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Activation measurement of neutron production and transport in a thick lead target and a uranium blanket during 4 GeV deuteron irradiation

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ABSTRACT

Several simple accelerator-driven system (ADS) setups were irradiated by relativistic proton and deuteron beams in last years at the Nuclotron synchrotron site of the Joint Institute for Nuclear Research (JINR) in Dubna, Russia. This paper is dedicated to a 4 GeV deuteron irradiation of a setup called Energy plus Transmutation (E + T), consisting of a lead target, natural uranium blanket, and polyethylene shielding. This paper represents the finalization of data analysis and concludes systematics of the proton and deuteron experiments carried out with the E + T setup. Activation detectors served for monitoring of proton and deuteron beams and for measurements of neutron field distribution in model ADS studies. Products of reactions with thresholds up to 106 MeV as well as non-threshold reactions were observed in the samples. The yields of the produced isotopes were determined using the gamma-ray spectrometry and compared with Monte Carlo simulations performed with the MCNPX transport code.

1. Introduction

Spallation reaction as an intensive source of high-energy neutrons has been studied with an increasing interest in last decades. These studies are motivated by the need of high-energy, intensive neutron fluxes for material research, transmutation of nuclear waste or production of nuclear fuel from non-fissile isotopes [1]. With great progress in accelerator technology, accelerator-driven systems (ADS), due to their inherent safety and other unique properties, seem to be a promising tool for effective nuclear waste incineration [2].

Systematic studies of neutron production, transport, and multiplication in spallation targets irradiated by relativistic proton, deuteron, and light ion beams have been carried out at the JINR Nuclotron accelerator facility. Use of various target and blanket materials, component geometries, and moderator compositions enables to study their influence on produced neutron field. Moreover, these simple and more complex systems can serve for benchmark studies of Monte Carlo particle transport codes.

The sub-critical assembly called Energy plus Transmutation (E + T) is a system of a thick lead target surrounded by a natural uranium blanket and a polyethylene moderator–reflector and wooden biological shielding. First, this experimental setup was irradiated with proton beams in the energy range 0.7–2.0 GeV [3–8] or [9]. Next logical step in the systematic studies of spallation reactions were experiments with a deuteron beam. The E + T setup was irradiated by 1.6–4.0 GeV deuterons. The results of the 1.6 GeV and 2.52 GeV irradiations were published separately [10,9,11,12] or [13]. In this paper, we focus solely on the results of the 4 GeV irradiation carried out in November 2009.

We studied high-energy neutron field created inside the E + T setup during the 4 GeV deuteron beam irradiation by means of the activation method. The obtained data were used for testing predictions of the MCNPX transport code [14], since the E + T setup was acknowledged as an IAEA benchmark target [15]. Experimental results combined with simulations were also utilized for tests of high-energy neutron cross sections of selected threshold reactions calculated by the TALYS code [16].

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2. Materials and methods

2.1. The Energy + Transmutation setup

The E + T setup consists of a cylindrical lead target (diameter of 84 mm and total length of 480 mm) and a surrounding subcritical uranium blanket (mass of 206.4 kg of natural uranium). The target and blanket are divided into four sections. Between the sections, there are 8 mm gaps for placement of activation samples and other detectors. Each section contains a lead cylinder 114 mm long and 30 identical natural uranium rods, which are encased in a hexagonal steel container with a wall thickness of 4 mm. The front and back end of each section are covered with hexagonal aluminium plates 6 mm thick. The four target-blanket sections are mounted along the target axis on a wooden plate of 68 mm thickness, which is moreover covered with a 4 mm thick steel sheet. The uranium rods are hermetically encapsulated in a 1 mm thick aluminium cladding. Each rod has an outside diameter of 36 mm, a length of 104 mm, and a weight of 1.72 kg.

The four target-blanket sections mounted on the wooden plate are placed in a wooden container filled with granulated polyethylene. The polyethylene serves for moderation and reflection of neutrons back to the setup. The inner walls of the polyethylene box are coated with a 1 mm thick cadmium layer for absorption of neutrons thermalized in polyethylene. The floor wall of the polyethylene box is covered by a textolite plate with a thickness of 38 mm. The front and back ends of the setup are open without shielding. Schematic drawing of the experimental setup and locations of the activation detectors inside the E + T assembly are depicted in Fig. 1. A detailed analysis of the influence of different construction parts and uncertainties in their geometrical and physical definitions on the neutron flux inside the setup can be found in [17].

2.2. Experimental method and data analysis

Neutron activation method was used to study high energy neutron production and transport in the system. Activation detectors in the form of thin foils were placed in the gaps between the E + T setup sections to measure the spatial and energy distribution of the inner neutron field, see Fig. 1.

The activation samples were made of aluminium, gold, bismuth, tantalum, indium, and cobalt. These elements were chosen because they are mostly naturally mono-isotopic or one of the isotopes is dominant in natural composition. They also have suitable physical and chemical properties with very few long-lived isotopes produced (in case of bismuth and tantalum) if the foils are intended to be reusable. Further important criteria for selecting these elements were the decay times of the isotopes that were produced in observed (n,xn) threshold reactions. Isotopes with half-lives shorter than roughly an hour or longer than a year were hardly measurable with the available equipment.

The activation samples had a square shape with one side of 20 mm (Al, Au, and Ta samples), 25 mm (Bi samples), and 12.5 mm (In and Co samples). The thickness of the foils varied in the range from 50 μm to 1 mm. Chemical purity of the materials was better than 99.99%. It was tested by the activation method and no significant impurities were found. Average weight of used foils was 0.63 g for Al, 0.36 g for Au, 0.80 g for Ta, 6.54 g for Bi, 0.56 g for In, and 1.36 g for Co foils. Before the irradiation, the foils were wrapped in two layers of thin paper. The outer paper layer minimized contamination of the samples by radioisotopes coming from the setup and it was removed after the irradiation. The inner paper layer prevented the transport of the produced isotopes out of the foil and also between adjoining foils. It was present during every measurement so that possible HPGe detector contamination was excluded.

The activation foils double wrapped in paper were stuck on 5 plastic plates, mounted on wooden holders and inserted into the slots in the setup. The activation detectors were placed in the setup in two main directions — longitudinal and radial; Al, Au, Ta and Bi, Co, In samples were grouped together. The first group was placed in a row in the upward direction from the target axis (directly on the vertical axis). The second group was placed in the right-down direction in an angle of 30° from the horizontal axis. The plates were put in longitudinal direction in the distances 0, 11.8, 24.0, 36.2, and 48.4 cm from the target beginning, and on each plate the detectors were placed in radial direction in the distances 3.0, 6.0, 8.5, and 10.7 or 11.5 cm from the target axis.

When the irradiation finished and the period of cooling time (for decrease of the setup radioactivity) was over, the activated detectors were transported in a shielded container to the spectroscopic laboratory in order to measure their γ-activities with HPGe detectors. Almost all samples were measured at least twice. The first measurement of each sample lasted only a few minutes and all the samples were measured within a few hours. The second measurement was performed up to several days after the irradiation. In this way we detected the maximum of the produced isotopes. Due to about a two-hour span between the end of the irradiation and the start of the measurement the isotopes with half-lives shorter than approximately one hour could not be observed.

Complex neutron field was created inside the setup during the irradiation. This field induced various nuclear reactions in the activation samples. Many radioactive nuclei were produced mainly through (n,γ), (n,α), (n,p), and (n,xn) reactions. We measured the yields in each sample from the characteristic γ-ray spectrum emitted during their radioactive decay. The irradiated foils were measured on HPGe detectors of Ortec GMX type (with 28.3% and 32.9% relative efficiencies for the 1332 keV γ-line). The detectors were placed in a lead shielding with the front wall opened for insertion of irradiated samples. This shielding suppressed the natural background substantially. The detector systems were calibrated before the experiment and after all measurements the calibration was checked once again to control the calibration stability. The detector systems were calibrated using point-like ^{54}Mn, ^{57}Co, ^{60}Co, ^{88}Y, ^{109}Cd, ^{113}Pb, ^{133}Ba, ^{137}Cs, ^{139}Ce, ^{152}Eu, ^{228}Th, and ^{241}Am standard laboratory sources which have several dozen γ-lines ranging from 80 keV up to 2700 keV. The accuracy of the peak efficiency determination was ~2% for more distant geometries and ~3% for the nearest geometry.

The acquired γ-ray spectra were analysed and the net peak areas were determined using the Deimos32 spectroscopy software [18]. All necessary spectroscopic corrections were applied according to Eq. (1) in order to obtain the total number of nuclei of certain isotope. The yield of each isotope was then calculated as a weighted average of multiple γ-lines. The final yield was then normalized to one gram of activation foil material and to one primary beam deuteron using the measured beam intensity so that the results were comparable with previous Energy plus Transmutation experiments. The experimental yields were determined according to the formula:

$$N_{yield}^{\exp} = \frac{S_p \cdot C_{abs}(E) \cdot B_a}{I_\gamma \cdot \varepsilon_P(E) \cdot COI \cdot C_g} \frac{t_{real}}{t_{live}} \frac{1}{m_{foil}} \frac{1}{N_d} \frac{e^{(\lambda \cdot t_0)}}{1 - e^{(-\lambda \cdot t_{real})}} \frac{\lambda \cdot t_{irr}}{1 - e^{(-\lambda \cdot t_{irr})}} \quad (1)$$

where S_p represents peak area, C_{abs}(E) — self-absorption correction, B_a — beam instability correction, I_γ — gamma emission probability, ε_p(E) — detector peak efficiency, COI — correction for real γ-γ coincidences, C_g — geometry (non-point like emitter) correction, m_{foil} — mass of foil, N_d — integral deuteron beam flux, t_{real} — real time of measurement, t_{live} — live time of measurement, t_0 — cooling time, t_{irr} — irradiation time, λ — decay constant; t_{real}/t_{live} is dead time correction, and the last two fractions represent decay during cooling and measurement and decay during irradiation. Further details concerning the correction factors can be found in [19,15]. The geometry corrections for every sample size and for each measurement distance from the detector end cap were calculated using MCNPX with models of the used HPGe detectors in the same way as described in [20].

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