Experimental Study of the Physical Properties of ADS Systems – Measurement of High Energy Neutron Flux Density by Using the $^{89}\text{Y}$ Threshold Detectors

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ABSTRACT

Study of deep subcritical electronuclear systems and radioactive waste transmutation using relativistic beams from the accelerator was performed. This work is a preliminary step toward the study of the physical properties of ADS systems (Accelerator Driven Systems). It is aimed to obtain fast neutron energy spectra inside the volume of the assembly using (n,xn) threshold reaction. Due to those experiments we can determine optimal energy for ADS systems and obtain the best position for transmutation processes. Normally in experiment like this one uses neutron activation foils made of gold ($^{197}\text{Au}$), cobalt ($^{59}\text{Co}$), bismuth ($^{209}\text{Bi}$). New type foil, yttrium ($^{89}\text{Y}$) was proposed. Yttrium detectors give good results for neutron energy from 10 MeV to about 50 MeV.

Results of few experiments with $^{89}\text{Y}$ foils are presented. The uranium experimental assembly “QUINTA” (consists of 512 kg of natural uranium rods arranged hexagonally and surrounded by a lead reflector) was irradiated with Dubna NUCLOTRON with 2, 4 and 8 GeV deuteron beam. Neutron energy spectrum inside whole 3D model was obtained with $^{89}\text{Y}$ threshold energy reaction. Also the comparison of the average neutron flux density per deuteron in three neutron energy ranges for the different deuteron beam is presented. Cross section data for neutron induced reactions were calculated using the TALYS code. We have started checking experimentally those cross section values.

1 INTRODUCTION

The main advantage of ADS systems is that a subcritical active core plus an external neutron source (accelerator) are more safe than other. The high-energy neutrons inside ADS systems are an ideal tool to induce fission in most trans-uranium isotopes and thus transmute most of the dangerous radioactive wastes from nuclear power station and other sources.

Study of deep subcritical electronuclear systems and their application for radioactive waste (RAW) transmutation using relativistic beams from the JINR Nuclotron, which we are involved in, is performed within the E&T RAW collaboration. This work is a one of the steps the study of the physical properties of ADS systems. Our deeply subcritical active core made of natural uranium is irradiated by a high energy pulsed beam of relativistic deuterons or protons.

We have started with experiments which refer to the study of neutron spectra at various points inside the volume of the assembly QUINTA (Fig. 1.).
EXPERIMENTS

A new “ready to use” assembly Quinta, located at the Joint Institute for Nuclear Research (JINR), Dubna, Russia, has been available for the E&T RAW collaboration since the end of the year 2009. It is an assembly massive uranium target and lead shielding (Fig 1). The Quinta assembly, consists of 512 kg of natural uranium (the level of sub criticality is about 0.2). Quinta assembly is divided on five sections. Each section has 114 mm length and are separated by a 17 mm air gap for the placement of our samples (activation detectors) mounted onto special plates (Fig. 1-2). We have 6 plates (measurements positions) because we have 4 gaps between assembly sections and two positions in front of and rear assembly. The natural uranium is inside aluminum cylinders, where each cylinder is 36 mm of diameter, 104 mm of length and has 1.72 kg mass. These cylinders are inclusive of a 1 mm thick outer aluminum cover for safety reasons. Quinta sections have 61 uranium cylinders enclosed in a hexagonal aluminum container with a wall thickness of 5 mm. Only the first section has 54 cylinders (removed the central 7 cylinders to create a beam window). This beam window is 80 mm of diameter and serves to reduce the loss of backward emitted/scattered neutrons. [2]

The Quinta assembly (target-blanket) has 638 mm length and mass of 538 kg including all construction materials. The assembly is surrounded by lead bricks 100 mm thick on all six sides of total weight 1780 kg. This working as a neutron reflector and as a biological safety shielding. In the front of the lead box is a square window 150 x 150 mm. Plates with samples can be removed quickly from the air gaps between the sections. The sample plates have numbers 0–5, starting from the direction of the accelerator beam (Fig. 2). [2]

The Quinta assembly was irradiated by deuteron beam with energies 2, 4 and 8 GeV. Total number of deuterons of the three irradiations are equal to 3.02(30)x10^{13}, 2.73(27)x10^{13} and 0.91(9)x10^{13} during the time of irradiation equal to 6.27h, 9.35h and 16.7h, respectively.

The total number of deuterons to hit the Quinta target was determined by the activation of aluminum via beam induced $^{27}$Al(d,3p2n)$^{24}$Na reactions. The 3 aluminum monitors were placed between the beam output and the front of the Quinta assembly. [2]

A total of twelve $^{89}$Y activation foils (purity > 99.99%) were placed in the Quinta target on the detector plates in front of, between the five sections, and on the rear of the target in two radial positions (4 and 8 cm) (Fig. 2) from the deuteron beam axis for each irradiations. Foils had form of pills made of compressed yttrium powder with dimensions diameter $9 \times 1.5$ mm$^3$, with weight $\sim 0.6 – 0.8$ g.
After each experiment, the foils were transported away for analyzing using a HPGe detector. Measurements began about 1 h after irradiation and continuing by few days. All spectra were analyzed using the DEIMOS [3] program. Finally we calibrate all the results to B parameters by the below calibration formula (1). Spatial distributions of $^{88}\text{Y}$, $^{87}\text{Y}$, $^{86}\text{Y}$ and $^{85}\text{Y}$ isotope production for the deuteron beam of 2, 4 and 8 (December 2012) GeV in the Quinta assembly were measured. The error of spatial distributions of $^{88}\text{Y}$, $^{87}\text{Y}$, $^{86}\text{Y}$, $^{85}\text{Y}$ isotope production are from 7E-08 to 1E-06 (error range 10-20%) it deepens of the point.

$$B = \frac{N_{1}}{m \cdot I} \cdot \frac{\Delta S(G) \cdot \Delta D(E)}{N_{abs} \cdot \varepsilon(E) \cdot COI(E,G)} \cdot \frac{(\lambda \cdot t_{ira})}{[1 - \exp(-\lambda \cdot t_{ira})]} \cdot \exp(\lambda \cdot t_{real}) \cdot \frac{t_{real}}{t_{live}}$$

(1)

Where

- $B$ number of nuclei per 1 gram of a sample material and per 1 primary deuteron
- $N_{1}$ peak area (line) – number of counts
- $N_{abs}$ the absolute intensity of given line in percent [%]
- $\varepsilon(E)$ detector efficiency function of energy (polynomial)
- $COI(E,G)$ cascade effect coefficient function of energy and geometry
- $\Delta S(G)$ calibrations function for thickness and shape of detectors
- $\Delta D(E)$ calibrations function for self absorption inside the detectors
- $I$ total number of primary deuterons
- $t_{1/2}$ half life time [s]
- $t_{ira}$ elapsed time of irradiation [s]
- $t_{+}$ elapsed time from the end of irradiation to the beginning of measurement [s]
- $t_{real}$ elapsed time of the measurement [s]
- $t_{live}$ “live” time of measurement [s]
- $m$ mass of the sample (target) in grams [g]
Figure 3: Spatial distribution (radial and axial) of $^{88}$Y production (B parameter) for the deuteron beam 8 GeV. On X axis we have number of measurements plates (distance in [cm] 1=13.1, 2=26.2, 3=39.3, 4=52.4 and 5=65.5). Y axis – radial distance in [cm].

All our results can be present in 3D graphs form. An example of the isotope production $^{88}$Y presented in Fig. 3. The maximum yield of production is at about 26 cm from the front of the QUINTA assembly (about 13 cm from $^{238}$U spallation target).

3 EVALUATION OF AVERAGE HIGH ENERGY NEUTRON FLUX IN THE $^{89}$Y FOILS LOCATION INSIDE THE U/PB ASSEMBLY.

Having determined isotope production per one gram of $^{89}$Y sample and per one primary deuteron at specified positions of the Quinta experimental assembly for the three isotopes $^{88}$Y, $^{87}$Y and $^{86}$Y, we can calculate three average high energy neutron fluxes in each $^{89}$Y foils location. The energy ranges are roughly determined by the microscopic cross section in function of energy for the (n,xn) reactions of the three isotopes $^{88}$Y, $^{87}$Y and $^{86}$Y. The three threshold reaction energy 11.5, 20.8 and 32.7 MeV for the reactions $^{89}$Y(n, 2n)$^{88}$Y, $^{89}$Y(n, 3n)$^{87}$Y and $^{89}$Y(n,4n)$^{86}$Y, appoint the first two energy ranges (11.5 - 20.8 MeV) and (20.8 - 32.7 MeV) of the neutron fluxes $\phi_1$ and $\phi_2$. The third energy range begins at the energy 32.7 MeV and ends at about 100 MeV. Microscopic cross section for energy 100 MeV for reaction $^{89}$Y(n,4n) $^{86}$Y is very low.

To calculate the neutron flux density we need the microscopic cross section for the $^{89}$Y(n, xn) reactions. Some cross section data for neutron reactions we can take from the EXFOR data base [4, 5]. Only the data for the cross section of $^{89}$Y(n, 2n) and several points of $^{89}$Y(n, 3n)$^{87}$Y reactions we could find [6, 7]. That is why our cross sections were calculated using the TALYS code [8, 9]. TALYS calculation for energy range 0-200 MeV see Fig. 4.
In general the nucleons number of $^{89}$Y isotopes ($N_y$) in the $^{89}$Y foils of volume $V_p$ [cm$^3$] in the our chosen energy range can be expressed:

$$N_y = B^y W_p S,$$  

(2)

Where:

- $\Phi$ – average neutron flux in the chosen energy range [n/cm$^2$·s]

$$\Phi = (E_2 - E_1) \overline{\psi} \quad ; \quad \overline{\psi} = \frac{\int E \psi(E) dE}{E_2 - E_1}$$

- $\psi(E)$ – neutron flux density [n/cm$^2$·s·MeV]
- $N$ – number of $^{89}$Y isotopes nucleons in volume unit [cm$^{-3}$]
- $\overline{\sigma}$ – average microscopic nucleons cross section for the reaction (n, xn) in the energy range ($E_1 - E_2$) [barns].

$$\overline{\sigma} = \frac{\int E E \sigma(E) dE}{E_2 - E_1}$$

- $\sigma$ – microscopic cross section for the reaction (n, xn) [barns]
- $t$ – deuteron irradiation time [s].

$$N = \frac{\rho_p}{h^9 G} A$$  

(3)

where:

- $\rho_p$ – specific weight of $^{89}$Y [g/cm$^3$],
\(^{89}\)G – gram-atom of \(^{89}\)Y \([g]\)

A – Avogadro’s number,

We assume that the average neutron flux in the chosen energy range \((\overline{\Phi})\) is constant versus time during deuteron irradiation.

From the other side the number \(N_y\) of \(^{88}\)Y, \(^{87}\)Y and \(^{86}\)Y isotopes produced in \((n, xn)\) reactions in the detector we can obtain as:

\[
N_y = B^y W_p S ,
\]

(4)

Where:

\(B^y\) – isotope production inside of samples per one gram of foils and per one beam deuteron,

\(W_p\) – weight of detector \([g]\); \(W_p = \rho_p V_p\),

\(S\) – total number of deuterons.

Combining the equation (2) with the equation (4) we obtain:

\[
\overline{\Phi} = \frac{B^y S \; ^{89}G}{\sigma A t} \; [1/cm^2\cdot s]
\]

(5)

The above equation (5) pertains only to one assumed isotope. When we consider three isotopes then we have to solve three equations. First of all we have to define the three energy ranges for the three average neutron fluxes which are of our interest. By choosing the first three threshold energies \(E_1=11.5\) MeV, \(E_2=20.8\) MeV and \(E_3=32.7\) MeV for the reactions \(^{89}\)Y\((n, 2n)\; ^{88}\)Y, \(^{89}\)Y \((n, 3n)\; ^{87}\)Y and \(^{89}\)Y\((n,4n)\; ^{86}\)Y and the fourth \(E_4=100\) MeV where the microscopic cross section is comparatively low with the maximum cross section of \(^{89}\)Y\((n,4n)\; ^{86}\)Y reaction we have defined the three average neutron fluxes \(\overline{\Phi}_1, \overline{\Phi}_2, \overline{\Phi}_3\) which are of our interest. Finally we can write the three algebraic equations:

\[
B^{88} C = \overline{\Phi}_1 \; \sigma_{11} + \overline{\Phi}_2 \; \sigma_{12} + \overline{\Phi}_3 \; \sigma_{13}
\]

(6)

\[
B^{87} C = 0 + \overline{\Phi}_2 \; \sigma_{22} + \overline{\Phi}_3 \; \sigma_{23}
\]

(7)

\[
B^{86} C = 0 + 0 + \overline{\Phi}_3 \; \sigma_{33}
\]

(8)

Where:

\(B^{88}, B^{87}, B^{86}\) – measured isotopes of \(^{88}\)Y, \(^{87}\)Y and \(^{86}\)Y respectively per one gram of foils and per one beam deuteron

\[
C = \frac{S \; ^{89}G}{A t}
\]

\(\sigma_{11}, \sigma_{33}\) – microscopic cross section of the measured isotopes for the reaction \((n, xn)\) in the three chosen energy ranges,

\(\overline{\Phi}_1, \overline{\Phi}_3\) – unknown average neutron fluxes in the three chosen energy ranges.
Solution of the three algebraic equations give us the average neutron fluxes in the three energy ranges expressed in \([\text{n/cm}^2\cdot\text{s}]\):

\[
\bar{\phi}_1 = \frac{C}{\sigma_{11}} \left[ B^{88} - B^{87} \frac{\sigma_{12}}{\sigma_{22}} + B^{86} \left( \frac{\sigma_{21} \sigma_{15}}{\sigma_{33} \sigma_{22}} - \frac{\sigma_{13}}{\sigma_{33}} \right) \right]
\]

(9)

\[
\bar{\phi}_2 = \frac{C}{\sigma_{22}} \left[ B^{87} - B^{86} \frac{\sigma_{23}}{\sigma_{33}} \right]
\]

(10)

\[
\bar{\phi}_3 = \frac{C}{\sigma_{33}} B^{86}
\]

(11)

Number of measured isotopes in the detector assigns the number of algebraic equations what in turn assigns the number of unknown neutron fluxes in the chosen neutron energy ranges which are possible to be evaluated. The more of measured isotopes in the detector the more precise evaluation of the high energy neutron spectrum is obtained. Using the microscopic cross sections for the reactions \(^{89}\text{Y}(n, 2n)\), \(^{(n, 3n)}\) and \((n,4n)\) generated by TALYS code (Fig. 4) and the experimental data (parameter B) we have evaluated the average high energy neutron flux in the \(^{89}\text{Y}\) foils located inside the Quinta assembly for the three energy ranges (11.5 - 20.8 MeV), (20.8 - 32.7 MeV) and (32.7 - 100 MeV). An example of comparison of average neutron flux density per unit energy of deuteron from accelerator deuteron beams with energy 2.0, 4.0 and 8.0 GeV in function of Quinta target axis at R=4 cm and the energy range 20.8 - 32.7 MeV is presented in Fig. 5. The curves of the neutron flux density per deuteron and its energy (2, 4 and 8 GeV) overlap in the energy ranges 20.8 - 32.7 MeV and nearly overlap in the energy range 11.5 - 20.8 MeV and 32.7 - 100 MeV for radius 4 cm in the area close to the spallation target. Another comparison we can find in [11].

4 CONCLUSION

The presented here research results are of great importance for future use ADS for long lived nuclear waste utilization. ADS parameters like what target material to be used, what impinging particle, what particle energy, what geometry should be selected to get the highest transmutation efficiency.

The general feature of the experimental spatial distribution of \(^{88}\text{Y}\), \(^{87}\text{Y}\), \(^{86}\text{Y}\) and \(^{85}\text{Y}\) isotopes production is that the maximum yield is at about 13 cm from the front of the \(^{238}\text{U}\) spallation target and that the yield is decreasing with increasing radial distance from the target axis. The same distance was observed when we used lead spallation target.

The shape of neutron flux density per deuteron in the Quinta assembly produced by the neutrons generated in the assembly irradiated by the relativistic deuteron beam of 2 GeV, 4 GeV and 8 GeV energies in general is the same.

Transmutation yield normalised to 1 GeV beam energy is generally the same for each beam energy from about 1 GeV (bigger energy give us similar transmutation efficiency). Energy range 1-1.5 GeV seems to be the optimal energy for ADS installations. No specific difference was observed in transmutation rate comparing proton (older experiments [11]) and deuteron beam.
Based on theoretical calculations natural uranium target should be more effective in spallation neutron production than the lead one. The experiment shows that neutron production is more sensitive to beam justification – beam direction and shape.

The experimental setups should emulate the future ADS reactors as close as possible. Therefore JINR Dubna planes a series of experiments with much bigger natural uranium setup BURAN, simulating fast reactor.

![Average neutron flux density per deuteron and its energy in function of Quinta target axis at R=4 cm for three deuteron energies (2, 4, 8 GeV) in the neutron energy range (20.8 - 32.7 MeV). Errors range 5-11%.

Figure 5: Average neutron flux density per deuteron and its energy in function of Quinta target axis at R=4 cm for three deuteron energies (2, 4, 8 GeV) in the neutron energy range (20.8 - 32.7 MeV). Errors range 5-11%.

REFERENCES


[5] Experimental Nuclear Reaction Data; EXFOR/CSISRS


