Calculations of ADS with deep subcritical uranium active cores – comparison with experiments and predictions

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Abstract. The main characteristics of the neutron field formed within the massive (512 kg) natural uranium target assembly (TA) QUINTA irradiated by deuteron beam of JINR Nuclotron with energies 1, 2, 4, and 8 GeV as well as the spatial distributions and the integral numbers of (n,f), (n,γ) and (n,xn)- reactions were calculated and compared with experimental data [1]. The MCNPX 27e code with ISABEL/ABLA/FLUKA and INCL4/ABLA models of intra-nuclear cascade (INC) and experimental cross-sections of the corresponding reactions were used. Special attention was paid to the elucidation of the role of charged particles (protons and pions) in the fission of natural uranium of TA QUINTA. Extensive calculations have been done for quasi-infinite (with very small neutron leakage) depleted uranium TA BURAN having mass about 20 t which are intended to be used in experiments at Nuclotron in 2014-2016. As in the case of TA QUINTA which really models the central zone of TA BURAN the total numbers of fissions, produced ²³⁹Pu nuclei and total neutron multiplicities are predicted to be proportional to proton or deuteron energy up to 12 GeV. But obtained values of beam power gain are practically constant in studied incident energy range and are approximately four. These values are in contradiction with the experimental result [2] obtained for the depleted uranium core weighting three tons at incident proton energy 0.66 GeV.

1. Introduction
The main characteristics of the neutron field formed within the massive (512 kg) natural uranium target assembly (TA) QUINTA surrounded by 10 cm lead blanket under irradiation by deuteron beam of JINR Nuclotron with energies 1, 2, 4, and 8 GeV as well as the spatial distributions and the integral numbers of (n,f), (n,γ) and (n,2n)- reactions were studied experimentally during last two years [1]. Here there are presented some results of simulations of these experiments by the code MCNPX 27.e [3] with INC model ISABEL, evaporation model ABLA and transport of the high energy particles by FLUKA (for detail see [4]). For the sake of precision the simulations were done also with combination INCL4/ABLA. In calculations all interactions of deuterons and produced by them neutrons, protons, pions and photons with whole TA QUINTA materials were considered.

2. The influence of charged particles on the fission of uranium in TA QUINTA
The deuteron beam generates in TA QUINTA not only neutrons, but high energy charged particles and photons too. In our simulations there were used the experimental cross sections for (n,γ), (n,f), (p,f),
(--f) and (γ,f) reactions from TENDL 2009 up to 20 MeV for respective entrance channel energies and for higher energies they were calculated by TALYS code. The flux of the charged particles was generated by MCNPX code with options described above. Note that the real beam profile and its position within TA are strongly influence on results and should be taken into account for each experiment precisely. The calculations show that the reactions 238U(p,f) and 235U(π,f) have an essential significance in the central part of the uranium target close to the axis of the beam. The results are presented in figure 1. It is seen that just on the axis (R=0) and up to R=4cm about 25% of total fission rate is related to protons and pions but their contribution sharply decreases to R=8cm and plays a negligible role at R=12 cm. The influence of the reaction 238,235U(γ,f) was insignificant.

3. The contribution of 235U(n,f)-reaction in total fission rates for TA QUINTA
In natural uranium has the central part of TA QUINTA there is ~0.7% of 235U. Due to presence in total energy spectrum of many slow neutrons it is necessary to account for 235U(n,f)-reaction in spatial distribution of fission rate within TA. The calculations show that the ratios 235U(n,f)/238U(n,f) are from 0.03 till 0.14 depending on the positions within TA and on deuterons energy E_d. For E_d=4 GeV and R=0cm the ratios are from 0.03 up to 0.07 depending on positions along beam axis, for radii R=4cm, 8cm and 12cm the ratios consist ~ 0.08, 0.1 to 0.12 and 0.12-0.14 respectively. For beam energy 8 GeV these ratios are rather similar. And the contribution of 235U(n,f)-reaction into total fission number for TA QUINTA is in limits 2-4%.

4. The distributions of the neutron flux and its mean energy <E_n> over TA
The neutron flux and its mean energy <E_n> have been calculated for all detector positions within and on the surface of TA QUINTA. This is important for analysis of data obtained with the silicon detectors used to measure the spatial distribution of total neutron flux that are rather sensitive to the value of <E_n>. The calculated values <E_n> vary from 1 to 30 MeV and from 1 to 36 MeV for deuterons energies E_d = 4 GeV and E_d =8 GeV respectively. The neutron flux varies essentially over TA and in general does not follow R^2 dependence. It is maximal in the middle of TA and reduces to three-four times by the end of the target. The radial flux dependence becomes weaker (change up two times) for last uranium sections of TA.

5. Spectral indices natU(n,f)/238U (n,2n) and 238U(n,γ)/238,235U(n,f)
Natural uranium samples placed on the aluminum plates within TA QUINTA were used [1, 5] for simultaneous activation measurements of natural uranium fission rate, (n,2n) and (n,γ)-reactions. This
provides a possibility to obtain so called spectral indices (SI) defined as the ratios of the respective reaction rates: $^{nat}\text{U}(n,\gamma)^{nat}\text{U}(n,f)$ and $^{238}\text{U}(n,2n)^{238}\text{U}(n,f)$. By definition these values do not depend on the uncertainties of beam monitoring (error $\sim 13\%$), on the intensity of neutron flux and on the exact knowledge of beam position on the target. So the spectral indices could be measured more precisely than absolute values of the particular reaction rates. In each TA position SI depend on the shape of neutron energy spectrum only and can be used to verify the correctness of the simulated spectrum. In figure 2 a comparison of experimental and calculated SI $^{nat}\text{U}(n,\gamma)^{nat}\text{U}(n,f)$ is presented for deuteron energies $E_d=4$ and $8$ GeV. One can see that calculations reproduce the whole character change the shape of the neutron spectra along the axis of the target. But the calculated values of SI and their dependence on incident energy do not agree with experiment. Namely experimental SI indicate some “hardening” of neutron spectra with growth of deuteron energy that does not reproduced by simulation.

**6. Total numbers of fission and neutron radiative capture in TA QUINTA**

By integration over the spatial distributions of measured $(n,f)$ and $(n,\gamma)$ reaction rates the total numbers of these reactions in TA QUINTA were deduced [1,5]. In figure 3 the calculated values of total numbers of fission $N_f$ and the total amounts $N_{Pu}$ of produced $^{239}\text{Pu}$ nuclei (normalized per one incident particle) are compared with experiment for deuteron energy range $E_d = (1-8)$ GeV. As follows from the picture the respective experimental values grow with $E_d$ remaining constant (within limit of their uncertainties) per unit of deuteron energy. If the calculated $N_{Pu}$ values reasonably reproduce the measured ones so the calculated ratios $N_f/E_d$ show a noticeable drop with growth of $E_d$. It means that simulated neutron spectra fail to reproduce high energy part of the experimental ones.

![Figure 3](image1.png)

**Figure 3.** Total numbers of fissions and $(n,\gamma)$- reactions in TA QUINTA

![Figure 4](image2.png)

**Figure 4.** Schematic lay-out of TA BremaiURAN

**7. Simulation of future experiments with quasi-infinite TA BURAN**

In eighties of last century there was designed and constructed unique massive (about 21 t) TA made from metallic depleted (0.03% of $^{235}\text{U}$) uranium – Big URANium (BURAN) target. It was intended for experiments on JINR Synchrophasotron beams. But these plans have not been realized. Now the project “Energy and Transmutation Spent Nuclear Fuel” endorsed at JINR for 2014-2026 is based on an upgrade and extensive use of TA BURAN. The schematic layout of it is shown in figure 4. Due to rather large dimensions of TA BURAN most part of neutrons and charged particles generated by incident high energy deuterons has to be absorbed in its volume. So this TA could be considered as deep-subcritical quasi-infinite active core proposed to use in new ADS (Accelerating Driving System) scheme [6]. For better planning of future experiment with TA BURAN the simulation of its main characteristics under irradiation by protons and deuterons has been done with the same variant of codes as described above. The results are given in Table 1. It is seen that all presented values excluding the neutron leakage stay near constant. It means that their absolute amounts per one incident deuteron grow linearly with increasing incident energy. The neutron leakage decreases essentially with
growth of $E_d$ that indicates on more effective “use” of neutrons inside of TA BURAN for higher incident energy. Beside present calculations confirm the assumption of a small neutron leakage from TA BURAN which is ~ 2% in comparison with ~ 60% for TA QUINTA.

Table 1. The integral characteristics of TA BURAN under irradiation by protons and deuterons ( per one incident particle/per one GeV)

| $E_p(d)$ , (GeV) | Protons | Deuterons |
|-----------------|---------|-----------|
|                 | 1       | 6         | 12        | 1       | 6         | 12        |
| Total neutron multiplicity | 126     | 128       | 121       | 125     | 132       | 121       |
| Total number of produced $^{239}$Pu nuclei | 70      | 73        | 69        | 70      | 75        | 70        |
| Total number of fission | 16      | 16.6      | 15        | 15      | 16.6      | 15.2      |
| Number of leaked neutrons | 3       | 2.3       | 1.9       | 3       | 2.3       | 1.8       |
| BPG = $E_{\text{release}}/E_{p(d)}$ | 3.82    | 3.75      | 3.5       | 3.82    | 3.85      | 3.55      |

Note that if the total neutron multiplicity and total number of fission increased about two times in comparison with TA QUINTA so the total number of produced $^{239}$Pu nuclei grew more than six times. This indicates an essential softening of the calculated neutron spectrum in TA BURAN.

Conclusion. The key elements of new ADS scheme [6] are the maximally hard neutron spectrum formed in the deep subcritical multiplying target from natural or depleted uranium and large enough the beam power gain (BPG ≥ 9). Both these parameters are necessary for effective utilization SNF with simultaneous energy production. But the calculated BPG ≈ 3.8 are lower even than the value of BPG ≈ 6 was obtained in [6] from the data [2] in which has been studied the massive (3t) metallic depleted uranium target irradiated by protons with energy up to 0.66 GeV. This energy is well below the threshold of fragmentation when the neutron multiplicity per unit of incident energy has not yet reached its asymptotic value. It seems probable that simulated neutron spectrum in TA BURAN is too soft underestimating its high energy part what in turn can lead to significant underestimation of BPW. So only future experiments with TA BURAN allow us to clear out real possibilities of new ADS scheme [6] for solving the problems of global energy.

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References
[1] 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
[2] Vassil’kov R., Gol’dansky V., Pimenov B. et al., 1978, Multiplication of neutrons in uranium irradiated by protons with energy, 300-660 MeV., Atomic Energy, v.44, p. 329 - 334 (in Russian)
[3] https://mcnpx.lanl.gov/
[4] Filges D., Goldenbaum F., 2010, Handbook of Spallation Reaction., WILEY – VCH, 2010
[5] Adam J., Artushenko 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)
[6] 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