STUDIES ON THE SPATIAL DISTRIBUTION AND NUCLEAR WASTE TRANSMUTATION CAPABILITIES OF THE SPALLATION NEUTRONS IN THE JINR “ENERGY PLUS TRANSMUTATION” SETUP


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Abstract

The “Energy plus Transmutation (EPT)” experimental setup of the Joint Institute for Nuclear Research (JINR), Dubna, Russia is composed of a lead target surrounded by natural uranium blanket of mass 206.4 kg. The target was irradiated with protons of different energies using the Nuclotron accelerator of the JINR. The special distribution of the neutrons in this assembly was studied experimentally using particle track and radiochemical techniques. The fission rate in the assembly was determined via calibration of the solid state track detectors in well known neutron fields. The nuclear waste transmutation capabilities of the neutron fields in the EPT-setup were determined at proton energy of 1.5 GeV. The experimental results are compared with Monte Carlo calculations using the MCNPX code.

1. Introduction

Accelerator driven systems (ADS) are considered to be one of the best options for cleaner, safer and economically viable methods for future nuclear energy production and nuclear waste incineration [1-3]. In such systems the large number of neutrons (spallation neutrons) that result from the interaction of high-energy ions (e.g. protons) with massive extended targets such as lead, are used to operate sub-critical nuclear assemblies. The chain reaction in an ADS is sustained by continuous operation of the driving accelerator and the system remains sub-critical at all time. These spallation neutrons have an energy spectrum covering a very wide energy range of keV to GeV (see e.g. [4]).

Determination of fission-rate in an ADS is essential for output power and effective neutron multiplication calculations. The latter is one of the most important parameters of the ADS which is aimed to operate under sub-critical conditions. In this paper determination of fission rate of natural uranium and transmutation of some nuclear waste nuclei in the fast neutron field of an experimental subcritical assembly known as “Energy plus Transmutation (EPT)” setup are reported.

The experiments described in this paper were carried out in the Veksler and Baldin Laboratory of High Energies (VBLHE), Joint Institute for Nuclear Research (JINR), Dubna, Russia, using the NUCLotron accelerator and EPT setup. The Monte Carlo calculations were performed using the MCNPX 2.5e code [5].

2. Experimental setup

Figures 1a and 1b illustrate the schematic drawings of the EPT installation. Detailed description of this setup is given elsewhere [6] and in this paper only a brief explanation of its components and their arrangements is given;

1) The system contains four cylindrical lead targets each with diameter 8.4 cm and length 11.4 cm.
2) A natural uranium blanket surrounds each of the four target sections. Each uranium blanket is composed of 30 uranium rods of diameter 3.6 cm (including the Al-cladding) and length 10.4 cm hermetically sealed in aluminum cladding. The uranium rods are arranged in the form of hexagonal (triangular) lattice
with pitch size of 3.6 cm. The weight of natural uranium in each blanket section is 51.6 kg and whole setup contains total of 206.4 kg of natural uranium. Each section of target-blanket is safely fixed within a steel right angle hexagonal prism container. The four target-blanket sections aligned, along the Z-axis (the target axis) with 0.8 cm gap between the sections. These gaps are used to place activation foils, track detectors and other sensors used in the study of the neutron field within the system.

3) The whole target-blanket system was placed within a wooden container filled with granulated polyethylene of average density \(0.7 \text{ g cm}^{-3}\) with dimensions and the arrangements as shown in Figure 1.

4) The inner walls of the container were covered with a Cd foil of thickness 1 mm, as shown in Figures 1a and 1b.

5) The whole setup is mounted on a platform that can be moved on a rail and its position on the platform can be adjusted with the help of appropriate screw devices.

![Figure 1. Schematic drawings of the “Energy plus Transmutation” experimental setup. (a) YZ cross-section and (b) XY cross-section.](image)

MCNPX calculation using the KCODE card show that the EPT assembly has an effective multiplication coefficient of \(k_{\text{eff}} = 0.20221 \pm 0.00081\).

3. Determination of natural uranium fission rate

Several experiments have been carried out using EPT setup at different incident proton energies in the range of 0.7 – 2 GeV. In this paper some of the results for 1.5 GeV protons will be given.

In order to study and determine the fission-rate in the EPT setup, metallic foils of natural uranium (diameter of 7 mm and thickness ~0.1 mm) were used in conjunction with track detectors. As the thickness of these foils was greater than the mean range (5.41 \(\mu\)m) of the fission fragments in uranium, the U-foils are considered to be “thick foils”. The U-foils were placed in close contact between two Fluorophlogopite (artificial mica) track detector sheets as shown in Figure 2b.

The U-foil mica sandwiches were mounted on plastic sheets (sample plates) of thickness ~0.2 mm, along the +Y axis at different radial distances \(R\), as shown in Figure 2a. Five plates each containing six samples were placed in front, back and in the three gaps between the target-blanket sections (see Figure. 1a and Figure 2c).

The target was irradiated with 1.5 GeV protons parallel to the target axis (Figure 1a). Total fluence of the protons striking the target during the irradiation was determined as \((1.10 \pm 0.06) \times 10^{13}\) by determination of
$^{24}\text{Na}$ activity in an Al-foil exposed to the primary protons via the $^{27}\text{Al}(p, 3pn)^{24}\text{Na}$ reaction [7,8]. The Al-foil was placed upstream at a distance of ~ 60 cm from the target surface.

After the irradiation the mica track detectors were etched in 7% HF at 60 °C for a period of 10-40 min and fission track density in each sample was determined using optical microscope. The mean value of the track density in two mica detectors of each of the U-foils was used in the further analysis. It is shown that this mean track density is independent of the angular distribution of the fission fragments [9]. Also it is shown that the kinetic energy of the fission fragments does not change with the energy of fission inducing neutrons. The fission track densities were converted to fission rates using a calibration factor ($w = (9.90 \pm 0.03) \times 10^{-5}$ tracks.neutron$^{-1}$.barn$^{-1}$) that was obtained in standard neutron fields [9].

![Figure 2](image)

(a) The schematic drawing of a sample plate on which the activation and track detectors were mounted, (b) natural uranium (fission foil)-mica track detector sandwich and (c) the image of the four blanket sections, outside the polyethylene shielding. In this image the gaps between the blanket sections and sample plates can be seen. On top of the blanket sections activation samples for neutron field monitoring and nuclear waste samples such as $^{129}\text{I}$ and $^{237}\text{Np}$ for transmutation studies are placed.

Figure 3 shows the obtained experimental and MC-results for the sensors placed on all five sample plates. Details of the Monte Carlo (MC) calculations using MCNPX 2.5e code [5], corrections and error analysis is given elsewhere [10]. In MC-calculations the fission induced by neutrons, protons and pions are included.

![Figure 3](image)

Figure 3. Variations of the $^{235}\text{U}$ fission-rate (fission induced by neutrons, protons and pions) as a function of radial distance measured from the target axis. Lines connecting the data points are drawn to guide the eyes.
The deviations between the experiment and calculations at radial distances less than 4 cm are due to underestimation of the track densities at these locations because of overlapping of the tracks due to very high track densities.

Figure 4 shows the variation of the fission rate (experimental and MC-results) with distance along the Z-axis (target axis) for the different radial distance. As it can be seen the trends and shapes of the distributions for experimental and calculated results are similar.

4. Transmutation of nuclear waste and other isotopes

In the EPT setup one primary proton of \( E_p = 1.5 \) GeV produces 50.8 neutrons, 7.2 protons and 0.54 pions. Figure 5a shows the average neutron spectrum on top of the uranium blanket. Figure 5b illustrates the variations of neutron flux on top of the blanket with distance along the target. In Figure 5c an example neutron flux in the gap between the 1\textsuperscript{st} and 2\textsuperscript{nd} blanket sections as function of distance from target axis is shown. In the Figure 5a the neutron energy is given in equal logarithmic energy bins with twenty intervals per decade. We have deliberately chosen rather narrow energy bins so as to make the effects of resonance peaks in the cross-sections of the material used in the transmutation studies more evident. From Figure 5 it is apparent that:

1) Within the uranium blanket and its surrounding volume before the polyethylene shielding the neutron field is fast and negligible amount of thermal neutrons are present in these volumes.
2) The region in the energy interval of 1 eV to 100 keV indicate that resonance region of the reaction cross sections are very well covered.
3) The integrated neutron fluence is 0.0152 neutrons/cm\(^2\)/proton, suggesting that for a proton current of greater than 10 mA (required for a power generating ADS) a neutron flux of \( > 9.5 \times 10^{13} \) will be obtained.

We used MCNPX code to calculate the transmutation rates of the \(^{232}\text{Th}, ^{237}\text{Np}\) and \(^{129}\text{I}\) in the above neutron field (Figure 5a). Transmutation of \(^{232}\text{Th}\) is important for fissile fuel breeding purpose while \(^{237}\text{Np}\) and \(^{129}\text{I}\) represent very long lived nuclear waste nuclei. The (n, \( \gamma \)) transmutation of these isotopes proceeds in the following manner.
Figure 6 shows the calculated transmutation results. Here the transmutation rate is given in units of number of nuclear reactions per atom of the sample per incident proton of energy 1.5 GeV. Also shown in Figure 6 are the variation of the neutron cross sections with neutron energy for some of the reactions and the nuclei that have been investigated.

From Figure 6 it can be seen that for all (n, γ) reactions neutrons of energy less than ~1 MeV have the largest contribution, dominated by the energies corresponding to resonance peaks in the cross-section plots. However for fission reactions neutrons of energy greater than ~1 MeV have a significant contribution. In these calculations the mix and match option of the MCNPX code was used, therefore the reaction cross-sections beyond the neutron energies available in data tables (generally 20 MeV) have been calculated by the code. Table 1 gives the transmutation rates for 127I, 129I, 232Th, 237Np, and 239Pu. In Table 1 the transmutation rate is given as, number of the transmuted nuclei in 1 g of the isotope per incident proton of energy 1.5 GeV (the B-value). In the MC-calculations of 127I, 129I and 237Np neutron spectra at location of these samples on the blanket was used while in the cases of 232Th and 239Pu the average spectrum on to the blanket (Figure 5a) was used.

Table 1. Transmutation rate (B-value) of some isotopes in the neutron field of the EPT setup. See the text for details of the rest of the data given in the table.

<table>
<thead>
<tr>
<th>Transmuted isotope</th>
<th>MC-Results* (n, γ) x10^6</th>
<th>Experimental results (n, f) x10^6</th>
<th>Mean cross-section (b)</th>
<th>σθ / &lt; σ &gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>127I</td>
<td>95.3</td>
<td>-</td>
<td>93 ± 7</td>
<td>0.76</td>
</tr>
<tr>
<td>129I</td>
<td>58.84</td>
<td>-</td>
<td>36.0 ± 1.9</td>
<td>0.39</td>
</tr>
<tr>
<td>232Th</td>
<td>62.3</td>
<td>2.0</td>
<td>-</td>
<td>1.57</td>
</tr>
<tr>
<td>237Np</td>
<td>292.1</td>
<td>27.0</td>
<td>378 ± 18</td>
<td>3.77</td>
</tr>
<tr>
<td>239Pu</td>
<td>66.3</td>
<td>209.9</td>
<td>-</td>
<td>1.0</td>
</tr>
</tbody>
</table>

* The statistical errors of the MC-results are less than 1%

As it can be seen the experimental and calculated results for 127I(n, γ) reaction are in total agreement while in the cases of the 129I and 237Np the agreement is not as good (The reasons for such deviations are under...
investigation). In Table 1 also the mean cross-sections $<\sigma>$ for transmutation of the relevant isotopes in the EPT neutron field are given. The $<\sigma>$ was calculated using the following equation

$$<\sigma> = \frac{\int_0^\infty \phi(E)\sigma(E)dE}{\int_0^\infty \phi(E)dE}$$

Where $\phi(E)$ and $\sigma(E)$ are the energy dependent neutron flux and neutron cross-section respectively. The ratio of $\sigma_{th}/<\sigma>$ represents ratio of the transmutation rate in thermal neutron field to that of fast neutron field of the EPT setup. It is evident that, except for the case of $^{237}$Np (n, f) reaction the transmutation rates in thermal field for both of the (n, $\gamma$) and (n, f) reactions is dramatically higher than those in the fast neutron field. These results are consistent with our earlier findings [11].

5. References


