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Measuring the Activity of Thorium by Its Decay Products Using a Multidetector γ-Ray Coincidence Spectrometer

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Abstract—A method for detecting thorium by its decay products has been tested on PRIPYAT six-crystal 4π γ-ray coincidence spectrometers. This method makes it possible to measure large-volume samples of any shape with a higher sensitivity and a shorter acquisition time than when an HPGe spectrometer with a comparable cost is used.

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INTRODUCTION

Since thorium is present in small quantities virtually everywhere and its total reserves are three to four times greater than the reserves of uranium in the Earth's crust, ^{232}Th activity measurements are of considerable ecological interest.

The radioactive series of ^{232}Th, which contains ten daughter radionuclides, undergoes a decay chain (seven α and five β decays accompanied by emission of γ rays) and ends with a stable isotope of lead—^{208}Pb [1]. If ^{232}Th is in radioactive equilibrium with its decay products, its activity is usually determined from a spectrum measured using a semiconductor γspectrometer by the most intense peak of γ rays from ^{228}Th (accompanying β^- decay of ^{228}Ac) with an energy of 911.1 keV (0.258) [2] or by a few peaks, e.g., with energies E_γ = 338.4 (0.113) and 911.1 keV (0.258) [3]. This is dictated by the necessity to avoid errors due to possible ^{220}Rn emanation from the measured sample. Semiconductor γ spectrometry is the second-most-used technique after α spectrometry. The data acquisition time is ≥10000 s.

The aforesaid basically refers to the case where a single detector is used to detect radiation. If a few particles are simultaneously emitted during a nuclear decay and detected by several detectors, it is possible to employ techniques based on detection of coincidences. In most cases, these techniques allow the minimum detectable activity to be lowered by improving the signal-to-background ratio [4].

The detection efficiency for a cascade is proportional to the product of the detection efficiencies for single quanta. Therefore, when techniques based on coincidences are used to make measurements, individual detectors included in a multidetector spectrometer must have high detection efficiencies and the geometric efficiency of the spectrometer must be as close to 4π as possible.

It is this type of multidetector spectrometers among which γ-ray spectrometers of the PRIPYAT series are classified [5]. In the previous paper [6], it was shown that, after appropriate engineering and methodical upgrading, the PRIPYAT spectrometer can be used to good effect for determining the concentrations of the decay products of natural radionuclides and, in particular, radon isotopes by detecting coincidences of γ rays emitted by such decay products as ^{214}Bi and others. A new data acquisition system for the PRIPYAT-2P γ spectrometer and a method for measuring the activity of ^{232}Th and its decay products using this spectrometer are described in this paper.

THE PRIPYAT γ-RAY COINCIDENCE SPECTROMETER

A measuring chamber of the PRIPYAT six-crystal scintillation 4π spectrometer of multiple γ-ray coincidences is shaped as a cube with a side of 175 mm. A scintillation detector based on a NaI(Tl) crystal 150 mm in diameter and 100 mm in length and an ФЭУ-49Б (i.e., a БДЭГ-39 scintillation unit with improved amplitude and time responses [7]) or ФЭУ-173-1 photomultiplier tube is positioned in each face of the cube. The energy resolution of new detectors included in the spectrometer is 8–12% for 662-keV γ rays from ^{137}Cs. The sensitive volume of the spectrometer is sufficient to make measurements of samples with an arbitrary shape and a volume of up to 5 l without preliminary sample preparation. The total solid

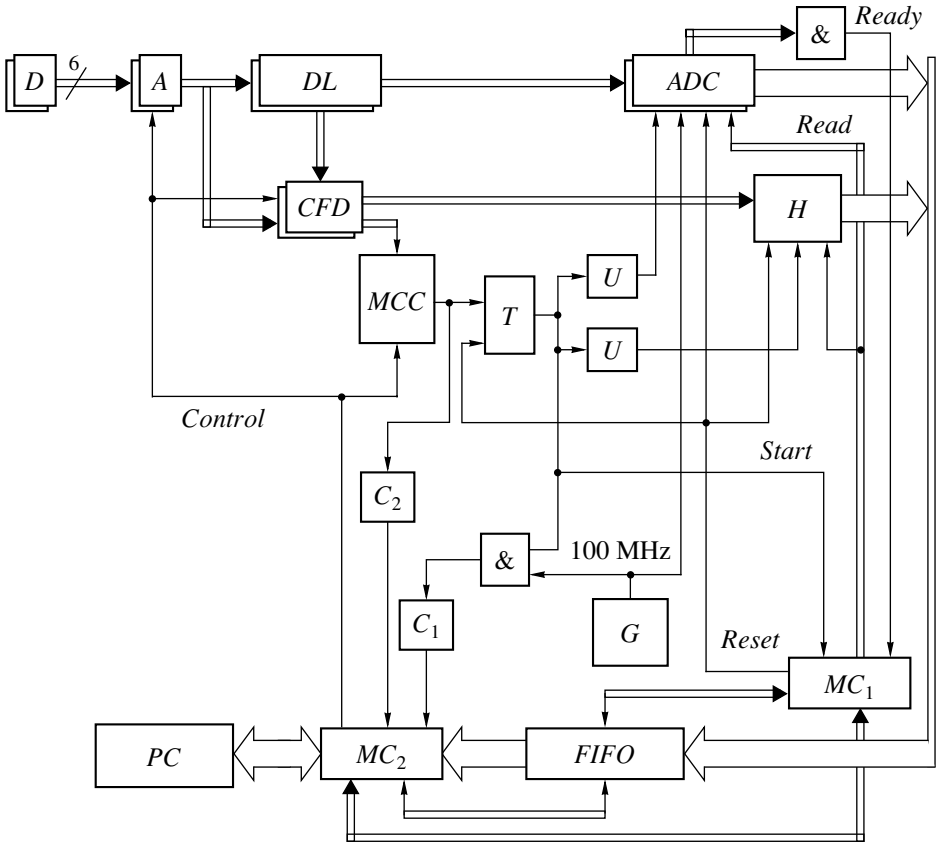

Fig. 1. General arrangement of the data acquisition system of the PRIPYAT-2P spectrometer: (*D*) detectors, (*A*) amplifiers, (*DL*) delay lines, (*ADC*) charge-to-digital converters, (*CFD*) constant fraction discriminators, (*MCC*) majority coincidence circuit, (*U*) univibrator, (*H*) hodoscope, (*T*) event flag trigger, (C_1) frequency divider of the dead time counter, (C_2) frequency divider of the event counter, (*G*) generator, (MC_1, MC_2) microcontrollers, (*FIFO*) buffer memory, and (*PC*) personal computer.

angle of the spectrometer is ~0.7 of the 4π. The photopeak efficiency of detecting 662-keV γ rays from ^{137}Cs is 0.25; for a point source, it varies over the sensitive volume of the spectrometer by 10% or less.

Until recently, the PRIPYAT spectrometer has been used mostly in express analyses of the ^{137}Cs and ^{40}K concentrations in a great number of variously shaped samples of different nature. This spectrometer has a very high γ-ray sensitivity and is capable of detecting cesium radionuclides in water at a level of 10 Bq/l over an acquisition time of ≤1 min.

A new data acquisition system and new measurement procedures have been recently developed for the spectrometer. The new data acquisition system (Fig. 1) has software-controlled (for each spectrometric channel) gains, detection thresholds, zero line shift, and a multiplicity of coincidences; as a result, the range of measurements can be varied over wide limits. In the default setting, the energy range of the spectrometer is 0.2–3.0 MeV.

Physically, the data acquisition system consists of six spectrometric channel units and one spectrometric channel control unit. The system can be nested into a computer and is powered by voltages of +12 and –12 V from the power supply unit of the computer. Control and data exchange between the control unit and the computer are exercised via the COM and LPT ports.

The operating principle of the data acquisition system is as follows. The current pulses arriving from the detectors are amplified by fast amplifier *A* with a software-controlled gain. The output pulse from the amplifier is analyzed by constant fraction discriminator *CFD* and charge-sensitive analog-to-digital converter *ADC*. The signal from the amplifier passes to the *ADC* through analog delay line *DL*. The *DL* has ten intermediate outputs, from one of which the signal is read out into the *CFD*. The *CFD* signals are fed into majority coincidence circuit *MCC*, which produced gate pulses for the *ADC* and the hodoscope that registers simultaneous operation of the detectors.

After the conversion is completed, all the converters produce the ready signal, and the control program of microcontroller MC_1 records data, which were registered in the hodoscope, from the *ADC* into buffer memory *FIFO*. The control program of microcontroller MC_2 transmits the data from the *FIFO* to the main computer program.

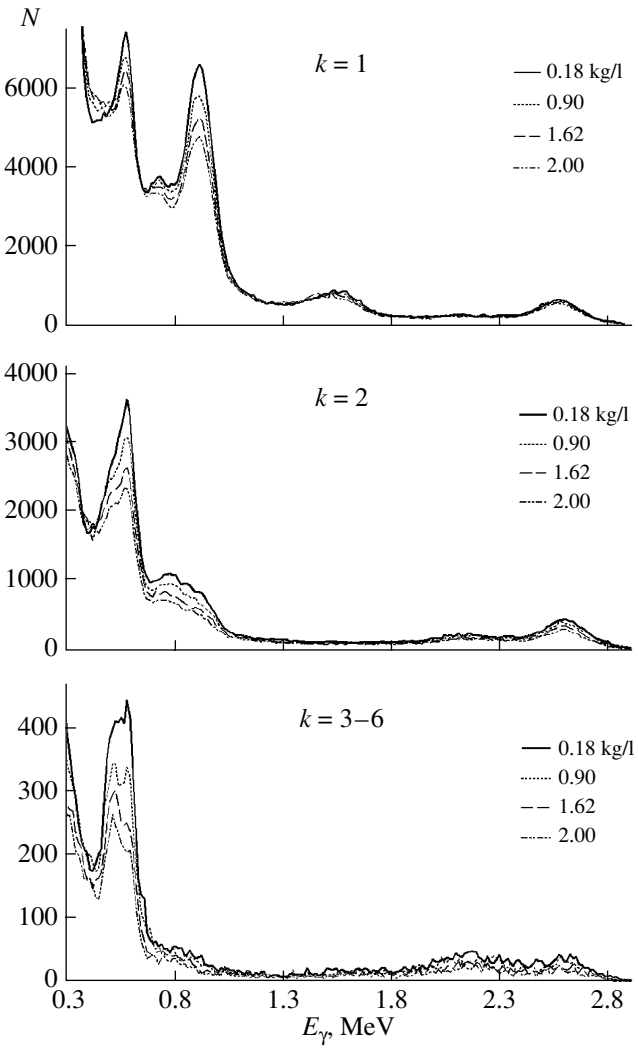

Fig. 2. Spectra of bulk reference samples with ^{232}Th (800 Bq) of different densities, measured by the PRIPYAT spectrometer in the modes of coincidences under investigation k (t = 1000 s).

If the detector signal contains short noise pulses, it is possible to suppress them in the recorded spectrum by discriminating the signal according to its duration. Discrimination is effected by a complementary register of the hodoscope, which detects the presence of a pulse at the input within a reference time interval. Should the pulse be absent, this event is rejected, and all converters *ADC* and the hodoscope are reset.

Information about each registered event arrives at the computer in the form of the quantity of activated detectors, their numbers, and the energies of γ rays responsible for the coincidence. The resolution time for coincidences is 40 ns, and the multiplicity of coincidence ranges from 1 to 6.

Under software control, information arriving from the detectors is presented in the form of sum (for each detector) one-dimensional integral spectra with multiplicities from 1 to 6. The mode selected for most practical applications (e.g., measurements of natural radionuclide concentrations in building materials) implies formation of three sum spectra with multiplicities of 1 and 2 and a window of multiplicity of 3–6.

The spectra obtained thereby can be processed in a spectrometric mode using the line spectrum processing procedures or (when the isotopic composition is known beforehand) in a radiometric mode using the matrix method and the data obtained by measuring standards of appropriate isotopes with different densities and masses.

DETECTION OF ^{232}Th BY ITS DECAY PRODUCTS

In the ^{232}Th series, coincident γ quanta appear in decays of ^{232}Th, ^{228}Ac, ^{228}Th, ^{224}Ra, ^{212}Bi, and ^{208}Tl nuclei [1]. However, the quantum yields of cascades have a significant value only for ^{208}Tl. Virtually all the γ rays from this isotope are emitted by cascade transitions with multiplicities of 2, 3, and 4; a 2615-keV quantum is emitted at the end of the cascade. Of the double cascades for ^{208}Tl, the strongest are 583 + 2615 keV (0.175) and 861 + 2615 keV (0.044); of the triple cascades, 511 + 583 + 2615 keV (0.078) and 277 + 583 + 2615 keV (0.022). The quantum yields of the cascades are presented in the brackets. ^{228}Th isotope produced by β decay of ^{228}Ac also has a complex structure of levels, deexcitation of which is followed by cascade transitions. However, the most intense double cascades end with transitions from a level of 58 keV, which lie below the detection threshold of the PRIPYAT spectrometer. The intensities of the other cascades are rather low.

In view of the aforesaid, one can see several good chances to determine the thorium activity in samples using the PRIPYAT spectrometer.

—Detection of 911-keV γ rays from ^{228}Th (β decay of ^{228}Ac) in a spectrum of single events. Taking into account the energy resolution of the PRIPYAT spectrometer, this spectrum peak is the unresolved multiple peak of γ rays with energies of 911 (0.258), 965 (0.05), and 969 (0.158) keV. The total quantum yield of these γ rays per decay of ^{232}Th atom is 0.47. A 860-keV (0.045) γ-ray peak of ^{208}Pb (β decay of ^{208}Tl) also overlaps this peak.

—Detection of 583-keV γ rays from ^{208}Pb (β decay of ^{208}Tl), primarily in the mode of double and triple coincidences with cascading γ rays. Quanta with energies of 2615, 511, 277, and 763 keV are emitted in cascade with this line. It should also be noted that 511- and 583-keV γ-ray peaks are virtually unresolved in double- and triple-coincidence modes (Fig. 2). Therefore, the total quantum yield for this line is 0.385.

—Detection of 2615-keV γ rays from ^{208}Pb (β decay of ^{208}Tl), primarily in the mode of double and triple coincidences with cascading γ rays (as in the previous

case). All γ rays are emitted in cascade with this line. The quantum yield for this line is 0.359.

Generally, it is convenient to use 239-keV γ rays from ^{212}Bi (β decay of ^{212}Pb) to detect thorium by means of the γ spectrometry method, since both the detection efficiency of the spectrometer at this energy and the quantum yield (0.47) are high. However, this peak lies at the boundary of the energy range detectable by the PRIPYAT spectrometer and is partially cut off. A slight drift of the threshold may cause instability of results.

Figure 2 presents the spectra of ^{232}Th reference samples with a volume of 1 l and an activity of 800 Bq, measured by the PRIPYAT spectrometer in coincidence modes with k = 1, 2, and 3–6. The reference samples were packed into hermetically sealed plastic containers with filler densities of 0.18, 0.90, 1.62, and 2.00 kg/l. The energy resolution of the spectrometer for 662-keV γ rays of ^{137}Cs was 15%. The background spectra obtained using this spectrometer in the corresponding coincidence modes are shown in Fig. 3.

The table contains thresholds of ^{232}Th detection (in terms of minimum detectable activity, A_{min}) by γ rays with energies of 583, 911, and 2615 keV, which were estimated according to the formula

$$A_{min} = \frac{m\sqrt{N_b}}{\varepsilon\sqrt{t}}, \quad (1)$$

where ε is the photopeak detection efficiency, N_b is the background counting rate in the photopeak region, t is the data acquisition time for the analyzed sample, and m = 2 is the number of rms deviations due to statistical background fluctuations (the confidence level P = 0.95). A sand-containing reference sample with a density of 1.62 kg/l, a volume of 2 l, and activity A = 1400 Bq was used to determine the detection efficiency of the spectrometer for the above-mentioned energies.

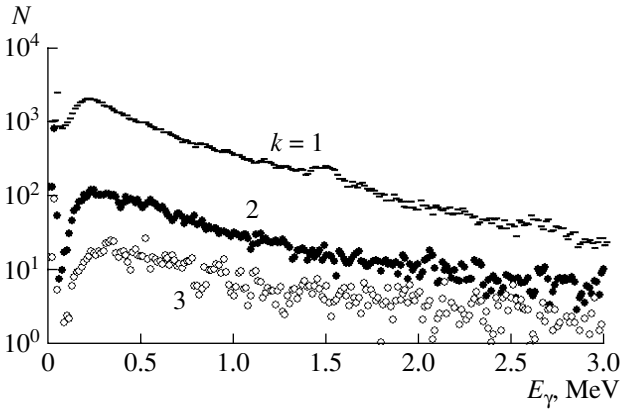

Fig. 3. Background spectrum measured by the PRIPYAT spectrometer in the modes of coincidences under investigation k (t = 4968 s).

DISCUSSION OF THE RESULTS

The indubitable advantage of the method for determining the ^{232}Th activity by 911-keV γ rays consists in a weak dependence of results on thoron (^{220}Rn) emanation from the sample, since most of overlapping γ-ray peaks, except for that with an energy of 860 keV (0.04), belong to ^{228}Th, the decay of which precedes the ^{220}Rn decay in the thorium series.

As regards the determination of the ^{232}Th activity by a 583-keV γ-ray peak, the double coincidence mode is the optimum variant (see the table). In the mode of k = 2, the background counting rate in the photopeak region decreases by a factor of 14 in comparison with the

Detection efficiency ε and minimum detectable activity A_{min} of ^{232}Th in different measurement modes k at confidence probability P = 0.95

E_γ, keV	k	N_b, s^{-1}	Spectrometric mode (t = 1000 s)		Radiometric mode (t = 100 s)	
			$\varepsilon \times 10^2$	A_{min}, Bq/kg	$\varepsilon \times 10^2$	A_{min}, Bq/kg
583*	1	20.88	1.33	6.7	15.06	1.9
	2	1.45	1.54	1.5	6.27	1.2
	3	0.25	0.30	3.3	0.63	4.9
911**	1	10.21	4.06	1.5	13.30	1.5
2615	1	0.94	0.81	2.3	2.13	2.8
	2	0.14	0.35	2.1	0.95	2.4
	3	0.06	0.04	17.4	0.07	20.9

*Overlapping of γ-ray peaks with energies of 511 (0.081) and 583 (0.304) keV, which cannot be resolved by the PRIPYAT spectrometer.

**Overlapping of γ-ray peaks with energies of 860 (0.045), 911 (0.258), 965 (0.05), and 969 (0.158) keV, which cannot be resolved by the PRIPYAT spectrometer.

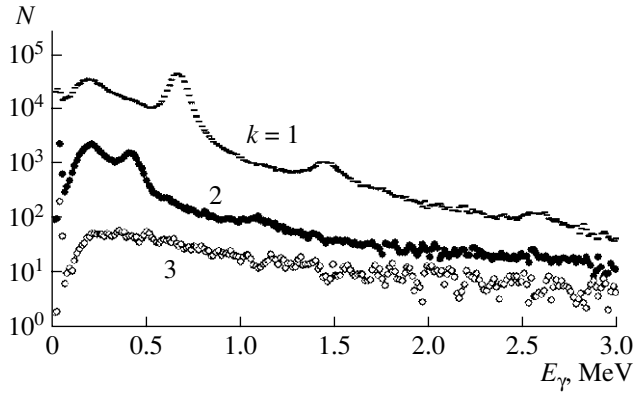

Fig. 4. Spectrum of the sample containing ^{137}Cs (203.4 Bq) in different coincidence modes k (t = 2000 s).

mode of k = 1, while the experimental detection efficiency for this photopeak increases only slightly. In this case, the ratios of the experimental detection efficiency for the 583-keV photopeak and the background counting rate in the double- and triple-coincidence modes are virtually equivalent, which causes A_{min} to increase according to Eq. (1).

For 2615-keV γ rays, the double coincidence mode is the optimum mode for determining the thorium activity (see the table). Although the total intensity of cascades in the mode of k = 2 is virtually equivalent to the total activity of the cascades with participation of 583-keV γ rays, the detection efficiency of the spectrometer's detectors in this energy range is slightly lower; therefore, the threshold of thorium detection using this line is higher. It should nevertheless be noted that the advantage of a 2615-keV line consists in that this energy range does not contain strong γ-ray lines due to decays of the other natural-occurring radionuclides (^{40}K and ^{238}U) and ^{137}Cs, which can affect the results of thorium measurements.

In what follows, for the sake of comparison, we present the results of thorium measurements by means of an ORTEC-30185-S coaxial HPGe spectrometer with a relative detection efficiency of 35%. The dependence of the spectrometer's detection efficiency on the γ-ray energy was determined using a multistandard in a Marinelli beaker with a capacity of 0.5 l and a density of 1 kg/l. In this case, the detection efficiency for the 911-keV photopeak used to determine the thorium activity was 0.013. A soil sample in a Marinelli beaker (0.5 l) with a density of 1.2 kg/l and an activity of 940.5 ± 30.5 Bq was used to determine minimum detectable specific activity A_{min}. In this case, the A_{min} value was calculated following the standard 3MDA method (program [8]) according to the formula

$$A_{min} = 4.5\frac{1+\sqrt{1+0.888N}}{T_j\varepsilon I}, \qquad (2)$$

where T_j is the live time, N are the background counts under the photopeak, ε is the detection efficiency, and I is the quantum yield. Over measurement time t = 11 840 s, minimum detectable specific activity A_{min} for this spectrometer was 2.1 Bq/kg.

Thus, comparison shows that detection of thorium by the PRIPYAT spectrometer takes much lesser (by an order of magnitude) time as compared to an HPGe spectrometer.

From the table, it is also apparent that virtually the same A_{min} values detectable in the spectrometric mode can be obtained in the radiometric mode over t = 100 s, which is an order of magnitude faster. In the limiting case when the thorium activity was determined by means of this spectrometer in the radiometric mode over the whole energy range (thorium only was present in the sample), the A_{min} value was 1 Bq/kg in both single- and double-coincidence modes over measurement time t = 30 s.

Since a reference sample of a 2-l volume was used to determine the experimental detection efficiencies of the spectrometer for the above-mentioned lines, it seems that the thorium detection threshold can be decreased almost twofold by employing 5-l samples.

The described technique provides a means for estimating the minimum activity of ^{232}Th and its decay products (^{220}Rn in particular) detectable by the PRIPYAT spectrometer in various samples with established radioactive equilibrium by measuring the total multiple integral spectra and can be used to determine the ^{232}Th activity (and specific activity).

The coincidence method allows the activity of ^{232}Th and its decay products to be measured in samples contaminated by ^{137}Cs without deterioration of the sensitivity. The use of this method has an obvious advantage in zones with radioactive contamination, since it helps exclude the effect of contamination by ^{137}Cs radionuclide, which is a one-cascade γ-ray emitter (662 keV) and makes a minor contribution to the coincidence spectra in the region of interest. As our measurements showed, the 662-keV peak, which does not virtually contribute to double-coincidence spectra, and the 583-keV peak are poorly resolved in spectra with k = 1 (Fig. 4).

The advantage of the method for measuring the activity of ^{232}Th and its decay products using PRIPYAT six-crystal 4π spectrometers of γ-ray coincidences is primarily associated with the high sensitivity of the double-coincidence method and with the possibility of measuring samples of any shape—in particular, large-volume samples (up to 5 l) without additional time and effort required for the sample preparation and calibration measurements for different configurations of samples.

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