# TIME-DEPENDENT SPECTRA OF NEUTRONS EMITTED IN INTERACTION OF 1 AND 4 GEV DEUTERONS WITH MASSIVE NATURAL URANIUM AND LEAD TARGETS

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Prompt and delayed neutrons (DN) from pulsed irradiation of geometrically identical natural uranium and lead targets by 1 and 4 GeV deuterons were measured at NUCLOTRON facility of Joint Institute for Nuclear Research. The massive (~300 kg) cubic shaped targets were surrounded by 10 cm lead blanket. Neutrons were measured by the assembly of <sup>3</sup>He counters embedded in moderator and by threshold activation detectors. The DN decay curves were analysed for both targets and information on fragment yields for lead target and fission properties for uranium one was extracted. The obtained experimental information could be useful for verification of INC and transport codes. The results of this experiment are important for development of advanced ADS systems and other applications.

KEYWORDS : ND2010, Nuclear Data, ENDF, ADS, delayed neutrons, nuclear fission

### 1. INTRODUCTION

In the recent years scientific and practical interest in the feasibility of accelerator driven subcritical (ADS) systems for transmutation of the long-lived components of radioactive wastes (RAW) and, in the long-term outlook, for solution of global energy problems has remained quite high. The alternative program of creation of fast-neutron reactors is developed rather slowly, and with clearly visible limitations of its capabilities. This work is a preliminary step toward the study of the physicals properties of ADS-systems, in which a deeply subcritical active core (AC) from natural uranium is irradiated by a pulsed beam of relativistic deuterons. The long-range goal is the study of the possibilities of such systems with maximally hard neutron spectrum, to carry out transmutation of RAW, and also to gain energy due to burning of AC material.

The results presented below consider first of all such aspects of the general task as the neutron spectra inside an extended uranium target (and lead target for comparison) and the time dependence of the neutron yield, including its delayed component. The stimulus for studying delayed neutrons in this context was the fact that in the case of low-energy nuclear fission that has been better studied DN present a very sensitive test for basic mechanisms of the process, in particular, the characteristics of fission fragment mass distribution.

It is obvious that the mechanism of interaction of a relativistic particle with a target nucleus, even in an elementary act, is incomparably more complex than in the case of usual nuclear fission. For further consideration it is convenient to use the simplified, although still rather realistic, model proposed in [1]. In this paper four stages of the intranuclear cascade are separated. The first one is an extremely fast spallation stage during which all possible hadrons are emitted, including very high-energy neutrons. Then the nucleus transfers to a strongly heated state with a temperature of order of 5 MeV and decays by emitting a wide set of light fragments, including fast neutrons. Part of emits delayed neutrons, these fragments whose measurement gives valuable information on the properties of this stage of intranuclear cascade. And finally after defragmentation a set of relatively weakly excited nuclei with a mean temperature of about 1.5 MeV is formed, which decay mainly by competitive neutron emission and nuclear fission, the latter producing prompt and delayed neutrons.

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The picture of interaction of a relativistic particle with a target nucleus described above is complicated; it becomes even more elaborate in the case of a massive extended target. Secondary interaction of all products of elementary intranuclear cascade produced at all stages of the cascade with the target material begins to play an important role. First of all, these are fast neutrons and all possible charged and neutral products of the first spallation stage followed by secondary intranuclear (INC) cascades.

The use of massive targets made it possible to study some integral characteristics of the nuclear processes going in such targets that are important for the solution of the problems, formulated at the beginning of this paper.

# 2. EXPERIMENT

#### 2.1 Measurements

The scheme of the experiment is shown in Fig. 1. The pulsed deuteron beam of the JINR Nuclotron [2] with energies of 1 and 4 GeV from beam line 1 hit target 2. Prompt and delayed neutrons were recorded by detector 3. The time profile and intensity of each deuteron pulse was monitored using calibrated ionization chamber 4 in coincidence with two scintillation telescopes 5. The beam position on the target and the integral deuteron flux were controlled by profilometer 6 and activation monitor 7 from aluminum foil placed before the target. For decreasing the background the opening for releasing charged and neutral particles leaving the target along the beam from the measuring zone was made in shielding wall 8.

The neutron detector consisted of 11 proportional 3Hecounters placed into the 50 x 50 x 60 cm Plexiglas moderator block. Each counter was equipped with the preamplifier and discriminator. The detector efficiency for registration of neutrons from the Pu-Be- source with a medium spectrum energy of 4.4 MeV was  $11.4\pm0.1\%$ . The DAQ system provided measurement of the neutron yield as a function of time for each deuteron pulse. The detector was surrounded by appropriate shielding from borated polyethylene 9.

Two geometrically identical three-sectional targets 1 from natural uranium (315 kg) and lead (187 kg) placed into lead matrix 2,3 with input window 5 (see Fig. 2) were used in measurements. The sets of threshold activation detectors 7 (at distances of 3 and 12 cm from the beam axis) were placed on easily extracted frames in the narrow gaps between target sections 6 for measuring the spatial distribution of neutron energy spectra inside the target volume. Activation detectors for measuring the integral neutron yield were attached on the surface of the sections.

For each deuteron energy lead and uranium targets were irradiated and background measurements were

performed.



Fig. 1. Layout of the experiment.



Fig. 2. The target design

The results obtained in on-line monitoring and the integral measurements on aluminum foils are consistent within experimental errors.

#### 2.2 Experimental Results

Figure 3 shows the time dependent spectra of neutron yields for both targets and two deuteron energies normalized to one incident particle. Special measurements showed that the contribution of the background to these spectra was negligible. The spectrum for the lead target at 1 GeV is not shown due to insufficient statistics. It is evident from Fig. 3 that the DN yield from the lead target is lower by approximately two orders of magnitude than that from the uranium target. It is obvious that this is mainly connected with fission of natural uranium in the target volume.

The threshold activation detectors measured the distributions of the reaction rates for four positions in both targets along the beam axis (see Fig. 2). The neutron energy spectra at the chosen measurement points were

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reconstructed from these distributions using the standard



Fig. 3. Time dependent neutron yield spectra. Index 1 corresponds to (Pb+d) for  $E_d$ =4 GeV; indexes 2 and 3 correspond to (U+d) for  $E_d$ =1 and 4 GeV, respectively.

method of reference spectrum [3]. Some typical examples of these spectra are shown in Figs. 4 and 5.



Fig. 4. Neutron energy spectra measured between the first and second target sections at  $E_d=1$  GeV. Pb-  $\bullet$  - 3cm,  $\blacksquare$  - 12 cm; U -  $\checkmark$  - 3 cm,  $\blacktriangle$  - 12 cm.



Fig. 5. Neutron energy spectra measured between the first and second target sections at  $E_d$ = 4 GeV. Symbols are the same as in Fig. 4

## 2.3 Analysis

First of all, it should be noted that in our experimental conditions the detector load from prompt neutrons was too high. So below we analyze only the DN parts of the whole time spectra. Using the fact that the total DN yield for uranium target is much higher than that for lead (see Fig. 3) we can analyze uranium data taking into account only nuclear fission as the DN source.

In present experiment information on long-lived (1-3) DN groups is lost; therefore the data can be analyzed only for short-lived DN groups 4-6 with characteristic periods of order of 2.5, 0.6 and 0.2 s.

The result of decomposition of DN spectrum for  $E_d=4$  GeV is shown in Fig. 6.



Fig. 6. Decomposition of decay spectrum for (U + d) at  $E_d = 4$  GeV in terms of three DN groups.

The fitted half-lives are  $t_{1/2}$ = 2.75 s and 0.62 s for 4-th and 5-th DN groups respectively.

Assuming that in the massive uranium target the rate of fission defines mostly by neutrons produced in initial and secondary intranuclear cascades, it is possible to determine the respective mean energy of neutron field comparing the fitted ratio  $R = a_4/a_5$  with the experimental ones measured in <sup>238</sup>U(n,f)-reaction at various incident neutron energies. As follows from Fig.7 the obtained ratio  $R = (2.91 \pm 0.29)$  corresponds to the mean neutron energy  $\langle E_n \rangle = (24 \pm 5)$  MeV that could be responsible for fission rate within the target.

The observed DN decay spectrum formed in fission of target nuclei under the action of neutrons of different energies is determined by product of the fission cross section  $\sigma_{nf}$  (E<sub>n</sub>), the DN multiplicity  $\upsilon_d$  (E<sub>n</sub>), and the flux density of neutrons inside the target  $\varphi$  (E<sub>n</sub>). Over a wide range of E<sub>n</sub>, at least, up to 15 MeV, the product of  $\sigma_{nf}$  (E<sub>n</sub>)  $\upsilon_d$  (E<sub>n</sub>) varies within several percent. Therefore, the value of  $<E_n >$  obtained above can be considered as the realistic mean energy of neutrons initiating fission for our geometry of the uranium target.

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It should be noted here that in our scheme of experiment we measured the DN spectrum averaged over the target volume.



**Fig. 7** Comparison of weight ratio of the fourth and fifth DN groups extracted from (U+ d) decay spectrum at  $E_d=4$  GeV and that obtained from U(n,f)-reaction. Symbols -  $\Box$  - [4],  $\Diamond$  - [5],  $\Delta$  - [6], \* - [7],  $\circ$  - [8].

So the value  $\langle E_n \rangle$  is a kind of an integral characteristic of the neutron spectrum of the target.

Of course, the total neutron energy spectrum below 10 MeV should be enriched by prompt fission neutrons produced in initial fission. And with increasing the radial target size the role of these secondary neutrons in production of delayed neutrons should become more important. For a quasi-infinite target the value of  $\langle E_n \rangle$  should be essentially lower. The value  $\langle E_n \rangle$  obtained above gives some indications that with our intermediate size of the target most of secondary neutrons leave the target volume without producing fission of target nuclei. It can be stated that the study of the decay spectra of DN predecessors provides an important and sensitive tool for investigation of basic characteristics of fission process in a massive fissile target used as the active core of an ADS system.

Concerning the total neutron spectra shown in Figs. 4 and 5 it should be noted that the direct DN contribution in these spectra are negligible. These spectra are formed by high energy neutrons emitted from first fast stages of INC, secondary neutrons of multiple inelastic scattering and (n,xn)-reactions induced by neutrons of the primary spectrum and also by neutrons evaporated from exited residual nuclei of the last stage of intranuclear cascade. An additional and important contribution can go from prompt neutrons of target nucleus fission.

The whole set of neutron spectra was obtained for two distances (3 and 12 cm) from the beam axis and four positions along this axis. The spatial measurement positions for U and Pb targets were the same. The comparison of these total neutron spectra partly presented in Figs. 4 and 5 demonstrates the pronounced contribution of prompt fission neutrons in the energy range (1 - 10) MeV for the uranium target. It can be seen from Figs. 4 and 5 that the role of these neutrons is more important for the central zone of the target than for peripheral regions. Beside that this effect becomes more pronounced with increasing incident deuteron energy. Note that the total neutron multiplicity also grows with increasing deuteron energy.

The difference between the overall shapes of the neutron spectra obtained for uranium and lead targets becomes stronger for higher deuteron energy especially for the central target zone. The whole set of neutron spectra will be analyzed in further papers.

#### 2.4 Concluding remarks

The presented experiment is a preliminary stage of the extensive studies with quasi-infinite uranium and lead targets that are planned to be performed at Nuclotron with proton and deuteron beams in an energy range from 1 to 10 GeV. This research program is aimed at the study of the possibilities to use such type of ADS systems for transmutation of radioactive wastes and for energy production.

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