SPECTROMETER FOR DETAILED STUDY OF RADON AND THORON SYSTEM DECAY PRODUCTS

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The results of development of an aspiration type spectrometer for investigation of daughter products of radon and thoron are presented. Main parameters of the device: active detector area of 32 cm^2, air pumping through the filter up to 100 liters/min, energy resolution of the detection unit of 50…70 keV. Measurements are made both during the pumping and after the pumping. Accumulation, processing, representation and storage of data are made using a personal computer incorporated into the device. Setup and control of the device are made on the computer keyboard. The results of measurements of spectra of the radon and thoron system, including dependencies of α-activity of various radio-nuclides are shown. With this device it's also possible to measure electron energy loss spectra.

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1. INTRODUCTION

Radon, thoron and their decay products present the greatest danger to the health of population among the known sources of radiation exposure. The relationship between 9 most radioactive decay products of radon and thoron in the premises is very complex. This complexity is caused by the significant number of radiating nuclides, by their comparable contribution to the common activity of system, by their short half-life periods $T_{1/2}$ (seconds, minutes), leading to rapid quantitative changes in composition of system, by dependence of quantity and composition of radio-nuclides on very large number of geological, climatic and other factors and also on the conditions of building and state of premises [1]. Therefore the determination of the concentration of all components of this system represents a complex task. At present this problem is not yet sufficiently solved. The instruments commonly used for measuring the volumetric activity of air in the premises, in the water or in the soil make it possible to conduct only the simplest estimation of radon danger and lack the possibility of detailed investigation the of radio-nuclides composition. At the same time these data are necessary for the correct radon monitoring, precise estimation of risk and, that is most important, for the correct choice among the numerous methods of decreasing the radon danger [2]. The spectrometric methods are most effective in determining these relationships [3]. The decay chains of radon and thoron are represented in Fig. 1.

Here we present more full data including information about β- and γ-radiation of radon and thoron decay products. Once again we note, that radon (^{222}Rn) and thoron (^{220}Tn) itself will not be considered further, as they both are not caught by the filter and yield no signals. But their α-radiating decay products have rather favorable distribution of α-radiation energy, permitting their confident separation. The exception makes only ^{212}Bi from a chain of thoron, which yields α-radiation practically coinciding by energy with that of ^{218}Po. However, it is necessary to take into account, that, firstly, emission intensity in a thoron chain is usually significantly lesser than that in a chain of radon. Secondly, α-decays make only third of all decays ^{212}Bi. Thirdly, the peak of ^{212}Po because of its very short half-life time $T_{1/2}$ completely follows the decay of ^{212}Bi. So it is enough to make small correction of intensity of ^{218}Po peaks to solve this problem completely. The similar situation exists and in a chain of radon: the decay of ^{214}Bi controls ^{214}Po decay due to short $T_{1/2}$ of a polonium. Therefore it is possible to consider the speed of ^{214}Po decay, which is measured on α-peak with energy 7.69 MeV, as that of ^{214}Bi.

There is a problem of experimental definition of ^{214}Pb decay speed. For this purpose β-radiation of this radio-nuclide can be used. It is possible not to take into account the radiation with energy threshold of 1.03 MeV because of its small intensity. It is necessary to note, that the definition of ^{214}Pb decay speed represents a nontrivial problem, as there is a commensurable contribution of ^{214}Bi radiation. The subject is more complicated with a thin silicon detector, where not the initial spectra of β-particles, but the spectra of their energy losses are measured. We take into account that the spectra of energy losses are measured as well as the spectra of energy losses for the energy thresholds

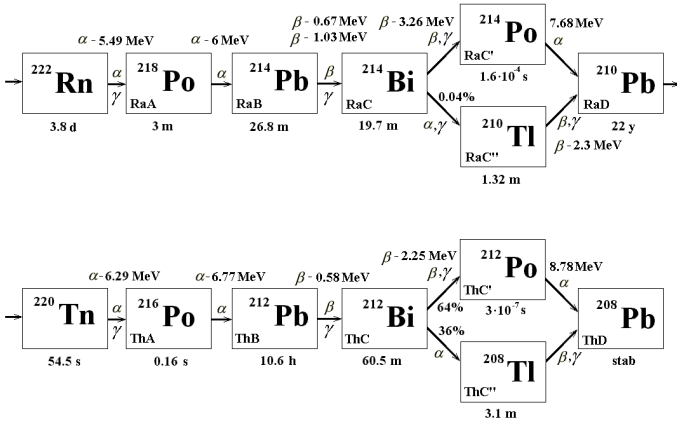

Fig. 1. *Decay schemes of radon and thoron*

thresholds of 0.67 MeV and of 3.2 MeV are different. Let's mark the paradoxical, at the first glance, fact – the signals of radiation with the greater thresholds energies will have a weight center at smaller energy, than signals with smaller thresholds energies. The method of separation of ^{214}Pb signals from that of ^{214}Bi is following: the spectral functions of response from ^{214}Pb and ^{214}Bi radiation sources are previously measured. For these spectra the ratio of numbers of signals of β-particles below and above the level 0.7 MeV are determined. Further, using this ratio, from an actual radon spectrum, measured with the filter, it is possible to determine ^{214}Pb and ^{214}Bi decay speeds separately. Certainly, this method of separation of radio-nuclides contributions is indirect and inaccurate.

2. DETECTION OF α- AND β-PARTICLES

For detection of α- and β-particles we have developed the instrument, in which radioactive aerosols precipitate on the filter and measurements of α- spectra and spectra of β-particles energy losses are made with its use. The possibilities of this instrument are extended in comparison with known instruments also due to the fact that the high speeds of air circulation are used (up to 100 liters per min) and measurements are performed not only after air circulation, but also during it. Need for recording a significant number of signals together with providing high enough energy resolution is a very complicated problem. The increased air pumping speed and the large area of filter and detector are used for this. In this work the silicon diode with area of 32 cm^2 is used as a detector. However, increase of detector active area is connected with significant difficulties. These difficulties concern all types of detectors, but with respect to silicon detectors they are manifested especially clearly. With an increase in the area of silicon detector its leakage current and noise also grow. Furthermore, the increase of electrical capacitance of detector also leads to increase of noise of charge-sensitive preamplifier. An increase of noise results in degrading of energy resolution and increase in the minimum energy of the detected particles. Even with high quality of technology, noise of 32 cm^2 detector together with that of preamplifier is about 100…130 keV at 20°C. At 40°C it is 250…300 keV. Concerning spreading of α-peaks, it is necessary to add the dispersion of energy losses in the filter and in the air. Final energy resolution can be more than 400 keV, which would complicate the separation of α-peaks. So the task of a substantial improvement of energy resolution arises. Poor resolution not only narrows the possibilities of α-spectra analysis, but also makes it impossible to record β-particles. Because of small β-particles signal amplitudes in silicon detector the channel of β-particles registration suffers from noise pulses, which imitate β-particles. It is possible to cut off these noise pulses by establishing sufficiently high threshold level in the signal discriminator, but in this case the substantial part of the signals of high energy β-particles would also be lost, which is unacceptable. The progress is possible in case of use of multi-sectional detection block [4], proposed by the authors of this work. The idea is the following: detector is divided into n independent parts (sections) with an area of each section of *s*. Each section has its own channel of preamplifier and shaper. Signals from all channels are then fed to special mixer, in which they are directed to one bus and further they are passed to the counter or the pulse-height analyzer. Mixer is designed in such a way that it passes noise only from that channel, which at the given moment is the source of signal, exceeding some threshold level. Thus, we obtain detector with an effective area of *ns*, while the noise at the mixer output corresponds to that with area *s* of a single section.

In this work the integrated matrix of 6x6 silicon diodes is used as the multi-sectional detector, developed and produced by us. Area of the matrix element is 1 cm^2. With the bias of 10 V the leakage current of the matrix element comprises less than 1 nA, and electrical noise is about 5 keV, which corresponds to the best models of leading producers. The spectra of noise given in Fig. 2 were measured with the same detector with an area of 32 cm^2 in 4 modes: single-channel (when all elements are connected to one channel of amplification), 2-channel (2 channels each of 16 cm^2), 4-channel (4 channels each of 8 cm^2) and 8-channel (8 channels each of 4 cm^2). The results of calculations of noise threshold energy E_{thr} from the data of spectra are brought to Table 1.

Table 1. *Noise for the diverse variants of multi-section detector*

Number of channel	E_{thr} (keV)	Ratio of E_{thr}(1 ch) /E_{thr}(n ch)	
		theory	experiment
1 channel	90.6	1	1
2 channels	62.2	1.4 … 2	1.46
4 channels	35.0	2 … 4	2.6
8 channels	25.3	2.8…8	3.6

First of all let us note that E_{thr}, which corresponds to the frequency of noise pulses 1 p/min, for the usual single-channel detector is too close to the energy losses of minimum ionizing particles (mip). This value is about 100 keV for the detector with the thickness of 300 μm. Consequently with this standard for the acceptable noise frequency a similar detector cannot be used for registering weak flows of mip. With the increase in the number of channels E_{thr} sharply decreases and in the 8-channel detector it is about 25 keV, which makes it possible to confidently record very low flows of mip in the wide temperature interval and to conduct α-spectra measurement with the sufficiently high energy resolution. The decrease of E_{thr} in the multi-sectional detector occurs with the factor of $\sqrt{n}$ for noise of detector and with the factor of n for noise of preamplifier. Since both components of noise are present, experimental results must give intermediate values that are practically observed (see Table 1). A similar effect with use of multi-sectional detector is also observed with respect to energy resolution. The values of energy resolution for the diverse variants are given in Table 2.

Table 2. *Energy resolution for the single-element and multi-sectional detectors*

Area of detector	Type of detectors	Resolution (keV)
16 cm^2	1 ch × 16 cm^2	70
	8 ch × 2 cm^2	30….40
32 cm^2	1 ch × 32 cm^2	150
	8 ch × 4 cm^2	40….50

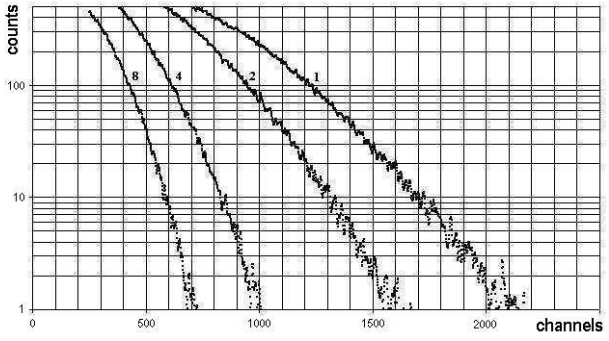

Fig. 2. *Noise spectra of detector with S=32 cm^2 with different number of sections. The weight of one channel is 0.03 keV/chan*

Cardinal improvement in the energy resolution because of the multi-sectional detector is evident. Thus, the principle of multi-sectional detection makes it possible to considerably increase the active area of detector practically without an increase in electrical noise. This makes it possible not only to drastically improve quality of α-spectra, but also offers the possibility of registering the electrons with silicon detectors with an area of tens and hundreds of sm^2. Previously for this purpose detectors with the area up to 5 cm^2 could only be used. This possibility can be used, besides of this work, for creation of new equipment for ecological monitoring, systems of radiation safety and nuclear physics.

3. INSTRUMENT FOR SPECTROMETRY OF THE RADIOACTIVE AEROSOLS

Instrument consists of two blocks and notebook type personal computer. In the first block the air pump and electronic circuits for pump control are placed. The second (measuring) unit houses the air filter, silicon detector, boards of multi-channel preamplifiers and shapers, mixer, spectrometer of signal pulses, power source. Control of the instrument's work, spectra processing and displaying results of measurements are done with the aid of PC. For the air pumping through the filter the MPR-100 type micro-pump is used, providing necessary air flow speed. The system ensures high flow stability and its weak dependence on aerodynamic load at the air input. The weight of instrument without PC is about 5 kg.

The possibility of conducting measurements during the air pumping required air gap creation between the filter and the detector, necessary for air access to the filter. In this case the significant losses of α-particles energy in the clearance appear. The dispersion of these losses because of the difference in the α-particles angles of incidence can give the prevailing contribution into the energy resolution worsening. Avoiding this effect is possible with the use of collimator, which limits the angles of incidence. Collimator design influences the detector effectiveness and in a very complicated way its energy resolution. Therefore the collimator construction should be optimized for particular requirements of the instrument. Entire detector block is designed to ensure rapid replacement of collimators. In certain cases there is need for pumping air through the filter in the remote air pump block, and then to transfer it into the measuring unit and to measure spectrum placing the filter closely to the detector. This possibility is also implemented in the instrument.

Experiments were carried out with the collimator, which has a comparatively long distance (14 mm) between the filter and the detector (lattice type collimator of 14 mm height with 3.3·3.3 mm^2 square openings). The effectiveness of detection is 40%. With the use of 8-channel construction of detector and amplifiers maximum energy of noise pulses with frequency of 1 p/min is about 25…30 keV, energy resolution for α-particles is 120…130 keV. The dispersion of energy losses in the air gives main contribution to the energy resolution.

Control of the instrument work and measurement results processing is produced using digital module of data acquisition, processing and accumulation (DAPA). DAPA is based on the micro-controller. It has 12-bit ADC, 32k RAM, RS232 interface for the connection

with the computer through the COM-port, logic for information readout control. DAPA can also be connected to PC using parallel interface (LPT-, EPP- and ECP-port). ADC has 256…4096 output code levels (programmable), conversion time of 5 μs and signal/noise ratio of 70 dB.

Micro-controller has an instruction set, compatible with 8051 industrial standard and can be in-circuit reprogrammed. MC crystal has 8-kbyte Flash/EE program memory, 640 bytes Flash/EE data memory and 256 bytes RAM. Furthermore, MK has a watch-dog timer, two 16-bite timers, power supply monitor and 32 programmed bi-directional logical in/out pins. SPI, I^2C and standard UART channels can be used as the interface. DAPA block has the mezzanine construction, which is widely used in the last developments of the leading firms. Major portion of the block is located on the mother board, and several input devices (discriminator, peak detector and ADC) is on separate boards, which construction and functions can easily be changed. Thus the versatility of device with minimum of redundancy is reached.

The instrument control program provides its tuning, adjusting and setting of operating modes from the computer keyboard. The program of spectra processing makes it possible to carry out operations of convolution, smoothing, automatic peak search and determination of peak parameters, together with number of other operations. The partition of spectrum into energy ranges and plotting the integral dependences $N_n = f(t)$, where N_n is the current number of signals in n-th energy range, is possible. Then these dependences can be approximated by smooth curves, and the time dependences of each separate α-radiating radio-nuclide activity on the filter can be obtained.

The experiments with the spectrometric filters AFA-RSP-20 are carried out using this instrument. The spectra obtained instantly during 30 min of air circulation and during 30 min after circulation are shown in Fig. 3a and Fig. 3b, respectively. In spectra obtained instantly during air circulation the peak of significant intensity with the energy of ~6 MeV is present, which corresponds to ^{218}Po as the immediate decay product of radon (^{222}Rn). As a result of short half-life time (3 min) its intensity after the end of circulation sharply falls. The kinetics of this and other peaks for one of the working premises on 2nd floor is given in Table 3.

The most intensive peak has energy of 7.7 MeV. This is ^{214}Po from the chain of radon. The half-life time of ^{214}Po is short (~10^{-4} s), but its kinetics is determined by the predecessor radio-nuclides: ^{214}Pb (26.8 min) and ^{214}Bi (19.7 min). Therefore changes in its intensity have some inertia: the intensity grows after the end of propulsion and then starts falling after tens of minutes. Sometimes one additional peak with the energy 8.8 MeV is manifested, which corresponds to ^{212}Po from the decay chain of thoron (^{220}Rn). This is weak and very inertial peak. The half-life time of ^{212}Po is short (~10^{-7} s), but the periods of the half-life of its predecessors are long: ^{212}Pb – 10.6 hours and ^{212}Bi – 60 min. Thus, all expected peaks are observed in the chains of radon and thoron. As it was expected, peak of ^{216}Po (6.8 MeV) is not observed as a result of its very rapid decay (0.16 s). ^{216}Po manages to decay earlier than it is captured by

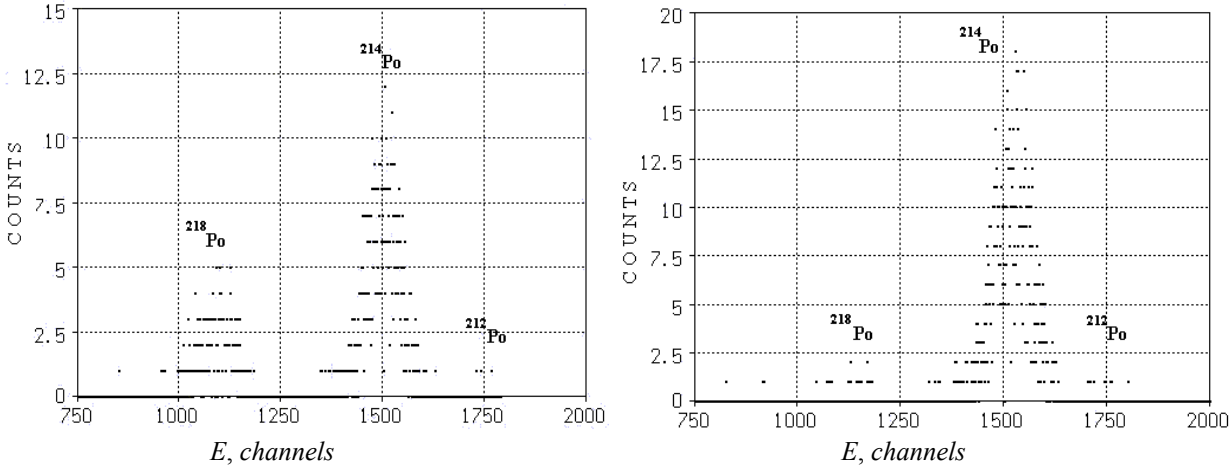

Fig. 3. *α-spectra of radon and thoron decay products. (a) – during 30 min expulsion of air, (b) – after expulsion, write time is 30 min*

Table 3. *Peak intensities (number of pulses) during 3 sequential periods of the measurements: 1 - air propulsion during 30 min, 2 - after propulsion, 30 min, 3 - next period after propulsion, 30 min*

Radio-nuclide	Chain	Period 1	Period 2	Period 3
^{218}Po	Radon	280	32	3
^{214}Po	Radon	778	1223	554
^{212}Po	Thoron	3	10	12

filter. The peaks of the maternal radio-nuclides of radon (^{222}Rn) and thoron (^{220}Rn), critical for the penetration into the premise, are absent because they are not captured by filter.

Coming from the first results it is already evident that the spectra make it possible to considerably clear up the picture of radio-nuclides distribution and their kinetics. As an example of possibilities of spectral studies let us note that it is possible to separate accurately the role of thoron chain, although its contribution to the general radioactivity can be very small. Moreover, long exposures of filters are not necessary.

4. CONCLUSIONS

An aspiration type spectrometer developed for detailed study of radon and thoron contamination in air has some advantages in comparison with known devices. Due to the high speed of air circulation through the filter the measurements of the activity of radon and toron decay products can be performed not only after air circulation but also during it. The principle of multi-sectional detection is proposed and realized in this instrument with the aim to provide the high efficiency of registration and high-energy resolution simultaneously. Due to this the quality of measured α-spectra can be essentially improved.

Designed spectrometer and multi-sectional detectors with large active area can be useful for measurements of α- and β-radiations at ecological monitoring as well as in the field of nuclear physics and radiation safety.

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СПЕКТРОМЕТР ДЛЯ ДЕТАЛЬНОГО ИССЛЕДОВАНИЯ ПРОДУКТОВ РАСПАДА РАДОНА И ТОРОНА

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Представлены результаты разработки спектрометра аспирационного типа для исследования дочерних продуктов распада радона и торона. Основные параметры прибора: активная площадь детектора 32 см^2, скорость продувки воздуха через фильтр – до 100 л/мин, энергетическое разрешение блока детектирования 50…70 кэВ. Измерения проводятся как после продувки воздуха через фильтр, так и во время продувки. В состав прибора входит персональный компьютер, с помощью которого производится набор, обработка, представление и хранение данных. Настройки и управление измерениями осуществляются с клавиатуры компьютера. Приведены результаты измерений спектров системы радона и торона, в том числе кинетические зависимости α-активности различных радионуклидов. Прибор дает возможность измерять также спектры энергетических потерь электронов.

СПЕКТРОМЕТР ДЛЯ ДЕТАЛЬНОГО ДОСЛІДЖЕННЯ ПРОДУКТІВ РОЗПАДУ РАДОНУ І ТОРОНУ

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Представлено результати розробки спектрометра аспіраційного типу для дослідження дочірніх продуктів розпаду радону і торону. Основні параметри приладу: активна площа детектора 32 см^2, швидкість продувки повітря через фільтр – до 100 л/хв, енергетична роздільна здатність блоку детектування 50… 70 кеВ. Виміри проводяться як після продувки повітря через фільтр, так і під час продувки. До складу приладу входить персональний комп'ютер, за допомогою якого виконуються набір, обробка, представлення і збереження даних. Настроювання і керування вимірами здійснюються з клавіатури комп'ютера. Приведено результати вимірів спектрів системи радону і торону, у тому числі кінетичні залежності α-активності різних радіонуклідів. Прилад дає можливість вимірювати також спектри енергетичних втрат електронів.