METHOD OF A WATER SAMPLES PREPARATION WITH HIGH CONCENTRATION OF ^{16}N

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ABSTRACT

On the basis of the accelerator KG-2,5 in IPPE created installation, allowing to obtain water samples with high concentration of ^{16}N. The article describes a technique and some of the experimental and calculated data.

INTRODUCTION

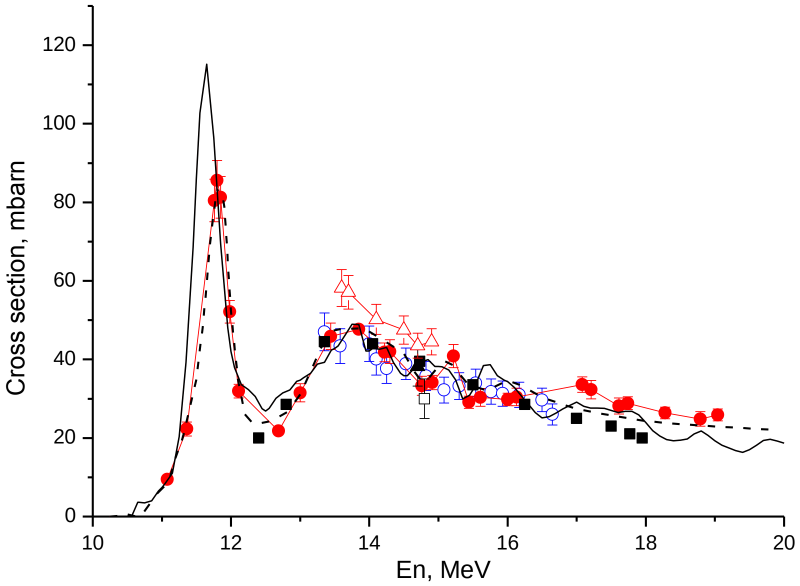
One of the leakage detection method and coolant flow meter in primary circuit of WWR is radioactivity control of ^{16}N in steam (correlation method). ^{16}N is produced in the reaction ^{16}O(n,p) as a result of fast neutron interaction with coolant (H_2O):

$$^{16}O + n \rightarrow {}^{16}N + p,$$
$$^{16}N \rightarrow {}^{16}O^* + \beta^- + \tilde{\nu},$$
$$Q = 10.4\ MeV.$$

E_{max} of β-particle in the decay of ^{16}N is 10.4 MeV. The oxygen (^{16}O*) in excited state decays with γ-rays emission. Gamma energies are E_γ=6.134 MeV (0.69 γ/decay) and E_γ=7.112 MeV (0.05 γ/decay) [1].

The only suitable way for calibration of correlation meter, is the creation of the installation that will contain radioactive nuclei in the circulating water and the decay of such nuclei followed by γ emission with energies close to or equal 6.13 MeV.

On the other side, the experimental data set for the ^{16}O(n,p)^{16}N reaction is very limited, and there are significant differences in estimations of different libraries (Figure 1). Thus, the possibility of a water samples preparation with high concentration of ^{16}N is interesting from a practical and scientific point of view.



Solid points – [2], open points – [3], solid triangles – [4], solid squares – [5], open squares – [6], open triangles – [7], solid line – ENDF B VII, dashed line – BROND.

Figure 1 – ^{16}O(n,p)^{16}N cross section – experimental points and theoretical evaluations.

EXPERIMENTIAL METHOD

The installation for measurement of ^{16}N concentration consists of: 1) accelerator KG-2,5; 2) target site, designed for fast neutrons generation; 3) reaction chamber filled with water for ^{16}N generation; 4) water transit system from the place of exposure to the measuring site; 5) hard γ-radiation spectrometer based on the NaI(Tl) crystal scintillation spectrometer.

The algorithm is following: accelerated deuterium ions (from KG-2,5) with energies of 1.4 MeV are sent on the lithium neutron generating target. Deuteron-lithium interactions are the source of neutron from $^{7}Li(d,n)2^{4}He$ (15.05 MeV) and $^{7}Li(d,n)^{8}Be$ (14.91 MeV) reactions. (Figure 2).

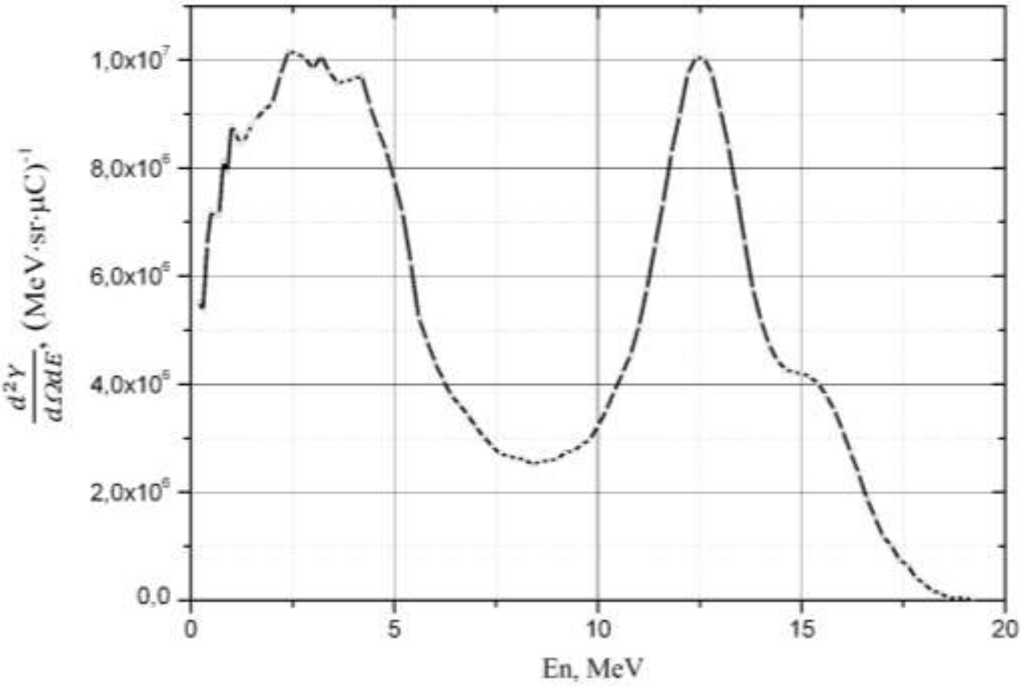
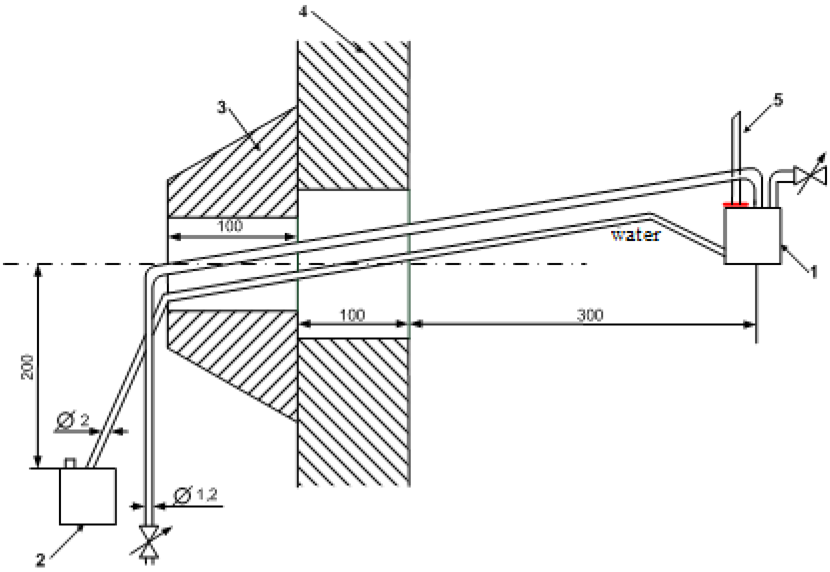


Figure 2 – Neutron energy spectrum from $^{7}Li(d,n)$ reaction (0º to the axis of the deuteron beam).

Then neutrons penetrate into a sealed reactions chamber, filled with water, in which the reaction $^{16}O(n,p)^{16}N$ takes place – source of ^{16}N nuclei [8]. Irradiation are conducted periodically, duration of each exposure is about 40 seconds (more than five ^{16}N half-lives). At the end of exposure water with ^{16}N moved from a position of irradiation to the measurement position. This is required to protect the measurement process from electromagnetic interference that exists near accelerator site. Time of delivery in all measurement not exceed three seconds. ^{16}N concentration, obtained in the water in the process of fast neutrons irradiation, is measured by means of γ- rays produced in the ^{16}N decay. Registering the event correlated to the full absorption of gamma-quanta with energy 6.13 MeV in the detector. The effectiveness of registration in the conditions of the known experiment geometry is known. That is why it is possible to determine the concentration of ^{16}N impurities in the measured sample of water. Adjusting to the time of sample delivery from exposure to measurement site and to the volume of the transferred water, we can define the initial concentration of ^{16}N at the end of exposure.

Figure 3 presents setup diagram for numerical measurement of the formed nuclei ^{16}N.



1 – water tank on the position of irradiation, 2 – water tank on the position of measurement, 3 – paraffin + B_4C, 4 – concrete, 5 – target.
Figure 3 – Scheme for measuring concentration of ^{16}N in water, after irradiation by fast neutrons.

In a room intended for activity measurement, there are two detectors hard gamma-quanta, one of which is based on a crystal of NaI(Tl) and the other from high-purity germanium (HPGe) and intended for spectrometry electronics. The signals from the detectors are passed in measuring room in which located the computer executing spectra accumulation registered radiation (in seconds).

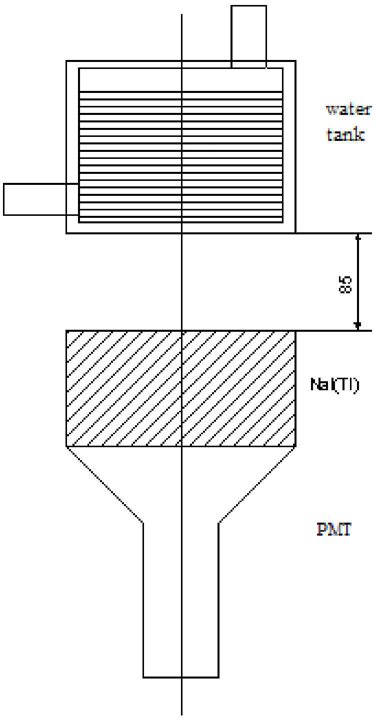


Figure 4 – Scheme of the mutual location of the water tank for measurement and detector.

The composition spectrometer with crystal NaI(Tl) (height – 100 mm, diameter – 150 mm) is PTM-49B, preamplifier, amplifier 1101, ADC712. Location of the detector relative chamber for irradiated water is presented in Figure 4. With such geometry managed to achieve necessary for the conduct of such measurements geometric registration effectiveness and to ensure the minimum time of transportation of irradiated water.

The second detector (HPGe) was situated at the distance of 80 cm from the tank side wall. In standard delivery detector of high-purity germanium (CAMBERRA GX5019 in the cryostat model 7500SL with preamplifier model 2002CSL), digital spectra analyzer DSA-1000. The size of Ge crystal: diameter – 65 mm, height – 65 mm.

For neutron flux measurements flat ionization chamber with a layer of uranium-238, posted on the cathode was used.

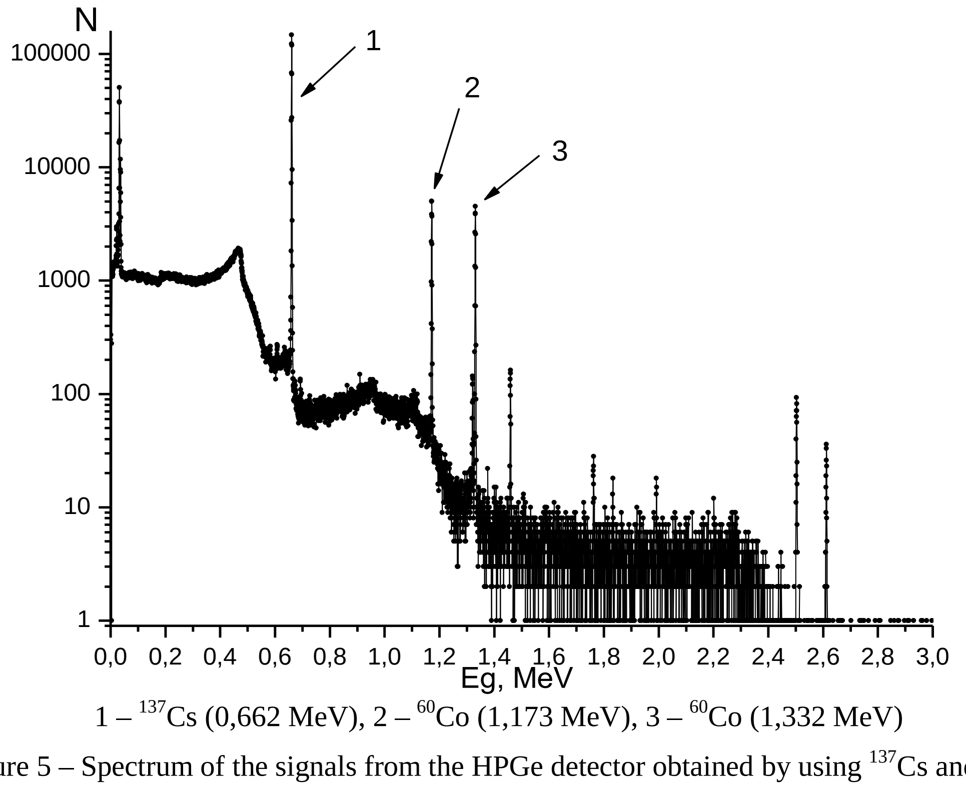
In the experiment used three different tank from stainless steel for water irradiation. Their geometrical dimensions presented in Table 1 (wall thickness – 3 mm).

Table 1 – Parameters of water tank, used in the experiment.

Tank №	Height, H, mm	Diameter, R, mm
1	150	150
2	110	175
3	300	100

RESULTS

The experiment consisted of two stages. In the first stage were measured with HPGe using tank 1 (Table 1). Spectrometer was calibrated, conducted measurement of the background (100 s) and γ-radiation spectra measurement from a water sample, irradiated by fast neutrons (t_{irr}=40 s, $t_{measurement}$=100 s, 100 μA current). Corresponding spectra are shown in Figures 5, 6 and 7.



1 – ^{137}Cs (0,662 MeV), 2 – ^{60}Co (1,173 MeV), 3 – ^{60}Co (1,332 MeV)

Figure 5 – Spectrum of the signals from the HPGe detector obtained by using ^{137}Cs and ^{60}Co.

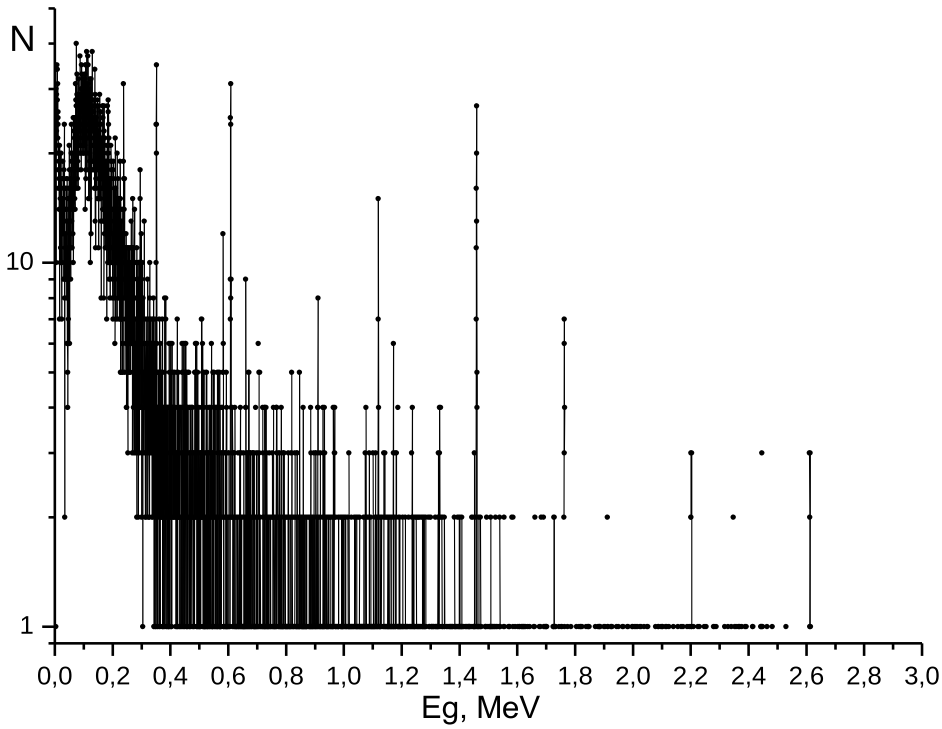
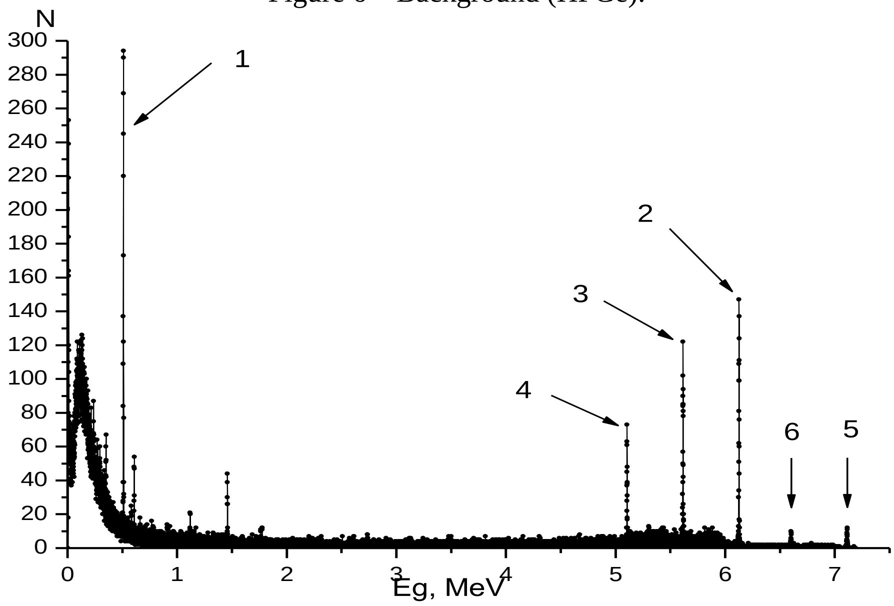


Figure 6 – Background (HPGe).



1 – annihilation peak, 2– total-absorption peak (6.13 MeV), 3 – single leakage peak of photons with energy 6.13 MeV, 4 – double leakage peak of photons with energy 6.13 MeV, 5 – total-absorption peak (7.11 MeV), 6 – single leakage peak of photons with energy 7.11 MeV.

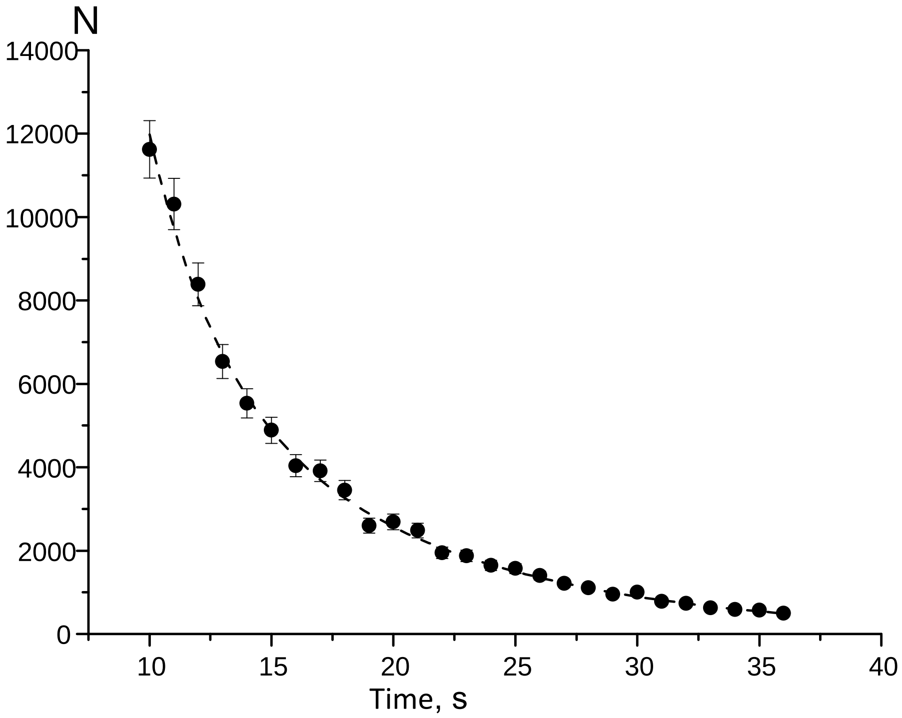
Figure 7 – The spectrum from the HPGe detector obtained from water samples irradiated with fast neutrons.

4.3 keV/channel, energy resolution defined by ^{60}Co (peak with energy 1.332 MeV), amounted to 2 keV.

From these figures it is clear that fast-neutron irradiation water radiates intensively hard γ-rays. Group of lines in the range from 5 to 7.5 MeV was not shown in the background measurements. For obtained spectrum analysis using data on the ^{16}N dacay probability [9].

Analyzing the obtained results, we can conclude that at energies above 2 MeV we observe only activity due to decay of the ^{16}N nuclei. This fact allows us to use a scintillation detector based on crystal NaI(Tl), which has a much worse energy resolution, but more efficient registration. Abnormal γ-lines in the hard part of the spectrum allows to determine the yield of ^{16}N simple integration of all events, the energy of which exceeds the threshold of 2 MeV.

In the second stage measurements have been performed for all three samples of water using a spectrometer with a NaI(Tl). Current on the target was 25 μA. For each experiment, measurements were performed for 30 seconds at 1 second intervals. This allows us to restore the ^{16}N decay curve, the disclosure of which exponent formed least squares method made it possible to determine the initial activity of the sample water. The measurement results for the tank 1 shown in Figure 8.



Point – an experiment, dashed line – curve obtained by least squares.
Figure 8 – The ^{16}N decay curve obtained with irradiation of tank 1.

The measurement results using the method of least squares can be described with expression:

$$N(t) = A_0 e^{-\frac{t}{B}} + N_{\Phi},$$

where A_0 – total number of γ-rays detected in the time immediately after the measurement, N_{ϕ} – counting rate of background events.

Results describing the exponential dependence of the obtained results are shown in Table 2. Numbering of samples corresponds to the Table 1.

Table 2 – Parameters of the initial count rate extracted from the fitting of the decay curves obtained in experiments with 1, 2 and 3 tanks.

Tank №	Initial counting rate (A_0), pulse/s	Initial counting rate error, pulse/s
1	17320	791
2	24450	1328
3	8497	483

The total amount of γ-rays emitted in the initial time can be determined using the expression:

$$\Gamma(0) = \eta \cdot A_0 ,$$

where η - absolute efficiency of γ-ray scintillation detector based on crystal NaI(Tl).

Concentration of ^{16}N atoms, achieved by the time the end of irradiation may be obtained by the following way:

$$N_{16N}(0) = \frac{\Gamma(0) \cdot T_{1/2}}{\beta \cdot \ln 2} = \Gamma(0) \cdot 13.96$$

where β is responsible for the fact that not all of ^{16}N decays accompanied by the emission of γ-rays.

The calculation result of the ^{16}N atoms concentration, obtained by irradiating a different water samples geometries are shown in Table 3.

Table 3 – Achieved experimental ^{16}N concentrations in different geometries (in terms of 100 μA current)

Tank №	Concentration of ^{16}N, atom/litre	Error, atom/litre
1	2.3E7	1.0E6
2	3.2E7	1.7E6
3	1.13E7	6.4E5

To compare the experimental results with the calculations used the code MCNP5, owned by the family of MCNP (Monte Carlo N-Particle Transport Code System, [10-12]), which is currently the most widely used code for the calculations by the Monte-Carlo implementing various schemes acceleration calculations. The calculation has been described in detail the geometry of each experiment, including the linear dimensions of each tank, its wall, the location of the neutron source and its dimensions. Results of calculations are shown in Table 4.

Table 4 – Comparison of the experimental values of the concentration of ^{16}N, obtained in different geometries and calculations made with different nuclear data libraries (in terms of 100 μA current).

Tank №	Experiment, atom/litre	Calculation with ENDF B VII, atom/litre	Calculation with BROND, atom/litre
1	2.3E7	4.5E7	3.9E7
2	3.2E7	3.5E7	3.1E7
3	1.13E7	2.4E7	2.1E7

CONCLUSION

The measurements showed that the established technique can reliably detect induced by fast neutrons in water activity of ^{16}N. For the most optimal geometry of irradiation, which was second tank geometry, with an available mode accelerator deuteron beam current of 1 mA, ^{16}N atom concentration will be $3.2 \cdot 10^8$ atoms per litre of the irradiated water. This value is comparable with the values obtained in the reactor ($5 \cdot 10^9$ Bq/litre) [13].

As shown by calculations, their result is highly dependent on the used nuclear data library. This fact is a consequence of the difference in the description of the reaction cross section taken in different libraries (see Figure 1). Thus, the results of calculations performed with the library BROND and ENDF B VII give relatively similar values for the geometry number 2 ($3.1 \cdot 10^7$ and $3.5 \cdot 10^7$, respectively). The difference in the values in this case is 13%. Geometry number 1 difference between the two variants of calculations is 15 %. Geometry number 3 – 14%. The experimental values differ from the calculations considerably more substantial. Geometry number 1 difference between the calculated value of concentration of atoms ^{16}N, obtained using the library ENDF B VII, and the experiment is 96%. Similar calculations were made with the library BROND, differ from experiment by 69%. Number 2 for the geometry corresponding to the difference is 9% and 3%. Moreover, in this case the library BROND gives a value which is lower than the experimental value. Number 3 for the geometry of the difference between theory and experiment are particularly large – 112% and 86% (library ENDF B VII and BROND, respectively). Analysis of the data shows that the calculations in general, correctly predict the atom concentration dependence of the ^{16}N on the geometry, but in general there is a tendency (except for one case mentioned earlier), according to which the calculated values are higher than the experiment.

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