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COINCIDENCE METHOD FOR DETERMINATION OF RADIONUCLIDES ACTIVITIES

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The radon and radium activity measurements using six-crystal gamma-gamma coincidence, 4 -spectrometer PRIPJAT and radioactivity measurements in different samples of meat and vegetation by 32-crystal spectrometer ARGUS, are described.

Keywords: *"coincidence", "spectrometer", "activity".*

Introduction

A radiation detector with 4 -geometry provides higher efficiency, and therefore shorter counting time than a detector without such geometry. However, its application is limited by the fact that obtained spectrum contains summing peaks of all γ-quanta registered in coincidence. Multiparameter information on coincident photon emission can be obtained only by a detection system where the 4 -geometry is made by many detectors, such are both the PRIPJAT and the ARGUS - γ-coincidence spectrometers of the Crystal Ball type in the Institute of Physics, Minsk [1,2]. There are other characteristics, as background conditions, energy and time resolution, makes it very suitable for investigation of rare decays and interactions, cascade transitions, low intensity radiations etc.

We are developing a method of ^{226}Ra and ^{222}Rn measurement by a multidetector 4 -spectrometer. The method is based on coincidence counting of γ-rays from two-step cascade transitions that follow -decay of ^{214}Bi. Its application to the PRIPJAT spectrometer, which has 6 NaI(Tl) detectors, is presented here, as well as the method of the determination of radionuclide activities based on the registration of the cascades intensity of γ-rays of different multiplicity using spectrometer ARGUS.

Experimental

Spectrometers: PRIPJAT and ARGUS

The outer view of the PRIPJAT spectrometer is shown in Fig. 1, and its structural scheme in Fig. 2. The spectrometer has outer dimensions of 250x145x186 cm^3 and

mass of 4200 kg. It contains 6 NaI(Tl) detectors, modular pulse-processing electronics in the CAMAC standard, and iron and lead passive shielding thickness up to 15 cm. Each of the NaI(Tl) crystals has a diameter of 15 cm and height of 10 cm. The front of each detector is positioned on a side of a cube with an edge of 17.5 cm, closing a solid angle of 0.7x4 sr, and forming a relatively large detection chamber within the spectrometer. 6 ADCs perform pulse height analysis, each with 256 channels. The system has an energy resolution of 10.5 % for the 662 keV line, and a 40 ns resolving time of coincidences.

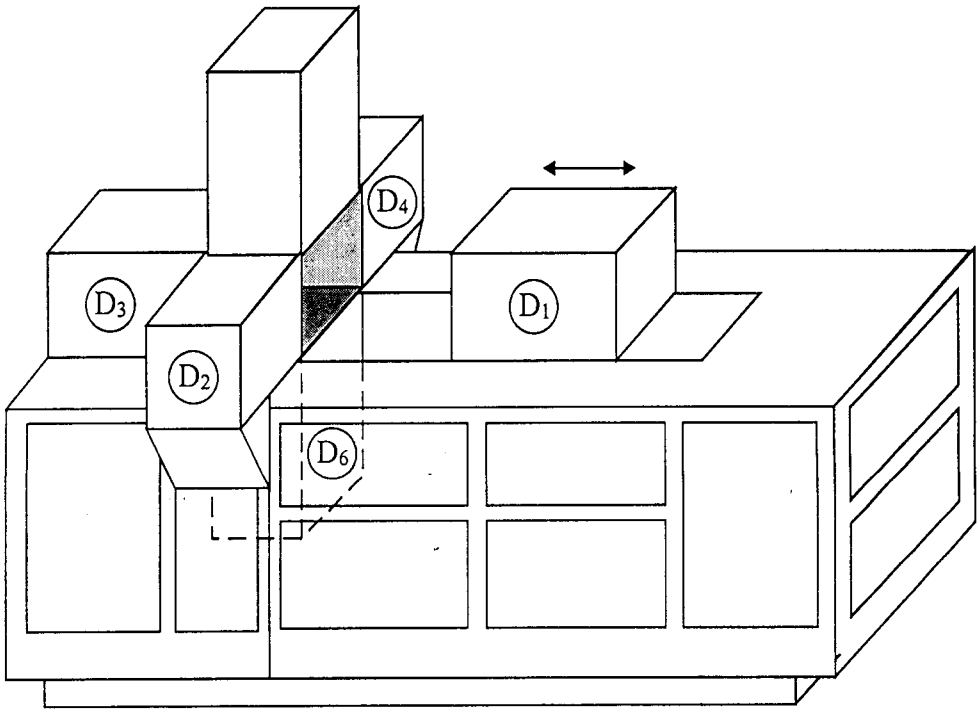


Figure 1. Outer view of the PRIPJAT spectrometer. D_1-D_6 are the measuring sections with NaI(Tl) detectors

The spectrometer registers γ-radiation event in the following way. A signal from the detector is branched and goes to the time and amplitude analysis. Signals from the photomultiplier tubes arrive at the inputs of the correspondent amplifiers. Logic pulses from the amplifier outputs are formed with lengths of 50 and 600 ns. A pulse of 50 ns goes to the input of both the counter and the hodoscope, while a pulse of 600 ns goes to the input of an ADC. Logic outputs with a pulse length of 50 ns are connected individually with corresponding hodoscope inputs. Each of these inputs has an adjustable delay line.

Software of the spectrometer enables two different modes of pulse counting: (1) integral {1-6}, when all pulses coming from the detectors are counted, and (2) coincidence mode, when separate γ-spectra of non-coincident {1-1} and of coincident pulses (with multiplicity from 2 to 6) {2-6} are produced simultaneously. Each of the counting modes gives six spectra from single detectors, as well as their sum spectrum.

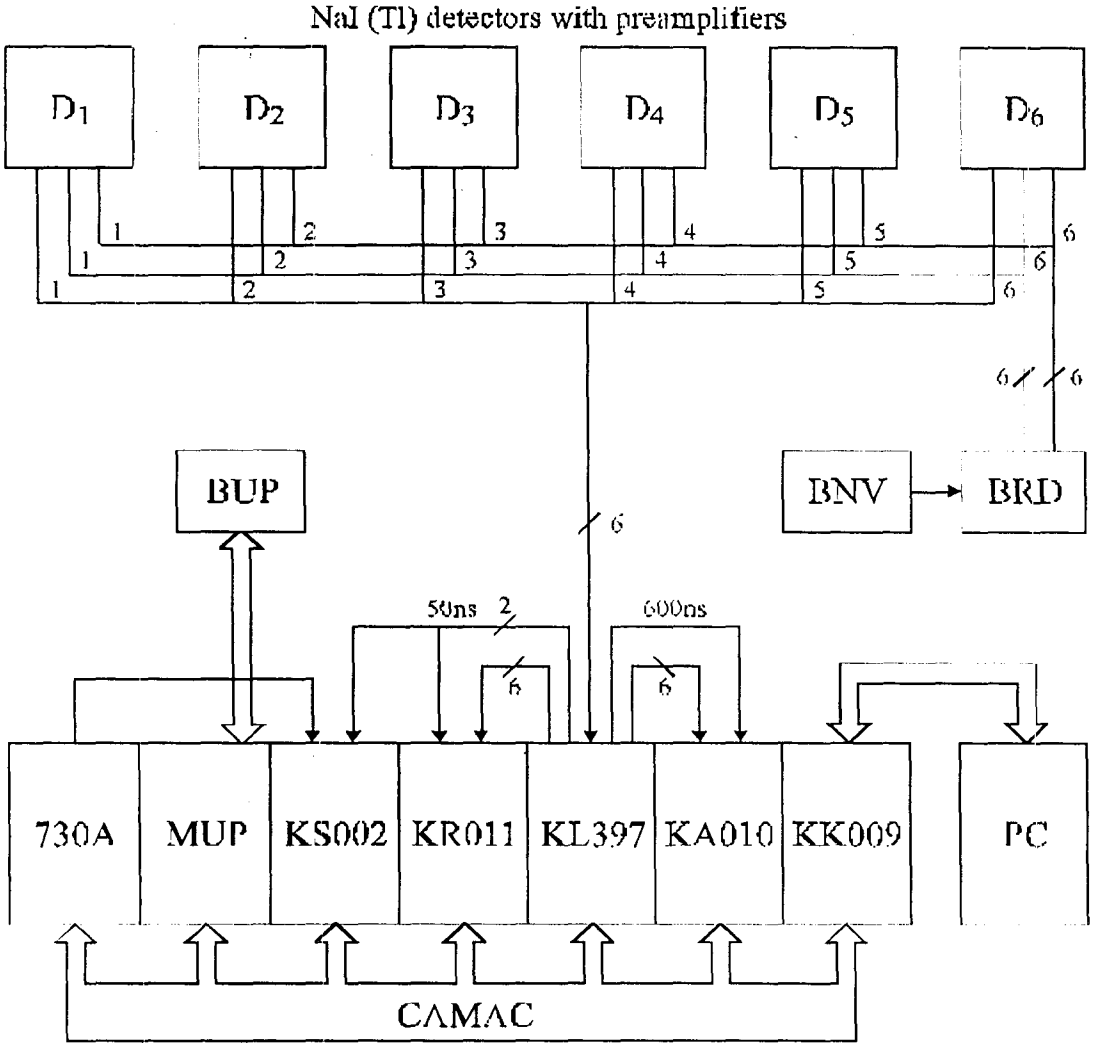


Figure 2. Structural scheme of the spectrometer PRIPJAT. BNV, BRD - high voltage supply; MUP, BUP - moving of the detector unit D_1; KL397 -6-channel amplifier; KR011 - 6-channel hodoscope; KA010 - 6-channel ADC, KS002 -dual counter; 730A-clock generator; KK009 - controller of the CAMAC system.

The spectrometer ARGUS (Fig. 3) consists of 32 pento- and hexahedral truncated steel pyramids, forming a closed cellular surface with internal diameter 0.44 m. Each of the pyramids embeds a NaI(Tl) detector with a 15x10 cm crystal and a low-background photomultiplier FEU-49B. In this view, in case of point source positioned in the center of spectrometer, a counting under solid angle of 0.9x4 sr is enabled.

The electronics (CAMAC standard) and a PC are also parts of the system.

The spectrometer ARGUS registers γ-radiation in the following way. The signals from the detectors pass through the time and amplitude analyses. The signals 50 ns

long, formed from the detector output signals, are entering the inputs of the majority coincidence circuit (MCC) which has capability of selecting event with k-fold coincidences (1 k 5). When a coincidence event of the given k takes place, the identification numbers of the detectors that have registered the coincidence are recording, and the amplitude analyses of the signals from the detectors are performing. The firs operation is carried out by a 32-channel hodoscope, and the second one by the 32 ADCs (256-channels each). The ADCs and hodoscope are controlled by the 600 ns signals from the MCC. Thus, for each coincidence event, the system records the number (multiplicity) and positions (angular correlation) of the detectors which register the event, as well as the signal amplitudes (photon energies).

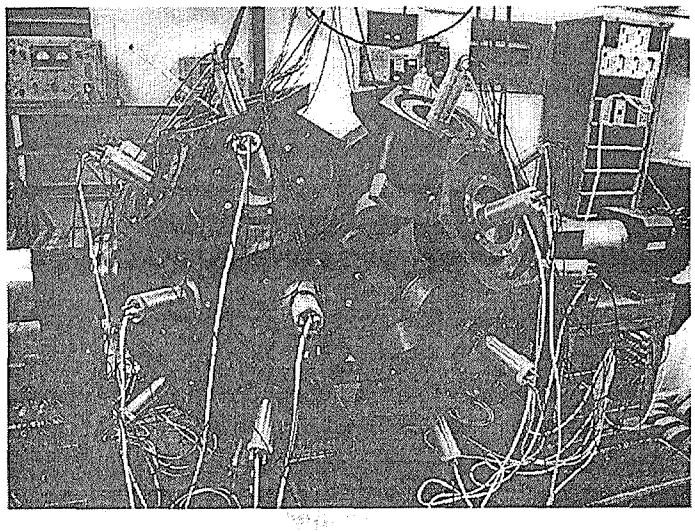


Figure 3. Spectrometer ARGUS

Gamma-background

Background spectra of the spectrometer PRIPJAT (Fig. 4) give the following total count rates in the energy region from 200 keV to 2000 keV: 22.1 counts/s in the integral spectrum {1-6}, 20.1 counts/s in the noncoincidence spectrum {1-1}, and 1.9 counts/s in the coincidence spectrum {2-6}. A theoretical calculation of the minimum detectable activity, as well as experiments, show that detection sensitivity of the PRIPJAT spectrometer when counting double coincidences is one order of magnitude higher than when counting triple coincidences. This means that contribution of the double coincidences to a total coincidence spectrum {2-6} is the highest.

The background spectra of the ARGUS spectrometer in the coincidence modes with multiplicities k = 1, 2, 3, 4 for counting time 2000 s and energy range (200 - 1500) keV, are given at Fig. 5. Background count rates in different working modes are: 378.4 cps for k = 1, 38.6 cps for k = 2, 18.4 cps for k = 3, and 12.6 cps for k = 4.

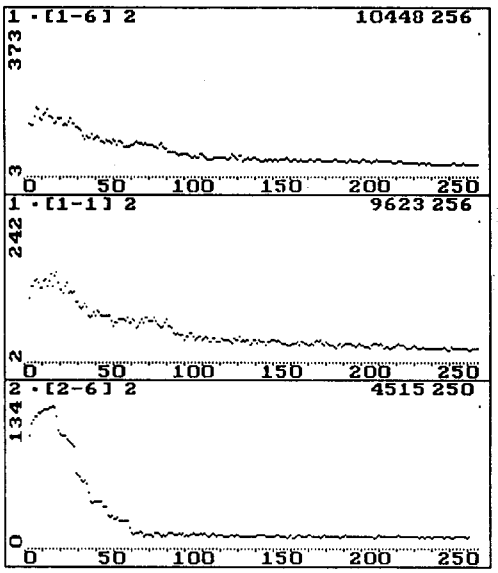
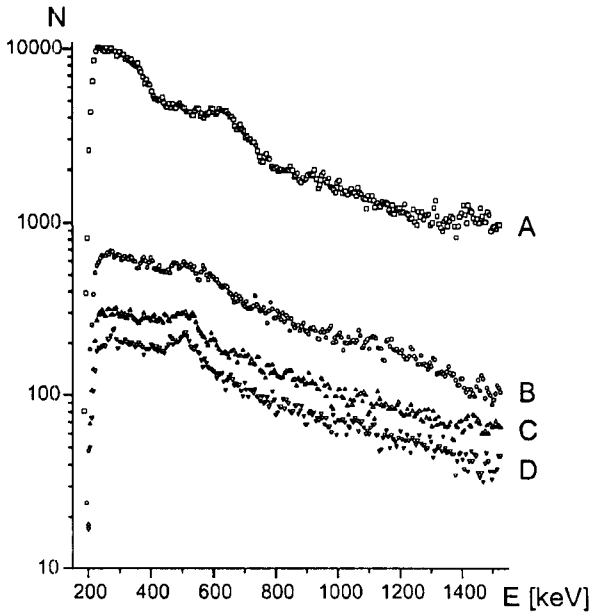


Figure 4. Background spectra of the PRIPJAT spectrometer



A: k=1, B: k=2, C: k=3, D: k=4

Figure 5. Background spectra of the ARGUS spectrometer

Coincidence method of radium and radon measurements

A careful examination of the decay schemes of ^{226}Ra and its descendants shows that there are only a few gamma cascades with transitions which have energies and intensities appropriate for easy detection by a coincidence arrangement with Na(Tl) detectors. All of them follow -decay of ^{214}Bi, and all of them are two-step gamma transitions with the same second transition - from the excited level 609 keV to the ground state of ^{214}Po with relative intensity I = 46. 1 %. Therefore, if radon in a probe

is in radioactive equilibrium with its descendants, the radon activity could be determined by the 609 keV photopeak in a spectrum of double coincidences, i.e., detecting simultaneously the 609 keV γ-ray and any one of the γ-rays in cascade with it. The same is valid for determination of ^{226}Ra in a probe (assuming no losses via radon emanation).

Although the coincidence mode of counting on the spectrometer PRIPJAT generally gives a spectrum of coincidences with multiplicity *m* from 2 to 6, in cases of radon measurement the spectrum is practically produced only by double coincidences. This is because the efficiency of γ-γ coincidence registration strongly decreases with increasing multiplicity of coincidences, and because the intensive cascades of three gamma rays do not exist in the decay of ^{226}Ra and its progeny.

The photopeak efficiency for 609 keV γ-ray detection by the PRIPJAT spectrometer in a different mode of counting has not been determined by theoretical calculations, because the detectors, although of the same type, have somewhat different efficiencies. Therefore, a semiempirical method was preferred, and a photopeak efficiency for 609 keV photon detection is calculated as: (m 1) = 0.101, (m=1) = 0.0675, and (m=2) = 0.0337 [3].

Accuracy of radon determination by coincidence γ-ray spectrometry is tested by measuring a radium probe of known activity (height 1.3 cm, diameter 0.1 cm, mass 0.09 g, activity 63000 Bq). This probe, with ^{226}Ra and ^{222}Rn in radioactive equilibrium, is positioned in the center of detection chamber of the PRIPJAT spectrometer and measured about 50 s in both the integral and coincidence counting modes. In the obtained spectra, the photopeak of ^{226}Ra at 609 keV is the most pronounced in the coincidence spectrum {2-6} (Fig. 6). Ratios of count rates under the 609 keV photopeak in a probe spectrum and under the corresponding energy region in a background spectrum (Fig. 4) are 59 for the integral spectra {1-6}, 38 for the non-coincidence spectra {1-1}, and 257 for the coincidence spectra {2-6}.

The following values of efficiency are obtained after correcting counts in the 609 keV photopeak for losses due to the pulse summing effect: (m 1) = 0.094, (m=1) = 0.062, and (m=2) = 0.035. In all cases the discrepancy between these experimental values and corresponding ones predicted by the coincidence method itself is lower than 9 %, and in the coincidence mode of counting is less than 4 %.

Also, radon determination in water is one of the possible applications of the coincidence method. In our experiment, 1 L of well water was hermetically closed in a glass vessel with height of 12 cm, diameter of 11 cm, and wall thickness of 0.2 cm. Counting started 4 hours later, after radioactive equilibrium between ^{222}Rn and its progeny in the sample was established. The sample was counted 30 minutes in both counting modes on the PRIPJAT spectrometer. The photopeak at 609 keV was the most pronounced in the coincidence spectrum {2-6}, and from the count rate under this photopeak, a radon concentration of (7.7 1.1) Bq/L in the analyzed well water is obtained. We did not apply any correction for gamma-attenuation in this experiment but, on base of an approximate formula taken from Ref. [4], we can estimate that the 609 keV photon attenuation in our 1 l water sample is not higher than 10 %.

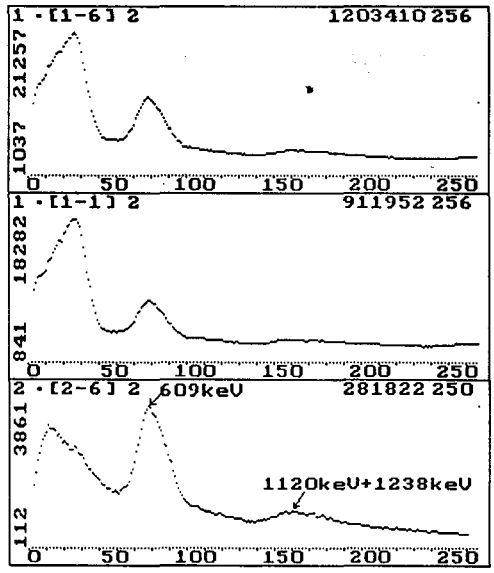


Figure 6. Spectra of the ^{226}Ra probe: {1-6} - integral spectrum, {1-1} - non-coincidence spectrum, {2-6} - coincidence spectrum

Radionuclides activities determination

It is necessary that a spectrometer can register γ-rays of different multiplicities, for using this method. If a sample contains M radionuclides whose activities are A_i, and if they emission cascades of γ-rays of different multiplicities and intensities, number of coincidences (S_k) registered in a measurement time t, is:

$$S_k = (\sum_{i=1}^{M} \varepsilon_{ik} A_i + F_k)t, \; k = 1,2,...,K, \qquad (1)$$

where F_k is background count rate in k coincidence counting mode, $_{ik}$ -registration efficiency, and K - the highest coincidence multiplicity. $_{ik}$ and K are characteristics of the spectrometer, enough precisely determined in a experiment with standard sources.

The error can be estimated:

$$\sigma^2_{A_i} = (\Delta A_i)^2 = \sum_{l=1}^{L} (\frac{\partial A_i}{\partial x_l})^2 (\Delta x_l)^2, \qquad (2)$$

where x_l are absolute errors of the magnitudes (coefficients of the system (1)).

The method is checked at the spectrometer ARGUS. The activities of radionuclides ^{137}Cs (E_γ = 662 keV - intensity 85 %), ^{134}Cs ($E_{\gamma 1}$ = 569 keV – intensity 15.4 % and $E_{\gamma 2}$ = 795 keV – intensity 85.4 %, $E_{\gamma 3}$ = 604 keV – intensity 97.5 %) and ^{106}Ru ($E_{\gamma 1}$ = 512 keV – intensity 20 % and $E_{\gamma 2}$ = 621 keV – intensity 9.8 %) in the

standard mixture, are measured. The experimental results are given in Table 1, and compared to true values show a good agreement.

Table 1.

Standard	t [s]	Activity (A_i A_i), [Bq] ^{137}Cs	^{134}Cs	^{106}Ru
			Standard	
I		7700 400	3100 160	1700 200
II		250 15	160 10	140 15
III		50 5	25 5	-
			Experiment	
I	100	7400 430	3000 175	2000 220
II	100	250 35	160 13	90 3
III	500	67 10	24 7	-

The registration efficiencies $_{ik}$ are experimentally determined using Standard I. The time measurement was 1000 s, and the results are given in Table 2.

Table 2.

Isotope	$_{ik}$ k = 1	k = 2	k = 3
^{137}Cs	0.495	0.009	0.0005
^{134}Cs	0.436	0.256	0.0315
^{106}Ru	0.136	0.036	0.0012

The background count rates in coincidence counting modes k = 1, 2, 3 were: F_k = 480, 35 and 11 s^{-1}, respectively. It confirms the advantages of spectrometer work in coincidence counting mode, compared to integral counting mode.

The measured activities of ^{137}Cs and ^{134}Cs in different samples of meat and vegetation are given in Table 3 and Table 4. These results are compared to the results by Ge(Li) detector and show a good agreement.

Notice: Between brackets errors for the experimental results given by coincidence method (equation (2)), as well as a statistical error (P = 6- 8 %) for the Ge(Li) detector results. Also, it is important, the spectra of these samples are taken two years after Chernobyl's accident.

Table 3. The measured activities in different samples of meat

Sample	Activity, [Bq] (t=100 s) ^{137}Cs	^{134}Cs	Activity, [Bq], Ge(Li) ^{137}Cs	^{134}Cs	t [s]
1	580(30)	220(15)	620(40)	240(20)	300
2	440(30)	160(10)	370(25)	130(15)	300
3	400(30)	150(10)	370(25)	120(10)	600
4	330(20)	130(10)	330(25)	100(10)	600
5	300(20)	120(10)	260(20)	80(10)	600
6	180(25)	70(20)	170(10)	60(5)	1200
7	120(25)	50(5)	130(10)	40(5)	1800
8	110(25)	40(10)	120(5)	40(4)	2000
9	70(20)	30(10)	70(5)	30(3)	3600
10	40(10)	16(4)	40(4)	12(4)	7200
11	30(10)	10(5)	30(5)	9(3)	7200
12	10(9)	8(7)	11(4)	8(8)	10800

Table 4. The measured activities in different samples of vegetation

Sample	Activity, [Bq] (t=100 s) ^{137}Cs	^{134}Cs	Activity, [Bq] Ge(Li) ^{137}Cs	^{134}Cs	t [s]
1	3500(190)	1140(60)	2800(190)	900(190)	100
2	3300(190)	1200(60)	3000(170)	940(60)	300
3	2700(150)	1000(50)	2700(180)	760(70)	100
4	2000(100)	760(40)	2000(130)	600(60)	100
5	1360(80)	470(30)	1030(80)	310(40)	100
6	720(40)	260(15)	590(30)	190(20)	300
7	640(40)	250(15)	590(35)	180(30)	3600
8	420 (20)	130(10)	320(20)	100(10)	600
9	350(20)	110(6)	360(25)	110(10)	600

Conclusions

The coincidence method of radon (and radium) measurement has a good accuracy and a sensitivity better than some standard methods that are currently in use. Moreover, practical insensitivity of the spectrometer PRIPJAT to the shape of analysed sample offers an additional advantage, because the need for sample of a standardised geometry is avoided.

Also, the method of the determination of radionuclides activities using spectrometer ARGUS, based on the registration of the cascades intensity of γ-rays of different multiplicity, applied when the isotope structure of the sample is known,

enables and provides express measurement and sensibility high enough for this purpose, as well as the accuracy, meanwhile not asking for the amplitude analysis of the registered radiation.

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