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Measurement of the high energy neutron flux on the surface of the natural uranium target assembly QUINTA irradiated by deuterons of 4 and 8 GeV energy

J. Adam[a,f,]* , A.A. Baldin[b], V. Chilap[c], W. Furman[a], K. Katovsky[d], J. Khushvaktov[a], V. Kumar[e], V. Pronskikh[a,g], I. Mar'in[a], A. Solnyshkin[a], M. Suchopar[f,h], V. Tsupko-Sitnikov[a], S. Tyutyunnikov[a], J. Vrzalova[a,h], V. Wagner[f], L. Zavorka[a,h]

[a]*Joint Institute for Nuclear Research, Dubna, Russia*
[b]*Institute for Advanced Studies "OMEGA", Dubna, Russia*
[c]*CPTP "Atomenergomash", Moscow, Russia*
[d]*University of Technology, Brno, Czech Republic*
[e]*G.G.S.Indraprastha University, New Delhi, India*
[f]*Nuclear Physics Institute, Rez, Czech Republic*
[g]*Fermi National Accelerator Laboratory, Batavia, IL, USA*
[h]*Czech Technical University, Prague, Czech Republic*

Abstract

Experiments with the natural uranium target assembly "QUINTA" exposed to 4 and 8 GeV deuteron beams of the Nuclotron accelerator at the Joint Institute for Nuclear Research (Dubna) are analyzed. The reaction rates of ^{27}Al(n,y1)^{24}Na, ^{27}Al(n,y2)^{22}Na and ^{27}Al(n,y3)^{7}Be reactions with effective threshold energies of 5, 27, and 119 MeV were measured at both 4 GeV and 8 GeV deuteron beam energies. The average neutron fluxes between the effective threshold energies and the effective ends of the neutron spectra (which are 800 or 1000 MeV for 4 or 8 GeV deuterons) were determined. The evidence for the intensity shift of the neutron spectra to higher neutron energies with the increase of the deuteron energy from 4 GeV to 8 GeV was found from the ratios of the average neutron fluxes. The reaction rates and the average neutron fluxes were calculated with the MCNPX 2.7 code.

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Keywords: Accelerator driven system (ADS); uranium target-blanket; spallation neutrons; neutron spectrum; gamma spectrometry; MCNPX.

* Corresponding author. Tel.:+7(49621)62175, *E-mail address:* iadam@jinr.ru

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1. Motivation

At JINR, Dubna during the last two decades in the framework of the «Energy + Transmutation of Radioactive Waste (E+T RAW)» collaboration, extensive ADS studies have been carried out with high-energy proton and deuteron beams. In particular, the transmutation rates of long lived fission products (LLFP) and transuranium (LLTRU) nuclides were measured in the neutron fields generated within lead-uranium by Adam et al. (2010) and by Bhatia et al. (2012) and lead-graphite by Adam et al. (2011) targets with moderators as well as in the natural uranium target assembly (TA) QUINTA (with and without the lead blanket) by Furman et al. (2012) irradiated by deuterons of energy 1 - 8 GeV. The effective incineration of LLFP and LLTRU needs a maximally hard neutron spectrum due to the high thresholds of (n,f) and (n,xn) reactions necessary for their transmutation.

In the present work, this problem is studied with an activation method (see Adam et al. (2005)). The yields of the product nuclei in $^{27}Al(n,y_1)^{24}Na$, $^{27}Al(n,y_2)^{22}Na$, and $^{27}Al(n,y_3)^{7}Be$ reactions have effective threshold neutron energies $E_{n,th}$ = 5, 27, and 119 MeV, respectively. These three monitor reactions have been chosen for measurement of the flux of spallation neutrons produced by 4 and 8 GeV deuterons colliding with the uranium target QUINTA. The results of these measurements are compared with the calculations performed by MCNPX2.7 to estimate the ability to reproduce the whole shape of the neutron spectra.

The target assembly QUINTA consists of four identical sections of hexagonal aluminum containers with an inscribed circular diameter of 284 mm. Each one contains 61 cylindrical metallic natural uranium rods. Each rod is 36 mm in diameter and 104 mm long with a mass of 1.72 kg. It is wrapped in an Al shell. The total mass of uranium in one section is 104.92 kg. The front fifth section has the cylindrical input beam channel of diameter 8 cm that consists of 54 uranium rods. Thus the total mass of uranium in the target assembly is 512.56 kg. The uranium target is surrounded by a lead blanket 10 cm thick.

2. Analysis and results

The QUINTA set-up was directly irradiated by deuterons with energies 4 and 8 GeV with integral beam intensities 2.73(10)E+13 and 9.1(4)E+12 respectively. Neutrons were produced by way of spallation, fission and evaporation processes after d+Unat collisions. Al foils with diameter 21 mm and about 4.5 mm thick were placed on the surface of TA QUINTA.

The reaction rate R is defined as the number of residual nuclei produced per sample atom per incident deuteron per second. The number of produced residual nuclei (^{24}Na, ^{22}Na and ^{7}Be) is established by measuring their emitted gamma-rays with an HPGe detector. All three measured reaction rates R together with their uncertainties and the respective calculated values with MCNPX are given in Table 1.

An analysis of the data represented in the table 1 leads to very important conclusions on the dependence of the shape of the neutron spectrum on the incident energy. While the ratio R(8GeV)/R(4Gev) for ^{24}Na having a low reaction threshold is less than two, for ^{22}Na and ^{7}Be product nuclei with much higher thresholds these ratios reach about 5 and 7, respectively. This gives evidence that the neutron spectrum on the surface of TA QUINTA becomes «harder» with increase of incident deuteron energy. Other essential conclusions are related to the inability of the code used to reproduce above dependence of the high energy tail of the neutron spectrum on incident energy.

Table 1. Comparison of the experimental and calculated rates of $^{27}Al(n,y1)^{24}Na$, $^{27}Al(n,y2)^{22}Na$ and $^{27}Al(n,y3)^{7}Be$ reactions obtained for 4 GeV and 8 GeV deuteron energies in units of E-29/atom/deuteron/s.

Product nucleus	Reaction rates	E_d = 4 GeV	E_d= 8 GeV	R(8 GeV)/R(4 GeV)
^{24}Na	R_1(exp)	7.5(4)	12.2(5)	1.64(12)
	R_1(calc)	7.473	16.75	2.24
^{22}Na	R_2(exp)	0.522(29)	2.73(22)	5.2(5)
	R_2(calc)	0.71	1.73	2.43
^{7}Be	R_3(exp)	0.069(11)	0.47(8)	6.8(15)
	R_3(calc)	0.0274	0.0736	2.69

The measured reaction rate R for a given produced nucleus ^{Z}A created in the $^{27}Al(n,y)^{Z}A$ process is a convolution of energy E_n of the neutron flux Φ [n/cm^2/MeV/deuteron/s] and the respective cross sections $\sigma_i(E_n)$ leading to the nucleus ^{Z}A.

Assuming that the neutron flux $\Phi_3(E)$ in the third energy interval (^{7}Be production) is constant and denoting this as Φ_3, it is possible to express the reaction rate for ^{7}Be production as:

$$R_3 = \int_3^4 \sigma_3(E)\phi_3(E)dE \cong \phi_3 \sum_3^4 \sigma_3(E)\Delta E = \phi_3 X_{34}(3), \tag{1}$$

where the limits of integration or summation are from 119 MeV up to E_{max} that is defined as the effective end of neutron spectrum (see below). The value $\sigma_3(E)$ denotes the sum of all cross-sections leading to final nucleus ^{7}Be at neutron energy E and $X_{34}(3)$ represents the sum of $\sigma_3(E)$ over whole third energy interval. So the neutron flux Φ_3 may therefore be calculated as:

$$\phi_3 = R_3 / X_{34}(3). \tag{2}$$

In the second neutron energy range the reaction rate for ^{22}Na production can be written as follows:

$$R_2 = \phi_2 X_{23}(2) + \phi_3 X_{34}(2), \tag{3}$$

where, as above, from Eq. (3) it has been assumed that the neutron flux Φ_2 is constant over the second energy interval. So from (3) one obtains:

$$\phi_2 = \{R_2 - \phi_3 X_{34}(2)\} / X_{23}(2) \tag{4}$$

Lastly, the reaction rate for ^{24}Na production is expressed as:

$$R_1 = \phi_1 X_{12}(1) + \phi_2 X_{23}(1) + \phi_3 X_{34}(1) , \tag{5}$$

that gives the neutron flux Φ_1 in the form:

$$\phi_1 = \{R_1 - \phi_2 X_{23}(1) - \phi_3 X_{34}(1)\} / X_{12}(1). \tag{6}$$

To extract information about the neutron spectra following the procedure described above one needs to obtain initially the relevant sums of the corresponding cross-sections in all three energy intervals. The calculations of the sums of cross-sections $X_{12}(1)$, $X_{23}(1)$ and $X_{23}(2)$ were performed with the use of experimental data from EXFOR and Hansmann (2010). The parts of sums $X_{34}(3)$ for the neutron energy range (119 to 200) MeV were calculated using the deterministic code TALYS 1.4 (see Koning et al.(1998)). The calculated sums of cross-sections are given in Table 2.

Table 2. The sums of cross sections, X_{12}, X_{23} and X_{34} in barn.

Product	X_{12}	X_{23}	X_{34} E_d = 4 GeV	X_{34} E_d = 8 GeV
^{24}Na	1.08(11)	1.08(11)	7.23(59)	9.20(75)
^{22}Na		1.23(15)	7.48(74)	9.48(67)
^{7}Be			1.72(20)	2.60(26)

The results for the neutron fluxes Φ_1, Φ_2 and Φ_3 calculated by formulas (6), (4) and (2), respectively, with use the data of Table 2 as well the experimental reaction rates from Table 1 are given in Table 3.

Table 3. The deduced average neutron fluxes Φ_i(exp) in the units [neutrons/cm^2/MeV/deuteron/s] 10^{-5} and their calculated values for the deuteron energies 4 and 8 GeV.

Product nucleus and neutron energy range	Fluxes	E_d = 4 GeV	E_d= 8 GeV	Φ (8 GeV)/Φ (4 GeV)
^{24}Na	Φ_1(exp)	4.8(8)	8.1(13)	1.69(39)
5MeV-27MeV	Φ_1(calc)	5.160	12.91	2.50
^{22}Na	Φ_2(exp)	0.18(7)	0.83(35)	4.7(5)
27MeV-119MeV	Φ_2(calc)	0.261	0.550	1.11
^{7}Be	Φ_3(exp)	0.040(8)	0.18(4)	4.5(14)
119MeV-800MeV	Φ_3(calc)	0.00271	0.00370	1.36

An analysis of the data shows that the deduced values of Φ_1 depend slightly on the choice of the effective end of neutron spectrum E_{max} but the fluxes Φ_2 and especially Φ_3 change significantly for two variants of the E_{max} choice. To choose between these options we take into account that the calculated reaction rates R_3 for ^{7}Be production presented in Table 1 changed by less than 1% as the E_{max} value increased from 800 (1000) MeV to 2(4) GeV.

One can see from Table 3 that if the flux Φ_1(exp) increases about a factor of two with increased deuteron energy from 4 to 8 GeV, the Φ_2(exp) and Φ_3(exp) values increase more than four times. It confirms the conclusion deduced above (see the discussion of Table 1) that the neutron spectrum in TA QUINTA becomes more “hard” with an increase of incident energy. Note that as in the case of the $^{27}Al(n,y_3)^7Be$ reaction rates the fluxes Φ_3(calc) do not reproduce absolute values of Φ_3(exp) nor their dependence on deuteron energy.

The calculations of the neutron spectra were performed with the use of the Monte Carlo code MCNPX 2.7, see D. Pelowitz (2011), combining the INCL4 intra-nuclear cascade model with the ABLA fission-evaporation model and cross-section libraries LA150 (up to 150 MeV) and ENDF/B-VII (up to 20 MeV). The geometrical and physical properties of the QUINTA setup and its materials were supplied to the code in an input file.

3. Conclusion

The experimental reaction rates for production of ^{24}Na, ^{22}Na and ^7Be in the ^{27}Al samples irradiated by neutrons produced in TA QUINTA when it is bombarded with deuteron beams from JINR Nuclotron have been measured in the range of deuteron energies (2 ÷ 8) GeV.

If the ratio R(8GeV)/R(4GeV) for ^{24}Na production (having a low reaction threshold 5 MeV) is less about two so for ^{22}Na and ^7Be product nuclei with much higher thresholds (25 and 119 MeV respectively) these ratios reach about 4.7 and 4.5 correspondingly. It gives strong evidence that the neutron spectrum on the surface of TA QUINTA becomes «harder» with increase of incident energy.

At 4 GeV deuteron energy for the effective thresholds $E_{th,eff} \leqslant 27$ MeV the MCNPX2.7 code reproduces R values rather well while for $E_{th,eff} = 119$ MeV the disagreement reaches 15. In the case of 8 GeV deuteron energy for highest thresholds of ^7Be the exp/(calc) ratio increases up to 5.0. This means that the used code is not able to reproduce the high energy part of the neutron spectrum of TA QUINTA.

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