and statistic errors), which is in accordance with the given intensity multiplied by the time of irradiation.

Two sets of five \(2 \times 2 \text{ cm}^2\) foils were placed after the first segment of the target, one foil on the central axis, and four others around it with the space 0.5 cm between their sides.
In the left and in the right foil the numbers of produced elements were 3 times smaller than in the central foil, in the lower foil it was 5 times smaller, and in the upper foil it was smaller by a factor of 0.8. This agrees with the results measured by the wire chambers: the beam diameter was roughly limited to 2 cm. From the results in the upper and lower foils, we concluded that the center of the beam was shifted upwards for cca 1 cm.

Three types of detector foils were used to study longitudinal neutron field: Al, Au \((2\times2 \text{ cm}^2)\) and Bi \((2.5\times2.5 \text{ cm}^2)\) foils in the square shape placed on the top of the target along its length. In the foils, neutrons produced radioactive isotopes via reactions \((n,\alpha)\), \((n,\gamma)\) or \((n,xn)\) (see Table 1). After the irradiation, we measured the \(\gamma\)-spectra of the foils, and could calculate the yields of produced isotopes.

<table>
<thead>
<tr>
<th>Material</th>
<th>Reaction</th>
<th>Product</th>
<th>Threshold (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{27}\text{Al})</td>
<td>((n,\alpha))</td>
<td>(^{24}\text{Na})</td>
<td>3.24996</td>
</tr>
<tr>
<td>(^{197}\text{Au})</td>
<td>((n,\gamma))</td>
<td>(^{198}\text{Au})</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>((n,2n))</td>
<td>(^{196}\text{Au})</td>
<td>8.11245</td>
</tr>
<tr>
<td></td>
<td>((n,3n))</td>
<td>(^{195}\text{Au})</td>
<td>14.78950</td>
</tr>
<tr>
<td>(^{209}\text{Bi})</td>
<td>((n,4n))</td>
<td>(^{206}\text{Bi})</td>
<td>22.55228</td>
</tr>
<tr>
<td></td>
<td>((n,5n))</td>
<td>(^{205}\text{Bi})</td>
<td>29.62538</td>
</tr>
<tr>
<td></td>
<td>((n,6n))</td>
<td>(^{204}\text{Bi})</td>
<td>38.13866</td>
</tr>
</tbody>
</table>

Table 1: Some reactions and their thresholds for Al, Au, and Bi foils [3].

The yields of produced isotopes were recalculated to the number of produced atoms of isotope \(A\) per 1 gram of the detector material and per 1 incident proton. This value is called production rate \(B(A)\) [4, 5], and is defined as:

\[
B(A) = \frac{\text{(number of } A \text{-atoms produced)}\}{(1 \text{ g sensor})\cdot(1 \text{ primary proton})].}
\]

In the figures (Fig. 1 and 2) are plotted the experimental production rates against the position along the target for all three types of detector foils (errors at the graphs are only statistic errors; systematic errors, such as imprecisely placed foils in the detectors, should contribute another 5%). All graphs show a specific shape: the maximum at around 10\(^{th}\) cm, and the point near 30\(^{th}\) cm, from where the neutron field starts to decrease faster - the particles lost so much energy by that point that they cannot cause further spallations. Moreover, the graphs of the elements produced with higher energy neutrons show a slower falling trend - higher energy neutrons are directed mostly in a forward direction, and cause the hardening of the neutron spectrum approaching the end of the target. The exception is the graph for \(^{198}\text{Au}\), which is produced via \((n,\gamma)\) reaction by low-energy neutrons (thermal, epithermal and resonance neutrons). The neutrons from the target were partly reflected back by concrete walls, resulting in a homogenous low-energy neutron field in the place with the target, which is seen as flat distribution of production rates of \(^{198}\text{Au}\).

### 2.2 Transmutation of iodine

Our basic interest here was to observe and measure the production rates of higher order reactions - \((n,5n), (n,6n),...\) in iodine samples. Because the isotopes produced via such
reactions are far away from the line of stability and have short lifetimes, we needed to use a series of prompt measurements immediately after the experiment. We could determine the yields of produced isotopes up to $^{118}$I with good accuracy (10%), and detect the products decayed from iodine isotopes up to $^{116}$I, which will be used to calculate the yields of produced iodine isotopes to the isotope $^{116}$I. The yields of produced isotopes for $^{129}$I were calculated with the substraction of $^{127}$I contribution in the samples with the mixture of radioactive and stable iodine.

The graphs in Fig. 3 show the production rates of measured iodine isotopes at the 9th cm and the 21st cm for $^{127}$I and $^{129}$I. The production rates for iodine isotopes are comparable with each other and with the production rates for other elements ($10^{-6}$ g$^{-1}$proton$^{-1} > B > 10^{-7}$ g$^{-1}$proton$^{-1}$).

### 3 MCNPX calculations

We have compared the experimental data with the calculations done by the simulation code MCNPX v2.3.0. MCNPX simulates the exact paths of incident particles through the setup, simulates nuclear reactions, and stores the history of the particles. From this history, we can extract neutron and proton spectra along the target (Fig. 4). If we convolute the calculated spectrum with the cross-sections for nuclear reactions, we can

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**Figure 1**: $B$-values for $^{24}$Na in Al foils along the target, and comparison with calculated values (MCNPX).

**Figure 2**: $B$-values for different isotopes in Au and Bi foils along the target.
calculate the production rates for our detectors, and compare them with experimental values (Fig. 1).

The calculations agree well with the experimental data (discrepancies do not exceed 20%). One general discrepancy of the MCNPX simulations is that they underestimate the neutron field after the 40th cm of a target [6], what was also our case.

Our calculations differed from experimental values in two more points. The field of low-energy neutrons is not correctly calculated, probably because in the calculations were not included the concrete walls, where neutrons from the target were moderated and partly reflected. Also, the calculated production rates for high-energy neutrons were systematically higher from the experimental values by a factor 1.2. This is supposed to be due to the dislocation of the beam to the upper part of the target, where monitor foils were placed. New simulations shall include these two corrections.

Figure 4: Longitudinal neutron spectra calculated with MCNPX. The graph on right shows the neutron energy range from 0-40 MeV in detail.

4 Conclusion

We obtained a lot of experimental data from the Phasotron experiment, and found out that it corresponds well to the expectations. This experiment is going to be a good tool to test the simulation codes. The calculations agree to the experimental data inside 20%, and we are trying to improve this agreement with a more accurate definition of the experiment...
in MCNPX. In the iodine samples, we measured the production rates for the reactions up to \((n,9n)\), and we are determining the production rates of higher order reactions with the analysis of the decay products of produced iodine isotopes.

The benchmark experiments done on Dubna accelerators have become routine experiments already. Each half a year, a new experiment is done, and we try to provide as much various experimental data as possible. Our future plans include targets from different materials (Bi/Pb), minor modifications of the setup, different energy and intensity of the beam, and different beam particles. Currently we are working on the experiment with deuterons from Nuclotron impinging upon the "Energy plus transmutation" setup. An another experiment is planned on Phasotron as well.

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References


