

# INFLUENCE OF HIGH ENERGY TALE OF NEUTRON SPECTRUM FORMED WITHIN THE QUASI-INFINITE ACTIVE TARGET ASSEMBLY BURAN IRRADIATED BY RELATIVISTIC PROTONS OR DEUTERONS ON BEAM ENERGY RELEASE

Zhivkov P.<sup>1</sup>, Baznat M.<sup>2</sup>, Chilap V.<sup>3</sup>, Furman W.<sup>4</sup>, Stoyanov Ch.<sup>1</sup>, Tutunnikov S.<sup>4</sup>

<sup>1</sup> Institute of Nuclear research and Nuclear Investigations, BAS, Bulgaria

<sup>2</sup> Moldova Academy of Sciences, Kishinev, Moldova

<sup>3</sup> Center Physical and Technical Projects “Atomenergomash”, Moscow, Russia

<sup>4</sup> Join Institute for Nuclear Research, Dubna, Russia

**Abstract:** Last decade a considerable interest in study of Accelerator Driving Systems (ADS) was observed, because they can be used for transmutation of nuclear waste and for energy production. Most of the efforts are concentrated on the simulations of physical properties of such systems and on the development of possible prototypes of ADS. The most advanced transport codes were used for these purposes, such as MCNPX, FLUKA, MARS, SHILD, as well the last versions of relevant nuclear models. One of the key characteristic of any ADS is the equilibrium neutron spectrum generated in the active core under irradiation with a beam of high energy particles. All nuclear models used in abovementioned codes for description of neutron induced fission account for only part of the spectrum with neutron energy  $E_n < 20\text{MeV}$ . In this paper it is estimated the influence of high energy tale of neutron spectrum formed within the quasi-infinite depleted uranium target assembly BURAN irradiated by protons or deuterons with energy from 1 to 12 GeV on main characteristics of this ADS .

## Introduction

During last fifty years the various types of ADS have been studied in Dubna experimentally and theoretically (see for instance [1 – 9]). There were studied ADS with natural and depleted uranium targets, lead-uranium and lead-graphite target assemblies irradiated by protons or deuteron beams with energy from 0.66 to 3.7 GeV. A lot of valuable information was obtained. Last four years the new project “Energy and Transmutation Radioactive Wastes” (E&T RAW) was launched at JINR [10, 11]. In the framework of this project during 2011 -2014 a wide variety of experiments have been done using massive (512 kg) natural uranium target assembly (TA) QUINTA irradiated by deuteron beams of JINR Nuclotron in incident energy range (1 – 8) GeV. The review of the results obtained till 2013 was presented in [12-16]. TA QUINTA has target of hexagonal shape about 30 cm in diameter and 65 cm in length surrounded by a lead blanket of thickness 10 cm.

The neutron spectrum was measured with threshold activation detectors located inside of TA as well as on the surface of the lead blanket. The rates of reactions (n,f), (n, $\gamma$ ) and (n,2n) <sup>nat</sup>U and other samples were measured. The experimental results were compared with the calculations [13, 14, 16] made with the different codes (MCNPX, Fluka, MARS, GEANT4) which used various nuclear models (ISABEL, INCL4, LAQGSM, Bertini and fission evaporation model ABLA). The overall agreement in limits of (10 – 30) % between of experiment and calculations for spatial distribution and integral values of <sup>nat</sup>U(n,f), (n, $\gamma$ ) and (n,2n)- reaction rates has been achieved. These reactions are determined mainly with the part

of the neutron spectrum energy below 20 MeV. But for  $^{209}\text{Bi}(n,f)$ -reaction and production of  $^7\text{Be}$  and  $^{22}\text{Na}$  nuclei in aluminum foils which is connected with high energy ( $E_n > 30$  MeV) part of the neutron spectrum a discrepancy between experiment and calculations exceeds 100%. This means that present codes are not able to reproduce high energy tail of neutron spectrum formed within natural uranium target of intermediate size such as TA QUINTA.

During the analysis of the experimental results some semi- phenomenological estimates [16] were made of the impact of high-energy part of the neutron spectrum ( $E_n > 20$  MeV) on integral numbers of fissions  $N_f$  for TA QUINTA and available at JINR quasi-infinite (with small neutron leakage) Big URANIUM (BURAN) TA of depleted uranium mass of about 21t. Calculations have shown that, if for the TA QUINTA with a large ( $\sim 80\%$ ) neutron leakage the effect of  $\sim 30\%$ , so for the quasi-infinite target the correct account of the role of high-energy neutrons can lead to an increase in  $N_f$  at  $\sim 100\%$ . Below this problem will be considered in more detail.

The lay-out of TA BURAN is presented in Fig. 1.

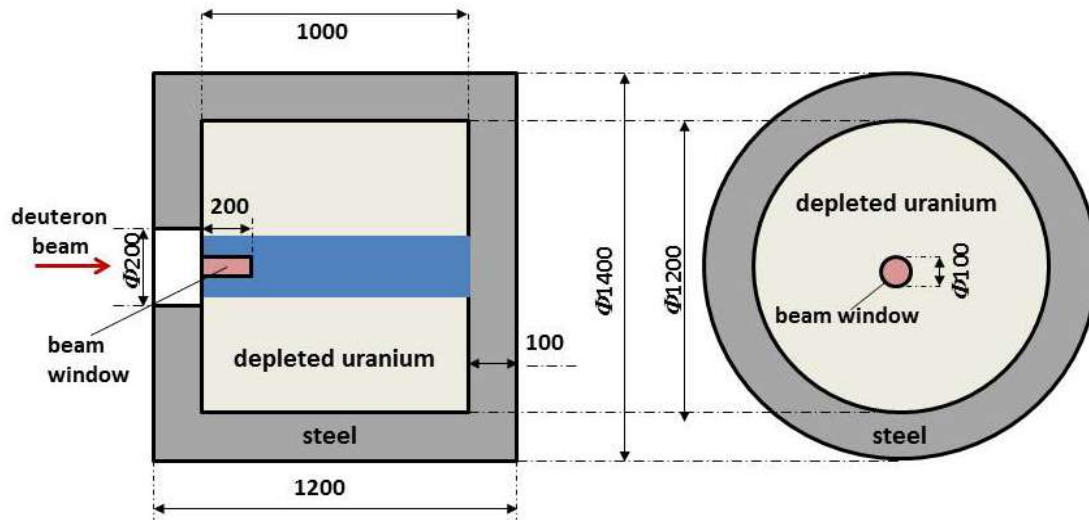


Fig1. The vertical and horizontal cuts of TA BURAN. It consists of the replaceable central part 18x80cm surrounded by a solid array of depleted uranium in 10 cm steel case. The central zone may be made of lead or uranium.

### Calculation of the reaction rates and total energy deposition

MCNPX 2.7e transport code [17] makes it possible to calculate the spatial distributions of any reaction rates within massive target irradiated by high energy incident particles as well as the respective integral characteristics. In present calculations the following models are applied ISABEL/ABLA + Fluka, QGSM with SMM (statistical multi fragmentation model), INCL4 and CEM03. A pre-equilibrium stage was involved too. Three main nuclear processes take place in the target: 1) intranuclear cascade (INC) whereby high-energy (spallation) neutrons, protons and  $\pi$ - mesons are generated, which preserve the direction of diffusion along the incident (proton/deuteron) beams; 2) isotropic emission of pre-fission neutrons before fission of uranium nuclei induced by high energy particles created at spallation stage and 3) post-scission neutrons evaporated from the fission fragments and other residual excited nuclei.

All generated neutrons participate in elastic and non-elastic interactions with the uranium nuclei. Some of these reactions such as  $^{235}\text{U}(n,f)$  or  $^{\text{nat}}\text{U}(n,\gamma)$  have no any neutron energy

thresholds but other meaningful reactions -  $^{238}\text{U}$  (n,f) and  $^{238}\text{U}$  (n,xn) have essential thresholds. Of course, the same reactions are induced by protons and  $\pi$ - mesons as well as by  $\gamma$ -quanta, but their contribution is much smaller than the neutron induced ones. MCNPX 2.7e code calculates all these reactions within whole target and obtained such important integral characteristic of given ADS as a total beam energy deposition  $E_{\text{dep}} = (E_{\text{fiss.}} + E_{\text{ion}})$  in the multiplying target. MCNPX2.7e transport code makes it possible to calculate all above discussed values in several ways.

The first option is a direct Monte-Carlo (MC) simulation of the product yields of each reactions within some volume of the target with coordinates (R,Z) followed by summation over whole target. As result the output files are displayed which consist of tables with the creation/losses balance for each particle that participated in the transportation. But this option takes into account only neutron induced reactions with cutoff neutron energy up to 20 MeV.

The second option permits to calculate the neutron, proton, deuteron, pion and photon spectra in each separate volume (R,Z). Using known cross-sections for all reactions under consideration from the databases TENDL, ENDF, EXFOR or others it is possible to calculate respective numbers of each reaction  $i$  per one incident (on target) proton/deuteron  $N_i(R,Z)$  within selected volume (R,Z) by the following formula

$$N_i(R,Z) = \sum_j \int_{E_{\min}^j}^{E_{\max}^j} \Phi_j(R,Z,E) \sigma_i(E) dE \quad , \quad (1)$$

where  $i$  denotes the type of reaction with a cross-section  $\sigma_i(E)$  and  $j$  refers to the spectrum of the particle inducing a reaction  $i$ . The values  $E_{\min}^j$  and  $E_{\max}^j$  are the minimal and maximal energies of the respective spectrum. An integration in (1) realizes a convolution of given cross-section  $\sigma_i(E)$  with the flux  $\Phi_j(R,Z,E)$  that assumed be constant within considered (R,Z) volume. Also it implicitly means the integration over selected volume (R,Z). Then after summation of  $N_i(R,Z)$  over whole target volume one can obtain the total numbers of respective reaction  $N_{i \text{ tot}}$ . Beside that MCNPX code calculates the total number (multiplicity) of neutrons  $N_{\text{tot}}$  generated in the target assembly.

The simulations have been done for the range of proton and deuteron energy (1 – 12) GeV. The results for deuteron beam are presented in table 1. (Note at once that the results for irradiation by protons with the same energy are similar). Beside values of  $N_{f \text{ tot}}$  and  $N_{\gamma \text{ tot}}$  calculated as described below in the table also are shown so called beam power gain (BPG) defined by the ratio  $\text{BPG} = E_{\text{dep}}/E_d$  that gives the energy efficiency of the studied ADS.

The values presented into row 3, 4 and 5 are calculated in standard option of MCNPX code with cutoff neutron energy up to 20 MeV for all inelastic reactions. An impact of high energy ( $E_n > 20$  MeV) tale of neutron spectrum is taken into account additionally by formula (1) for  $N_{f \text{ tot}}$  values shown in the sixth row. The contributions from (p+d+ $\pi$ + $\gamma$ ) induced fission presented in the seventh row is accounted for in the same way. The beam power gain  $\text{BPG}_{\text{MCNPX}}$  given in row 8 includes the fission yield summed over row 6 and 7. As seen from the table the values  $N_{\text{tot}}$ ,  $N_{\gamma \text{ tot}}$  and  $N_{f \text{ tot}}$  increase proportionally to incident deuteron energy  $E_d$ . But beam power gain  $\text{BPG}_{\text{MCNPX}}$  shows a slight drop with energy  $E_d$ . This is due to a relative decrease in the ionization losses  $E_{\text{ion}}$  during the growth of incident energy [10]. Note that the absolute values of  $\text{BPG}_{\text{MCNPX}}$  are lesser than the minimal value of  $\text{BPG}_{\min} \approx 9$  providing a zero energy balance of whole ADS.

Table 1. The results of simulation of the total numbers (per one incident deuteron) of fission  $N_{f\,tot}$  and radiative capture  $N_{\gamma\,tot}$  events as well total neutron multiplicities  $N_{tot}$  and  $BPG$  for TA BURAN.

1	$E_d$ [GeV]	1	2	4	6	12
2		MCNPX calculations				
3	$N_{tot}$	129	288	567	823	1536
4	$N_{\gamma\,tot}$	73	164	325	473	883
5	$N_{f\,tot}, E_n < 20\text{MeV}$	15.3	34.5	68	98	184
6	$N_{f\,tot}, \text{total } E_n \text{ spectrum}$	18.9	42.7	84	122	227
7	$N_f(p+d+\pi+\gamma, f)^{235,238}\text{U}$	1.2	2	2.3	5	9.7
8	$BPG_{MCNPX}$	6.15	6.26	5.7	5.54	5.14
9		MCNPX calculations + estimated impact of high energy neutrons				
10	$N_{tot\,est}$	200	450	884	1297	2386
11	$N_{\gamma\,tot\,est}$	123	276	545	803	1472
12	$N_{f\,tot\,est}$	37	83	163	216	439
13	$BPG_{est}$	9.4	10.3	9.8	8.9	8.8

As follows from the table the contribution of high energy part ( $E_n > 20$  MeV) of neutron spectrum leads to additional 3.6 fission events that consists of about 25% of  $N_{f\,tot}$  (compare the rows 5 and 6 for  $E_d = 1$  GeV). But fission events in this range of  $E_n$  accompany by prompt neutron emission with a multiplicity  $\mu$  growing fast when  $E_n$  increases (see Fig. 2 and refs. [18,19]). Using the calculated neutron spectrum averaged over TA volume, the cross section  $\sigma_{nf}(E_n)$  for  $^{238}\text{U}$  and the data from Fig. 2 one can estimate the mean value of  $\mu \approx 8$  for the range of  $E_n \sim (20 - 200)$  MeV. So about thirty additional neutrons have to appears as a result of these high energy fission acts.

If we assume that 25% of these additional neutrons induce (n,f)-reactions and the other 75% are moderated and disappear via (n,  $\gamma$ )-process so it easy to calculate using  $\mu_0 = 2.5$  for low energy fission that values of  $N_{tot}$ ,  $N_{\gamma\,tot}$  and  $N_{f\,tot}$  as well the respective BPG increase essentially due to the chains of secondary fissions. The results of such estimations are presented in table 1, rows 10 – 13.

In above described consideration we neglect any effects related with (p,f)- and ( $\pi$ ,f)-reactions as well as with small possible leakage of neutrons from TA BURAN. Beside that the estimates have been made in not self-consistent manner. But we believe that these estimates indicate a significant impact of high energy part of neutron spectrum on main basic characteristics of any quasi-infinite deep subcritical ADS such as TA BURAN irradiated by relativistic incident particles.

## Conclusion

As noted above in Introduction the present codes are not able to reproduce high energy tale of neutron spectrum formed within natural uranium target of intermediate size such as TA QUINTA. Apparently, with this is associated inability of standard codes to explain the total numbers of fissions obtained by experiments in [1] with almost quasi-infinite targets made from metallic natural and depleted uranium irradiated by 660 MeV protons.

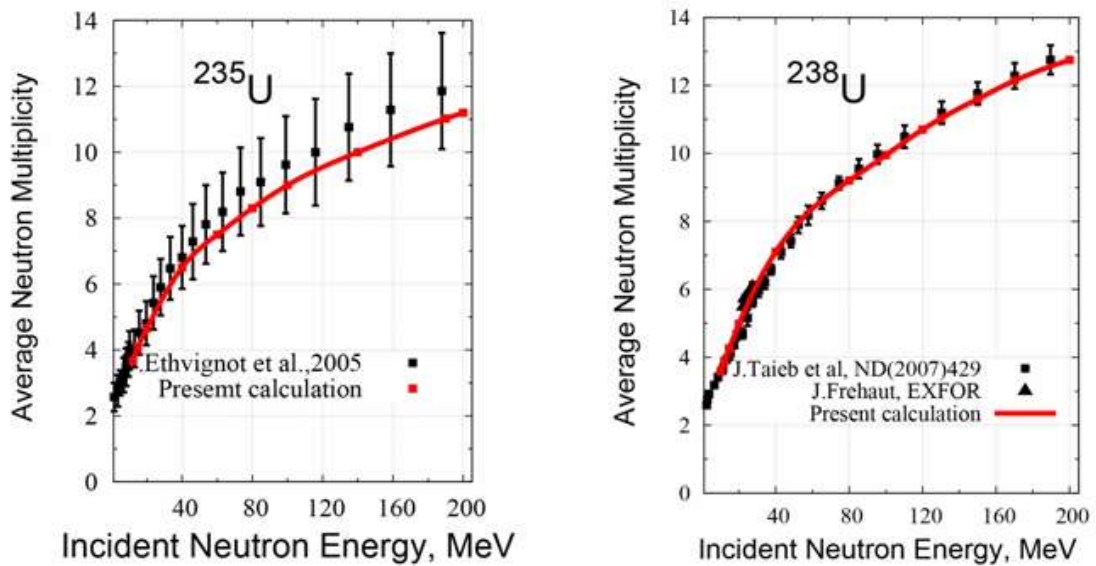


Fig. 2. Fission neutron multiplicity for  $^{235,238}\text{U}$  (n,f)-reactions as function of incident neutron energy.

The difference between  $N_{f\ tot}$  calculated by MCNPX code and the respective experimental value from ref. [1] is about two times. But with the same approach as used by the end of previous Section it is easy to reproduce the data [1]. Taking in account results of experiments [2] where for a lead target  $\varnothing 20 \times 60 \text{ cm}$  irradiated by protons and deuterons with energy from 1 to 3.7 GeV there was obtained that the high energy tale ( $E_n > 20 \text{ MeV}$ ) of neutron spectrum consists about 10%. The MCNPX calculations for TA BURAN gives for this share only about 1%. So if in reality this tale for TA BURAN is closer to numbers from [2] the estimations made above and presented in table 1 become more plausible. But of course only future experiments with TA BURAN permit to clear out this question finally.

This work has been partially supported by "the Alexander von Humboldt Foundation under the Equipment Subsidies Program".

## References

- [1] Vassil'kov R., Gol'dansky V., Pimenov B. and Pokotilovsky Yu., *Multiplication of neutrons in uranium irradiated by protons with energy 300-660 MeV*, Atomic Energy, v.44, (1978), p. 329 - 334 (in Russian).
- [2] Yurevich V., *Production of neutrons in thick targets by high-energy protons and nuclei*, Physics of Particles and Nuclei v.41 (2010) 778-825.
- [3] Barashenkov V.S. Nuclear-physical aspects of electro-nuclear method, Physics of Particles and Nuclei v.9 i.5 (1978).
- [4] Barashenkov V. S, Problems of electro-nuclear technology, JINR P2-94-56, Dubna, 1994.

- [5] Krivopustov M.I. at all., *First results studying the transmutation of  $^{129}\text{I}$ ,  $^{237}\text{Np}$ ,  $^{238}\text{Pu}$ , and  $^{239}\text{Pu}$  in the irradiation of an extended  $^{nat}\text{U}/\text{Pb}$ -assembly with 2.52 GeV deuterons*, Journal of Radioanalytical and Nuclear Chemistry, Vol. 279, No.2 (2009) 567–584.
- [6] Krivopustov M.I. at all., *First experiment on calorimetry of the uranium blanket of U/Pb-target assembly “Energy plus Transmutation” at JINR synchrotron proton beam with energy 1,5 GeV*, «Kerntechnik», v.68, (2003), p.48-55.
- [7] Adam J., Barashenkov V.S., Ganesan S. et al., *Measurement of the neutron fluence on the spallation source at Dubna*, KERNTECHNIK 70 (2005), 127-132.
- [8] Adam J., Barashenkov V.S., Kumawat H., Kumar V. et al., *Eur. Phys. J., A* **23**, 61 (2005).
- [9] Adam J., Bhatia C., Katovsky K., Kumar V. et al., *A study of reaction rates of (n,f), (n, $\gamma$ ) and (n,2n) reactions in  $^{nat}\text{U}$  and  $^{232}\text{Th}$  by the neutron fluence produced in the graphite set-up (GAMMA-3) irradiated by 2.33 GeV deuteron beam*, *Eur. Phys. J. A* **47**, 85-104 (2011).
- [10] Adam J. et al. (“E&T – RAW” Collaboration), *Study of Deep Subcritical Electronuclear Systems and Feasibility of Their Application for Energy Production and Radioactive Waste Transmutation*, Preprint JINR E1-2010-61, Dubna, 2010.
- [11] Baldin A., Belov E., Galanin M. et al., 2011, *Nuclear relativistic technologies (NRT for energy production and utilization of spent nuclear fuel. Results of first experiments on physical background of NRT.*, 2011, Letters to PEPAN, v. 8(6), p.1007 – 1023
- [12] Furman W., Adam J., Baldin A. et al., 2012, *Recent results of the study of ADS with 500 kg natural uranium target assembly QUINTA irradiated by deuterons with energies from 1 to 8 GeV at JINR NUCLOTRON.*, In Proceedings of XXI IHEP, Dubna 2012, PoS(MC2000)086.
- [13] Zavoroka L., Adam J., Furman W. et al., *A summary of experimental results on the reactions in uranium samples irradiated with a deuteron beam of energies up to 8 GeV at the QUINTA target*, PoS(Baldin ISHEPP XXI)089.
- [14] Suchopar M., Wagner V., Vrzalova J. et al., *MonteCarlo simulation of natural uranium setups irradiated with relativistic deuterons by means of MCNPX code*, PoS(Baldin ISHEPP XXI)091.
- [15] Adam J., Artyushenko M., Baldin A. et al., 2013, *Investigation of spatial distributions of fission and radiative capture in massive uranium target irradiated by deuterons of energies 1 – 8 GeV*, Preprint of JINR P1-2012-147, Dubna, (in Russian).
- [16] Zhivkov P., Furman W., Stoyanov Ch., *Calculation of ADS with deep subcritical uranium active cores – comparison with experiments and predictions*, Proc. Of XX International School on Nuclear Physics, Neutron Physics and Applications, Varna, J. Phys.:Conf. Ser.,**533**,012053, (2014).
- [17] <https://mcnpx.lanl.gov/>
- [18] Ethvignot T., Devlin M., Duarte H. et al., *Neutron Multiplicity in the Fission of  $^{238}\text{U}$  and  $^{235}\text{U}$  with Neutrons up to 200 MeV*, Phys. Rev. Lett. 94, 052701
- [19] Taieb J. et al., ND-2007 (2007) 429.