DEVELOPMENT OF NUCLEAR RADIATION MONITORS FOR RADIATION EARLY WARNING SYSTEMS

by

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Scientific paper
https://doi.org/10.2298/NTRP2203193G

The results of the development of modern precision monitors of alpha, beta and gamma ray radiation for setting up early warning systems for radioactive contamination in the atmosphere and rapid assessment of emerging threats, are presented. Proportional counters, scintillation $SrI_2(Eu)$ crystals and semiconductor Si, CdZnTe, and HPGe detectors are used for the development. The designed monitors provide information both on dose rate values in real time and on the activity of specific radionuclides. The software controls the measurement mode, as well as diagnoses the condition of the monitors themselves.

Key words: radiation early warning system, radioactive aerosol, environment monitoring, water radiation monitoring, ambient dose equivalent

INTRODUCTION

During the testing of nuclear weapons (in Nevada, Novaya Zemlya, Mururoa, Semipalatinsk), or as a result of man-made disasters at nuclear power facilities (Chernobyl, Fukushima) or in the extraction of uranium and other minerals containing radioactive elements, significant amounts of radioactive elements can be released into the Earth's atmosphere. Radioactive elements combining in the atmosphere with various gases in aerosol compounds, can rotate in the atmosphere around the planet for a long time, falling arbitrarily on the Earth's surface, depending on atmospheric conditions.

This circumstance makes it extremely difficult to estimate the dose received by the population as a result of radiation incidents, especially if the place of radioactive fallout is located thousands of kilometers away from the epicenter. The tasks of determining the doses received by the population not only in the first hours after the accident but also for a long time after it or at a great distance from the site of the radiation disaster, are solved within the framework of environmental monitoring of the territory where the situation is monitored. Toa implement this type of monitoring and timely organization of measures to protect the population from ionizing radiation, systems called "Radiation accidents early warning systems" are used [1, 2]. Such systems are a network of ionizing radiation monitors, continuously transmitting information about the level of background radiation, the volumetric activity of radionuclides, meteorological information, and other measured parameters to the central control post [3-5].

The first and main task of radiation early warning systems (REWS) is to record the fact of exceeding the natural levels of ionizing radiation characteristic of a given area due to a passing radioactive cloud. The second task is to identify all the radionuclides contained in the cloud in order to determine their activity for the subsequent assessment of the dose received by people. Since in monitoring of a human the main measured value is the dose of external and internal irradiation, it is necessary to determine the list of all radionuclides that have the greatest impact on the formation of the total dose and include monitors and units for detecting all relevant types of ionizing radiation, into the REWS.

A joint project of European countries on the problems of radiation accidents early warning systems located in Europe was fielded to develop and implement a new generation of detector systems capable of providing information both on dose rate values in real-time and on specific nuclides [6]. It was proposed to use gamma dose rate meters (GDRM) on the basis of new scintillation crystals with improved spectrometric characteristics, as well as semiconductor CdZnTe detectors applying a simple method of obtaining a dose rate from pulse amplitude spectra, without the use of labor-intensive spectral deconvolution methods.

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Technical requirements for GDRM were also formulated within the framework of this project, on the basis of the IEC EN 60846 Standard, but taking into account the peculiarities of their use in REWS [7].

This paper presents the results of the development of modern precision nuclear radiation monitors complex with improved performances for the creation of the atmosphere radioactive contamination REWS and rapid assessment of emerging threats. This complex includes: (a) GDRM on the basis of Geiger-Muller counters, semiconductor CdZnTe and scintillation SrI_2(Eu) detectors; (b) a stationary automated aerosol monitor for the detection of alpha-, beta and gamma-emitting radionuclides; (c) an automated gas monitor for the detection of radioisotopes of iodine; (d) a mobile automated aerosol monitor for the detection of alpha- and beta-emitting radionuclides; (e) monitor of radioactive contamination of the aquatic environment. The developed monitors are the basis for the creation of modern REWS.

PERFORMANCE OF GAMMA DOSE RATE METERS

As an overview of the means and methods of radiation monitoring of the stations included in the European radiological data exchange platform (EURDEP) shows [8], most of the stations are mainly equipped with GDRM on the basis of Geiger-Müller counters and scintillation crystals (primarily NaI(Tl)). Geiger- Muller counters, although lowly sensitive and unable to determine the type of radionuclides present [9], are capable of operating in radiation fields with an upper dose rate measurement range of up to 10 Svh^{-1} [10] that scintillation GDRM cannot provide. Since, in addition, Geiger-Muller meters are relatively inexpensive, simple, and reliable in operation, the need for their further use in REWS is beyond doubt.

Development of GDRM based on scintillators with an improved resolution instead of NaI(Tl) crystals for environmental monitoring has already begun [11-14]. So far, mainly $LaBr_3$(Ce) and $CeBr_3$ crystal-based devices are commercially available [15, 16]. In accordance with the recommendations of the EURAMET project [6, 7], for application in our REWS, we used the SrI_2(Eu) scintillator, the characteristics of which have been progressing recently [11]. In addition, GDRM on the basis of Geiger-Muller counters and semiconductor CdZnTe detectors have also been developed. Photos of the developed GDRMs are presented in fig. 1, and their characteristics are in tab. 1.

GDRM-GM/2 meter (a) developed on the basis of a two-element Dose Rate Probe 70 091 (OEM Kit) [17]. The meter contains large (L) and small (S) proportional counter with dimensions (Ø26 167) mm and (Ø9 58.5) mm, respectively, as well as a microcontroller board, containing all elements which are required to operate a meter for measuring the ambient equivalent dose rate. GDRM-GM/2 meter provides registration of gamma radiation dose rate in the range of 0.15 μSvh^{-1}-5 Svh^{-1} and can be used in environmental monitoring equipment only as a dosimeter.

GDRM-CdZnTe/6000 meter (b) has been developed on the basis of clustered high-efficiency gamma-radiation spectrometer MultiSPEC6000 [18]. It is an assembly of serial micro spectrometers μSPEC1500, containing signal preamplifiers and multichannel analyzers in each channel, combined by a special holder. All micro spectrometers work in parallel, creating one common spectrum. With a total detector volume of 6400 mm^3, its energy resolution at 662 keV does not exceed 3 %. With such resolution, this monitor could indeed be used in REWS both as a GDRM in determining the dose rate of external radiation, and as a spectrometer in the measuring channels of equipment for identifying radionuclides and determining their activity, as recommended in the IAEA Source and Environmental Monitoring Program [5].

However, in terms of gamma radiation registration sensitivity, CdZnTe detectors are significantly inferior to scintillation crystals [19]. This can also be seen from the comparison of the characteristics of our developed meters (tab. 1). Registration sensitivity for GDRM-SrI_2/1.5 is 327 cps*/(μSvh^{-1}) for the 580-700 keV range and 683 cps/(μSvh^{-1}) for the 20-700 keV range. However, for the GDRM-CdZnTe/6000, these parameters are only 140 cps/(μSvh^{-1}) and 570 cps/(μSvh^{-1}) for the respective ranges – and this is virtually for the largest possible size 6400 mm^3 of a CdZnTe detector.

In the works [20, 21], where the application of the CdZnTe detector with a volume of 1000 mm^3 was also investigated for environmental monitoring tasks, a clear conclusion was made, too, that sensitivity of the CdZnTe detector, which only has an active volume of 1000 cm^3, is so low that in a typical natural environment, the peaks are not visible even when this detector is operated for several hours.

It should be added that the operating temperature range for CdZnTe detectors is + 5 °C- + 50 °C and to ensure their stable operation in a wider temperature range, they need to be stabilized thermally, which, actually, is the same as with the scintillator detectors. In addition, the cost of a GDRM-CdZnTe/6000 meter with a volume of 6400 mm^3 significantly exceeds the cost of a GDRM-SrI_2/1.5 scintillation meter with crystal dimensions 1.5" 1.5". Thus, the relatively low sensitivity of CdZnTe detectors makes them unsuitable in REWS for environmental monitoring.

GDRM-SrI2/1.5 meter (c) is developed on the basis of a scintillation SrI_2(Eu) crystal with dimensions of Ø 38 mm 38 mm (Ø 1.5" 1.5") [22]. In addition to the crystal itself, the meter includes a photomultiplier, a signal amplifier, a multi-channel

*cps – counts per second

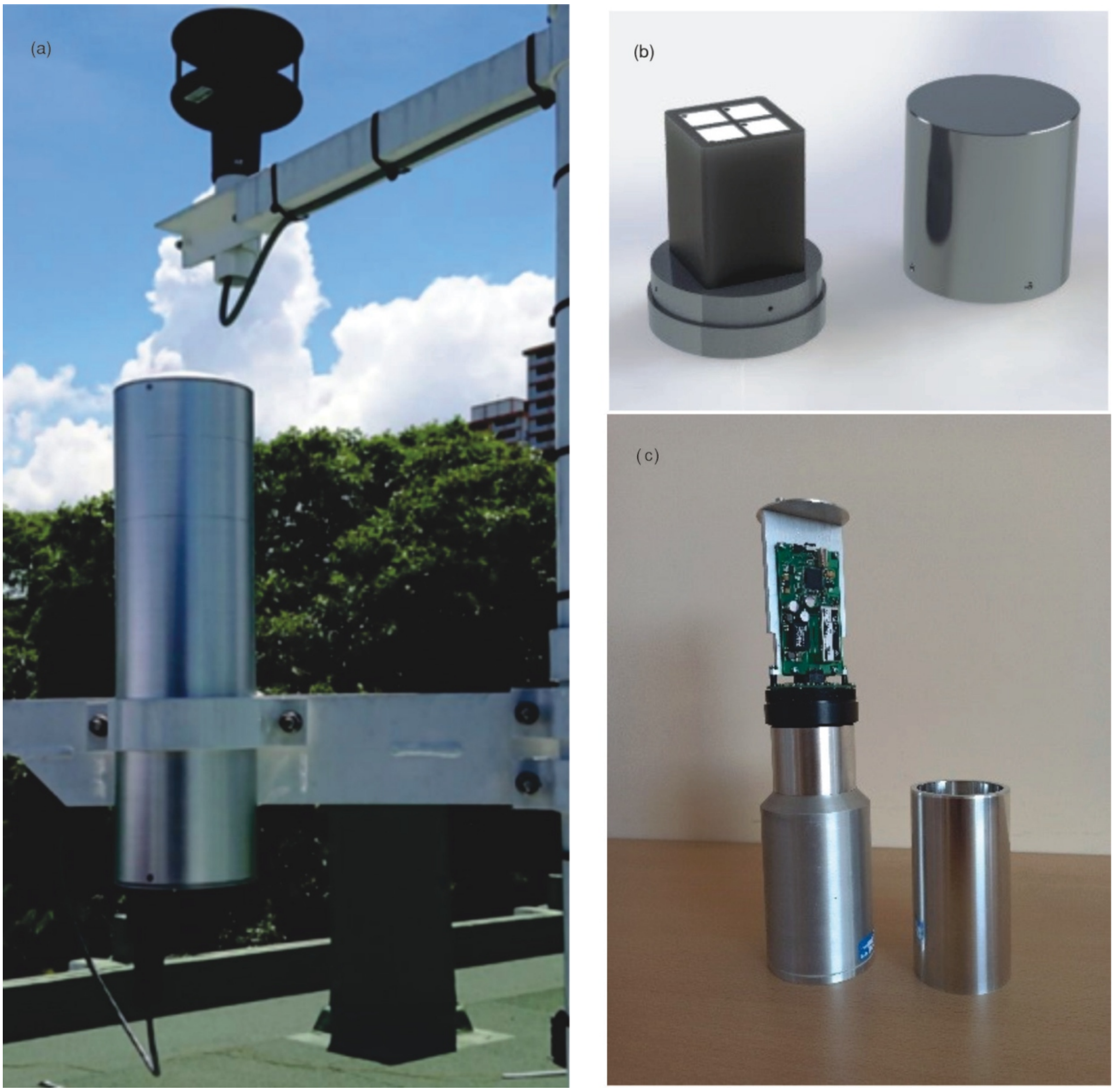

Figure 1. Photos of gamma dose rate meters on the basis of: (a) Geiger-Muller counters, (b) semiconductor CdZnTe detector – with open cap, and (c) SrI_2(Eu) scintillation detector – with open top cap

Table 1. Performance of the developed gamma radiation dose rate meters

Performance	GDRM-GM/2	GDRM-CdZnTe/6000	GDRM-SrI_2/1.5
Energy range	24.6 keV-10 MeV	20 keV-3,0 MeV	20 keV-3,0 MeV
Dose rate range	0.15 μSvh^{-1}-5 Svh^{-1}	10 nSvh^{-1}-0,1 mSvh^{-1}	40 nSvh^{-1}-150 mSvh^{-1}
Detector dimensions	L-(Ø 26 167) mm S-(Ø 9 58.5) mm	6400 mm^3	(Ø38 38) mm (1.5" 1.5")
Energy resolution (662 keV)	None	2-3 %	2,8-3,2 %
Registration sensitivity	L-7.16 cps*/(μSvh^{-1}) S- 0.60 cps/(μSvh^{-1})	140 cps/(μSvh^{-1}) for range 580-700 keV (peak 662 keV), 570 cps/(μSvh^{-1}) for range 20-700 keV	327 cps/(μSvh^{-1}) for range 580-700 keV (peak 662 keV), 683 cps/(μSvh^{-1}) for range 20-700 keV
Background	L-0.37 cps S-0.03 cps	0.014 cps	0.16 cps
Powersupply: – voltage – power	(10-30) V, 1.5 W	(4.5-5.2) V, 2.0 W	(4.5-18) V, 1.5 W
Interface	RS232, RS485	USB	RS485, USB
Temperature range	–30 °C to +60 ºC	+5 °C to +50 °C	–25 °C to +60 ºC
GDRM dimensions	(Ø 100 385) mm	(Ø110 123) mm	(Ø 80 330) mm
Weight	1.5 kg	0.75 kg	3 kg

analyzer, a high-voltage power supply unit, and a temperature stabilization unit for a dedicated peak. The last one allows stabilizing the monitor characteristics in the required temperature range by placing it in a thermostat with a programmable stabilization temperature [23]. When the thermostat is turned on, power consumption increases from 1.5 W to 25 W. The output of the meter is RC485 with galvanic isolation.

The choice of the SrI_2(Eu) scintillator was due to its better energy resolution and a higher gamma-radiation registration sensitivity of all comparable scintillators. The energy resolution at 662 keV energy achieved so far for SrI_2(Eu) is 2.8 %, which is better than the resolution for $CeBr_3$ (4.5 %) and equal to the resolution for $LaBr_3$(Ce) (2.7 %) [12]. $LaBr_3$(Ce), moreover, has its own lines in its own background spectrum in the energy range of 1.4-1.6 MeV, which, other things being equal, invariably worsens the minimum detectable activity (MDA) of the registered radionuclides. Our results showed that of the 25 meters fabricated on the basis of crystals from SrI_2(Eu), 14 meters had an energy resolution at 662 keV not worse than 3.2 %, and another 11 – no worse than 3.1 %.

Meters on the basis of SrI_2(Eu) detectors are certainly superior to replaceable meters with NaI(Tl) detectors because the energy resolution of the former is almost three times higher than the energy resolution of the latter (3 % and 8 % at 662 keV energy, respectively). This makes it possible to determine not only the dose rate of the registered radiation but also the list of radionuclides that most affect the formation of the total dose. Such energy resolution makes it possible to easily identify radionuclides ^{144}Ce, ^{132}Te, ^{143}Ce, ^{131}I, ^{140}Ba, ^{134}Cs, ^{137}Cs, ^{95}Zr, ^{54}Mn, ^{136}Cs, ^{60}Co, ^{140}La, emitted into the atmosphere during accidents at nuclear power plants [1].

Thus, the developed meter can be used in REWS not only as a GDRM in determining the dose rate of external gamma radiation but also as a SrI_2(Eu) spectrometer in the measuring channels of equipment for the unification of radionuclides and the determination of their activity.

DEVELOPMENT OF AN AUTOMATED MONITOR OF AEROSOL RADIOACTIVITY

It was shown in the work [24] that the control of alpha and beta active aerosols' content is the most sensitive way to register changes in the radiation background, which will make it possible to notice an emergency situation at the very initial stage of its course. This is due to the low activity of alpha and beta active natural radionuclides in the air in aerosol form. Therefore, even a small additional flow of such aerosols into the atmosphere will be recorded by the sensors of the early warning system. The sensitivity of the system is increased also owing to the fact that the contribution of alpha and beta active natural aerosols, due to the decay products of radon, is subtracted from the measured total values of activity [25]. Thus, introducing an alpha-beta-radioactive aerosol monitor into the REWS will solve two problems simultaneously: to timely record the fact of the arrival of a radioactive cloud and to determine the concentration of the main dose-forming radionuclides released during the accident, such as ^{90}Sr or ^{239}Pu.

The aerosol radioactivity monitor devised by us, fig. 2, allows to automatically monitor both gamma-ray and alpha and beta emitting radionuclides in aerosols deposited on filters. The filter change system is controlled automatically by the timer and/or by the controlled degree of contamination of the filter, as well as the control and measurement of the pumped air volume in real time. The operation of the entire aerosol radioactivity monitor is controlled using a built-in industrial panel computer running Windows 10 IoT. The installed software supports carrying out the calibration of detectors, checking the operability of all components of the monitor, monitoring the measurement of the alpha, beta, and gamma radiation spectra, as well as reading information about the measured radionuclide activity both at the location of the monitor and remotely on the top-level computer over a 4G/LTE or Ethernet link in ANSI 42.42 format.

Figure 2. Photograph of the aerosol radioactivity monitor

To determine the concentration of alpha- and beta-emitting radionuclides in the developed monitor, two independent spectrometric channels with ion-implanted silicon detectors with an area of 600 mm^2 and an energy resolution of less than 25 keV at the 5.5 MeV energy each, are used. Registration efficiency for such a detector for alpha radiation at ^{241}Am is 20 %   5 %, for beta radiation for ^{90}Sr/^{90}Y – it is 30 %   5 %. The second spectrometric channel is auxiliary and is used to compensate for real time background gamma radiation in the range of beta radiation energies. The measurement range of alpha radiation volumetric activity, performed in the energy range from 2 MeV to 10 MeV, is from 10^{-2} to 10^{5} Bqm^{-3}. The measurement range of beta radiation volumetric activity in the energy range from 70 keV to 3 MeV is from 10^{-1} to 10^{5} Bqm^{-3}.

The gamma radiation measurement unit is designed to be unified so that it allows the installation of different gamma radiation spectrometers depending on the problem being solved. In most applications, following IAEA recommendations [5], scintillation SrI_2(Eu) spectrometers are used to detect gamma radiation from deposited nuclides. However, if it is necessary to increase the number of identifiable radionuclides and improve the accuracy of the determination of volumetric activity, the design of the monitor allows the installation of an HPGe spectrometer (30 % efficiency) with electric machine cooling instead of a scintillation spectrometer, fig. 3. The gamma energy range of an HPGe detector is from 3 keV to 3 MeV, and its energy resolution is less than 1000 eV and less than 2000 eV for the 122 keV and 1.33 MeV energies, respectively [26]. Aerosol radioactivity monitor with such detector has a minimum volumetric activity for gamma radiation of 0.005 Bqm^{-3}.

Figure 3. Photo of the HPGe detector [26] with electric machine cooling

DEVELOPMENT OF AN AUTOMATED IODINE CONCENTRATION MONITOR

The automated gas monitor for detecting and determining the concentration of radioisotope iodine in the air is fully unified with the aerosol radioactivity monitor in the part of the control computer, the filter supply mechanism, the control of pumped air, and the implementation of the gamma ray spectrometric channel. Both monitors also have a single external design, fig. 2. To determine the concentration of radioactive gaseous ^{131}I, a developed spectrometer with a SrI_2(Eu) detector is used, but other scintillation detectors may also be used.

The main difference between an iodine monitor and an aerosol radioactivity monitor is the use of carbon filters. To be able to detect gamma radiation from ^{131}I with high sensitivity, iodine must be absorbed by the carbon material inside a special cartridge. This carbon material is selective, it does not capture other gases, so the sample will be enriched only with ^{131}I, which ensures high sensitivity of its registration. To achieve an effective capture process, a certain time is needed, as well as a fair volume of charcoal and sufficient time for the selected air to flow.

Besides, it is also important to minimize the number of particles that will collect at the air inlet in the iodine monitor, in order to minimize side contamination of the cartridge. For this purpose, an original design of the sampling head has been devised, which carries out the first filtration and blocks all particles larger than 10   m. The second filtration is provided by a filter built into each cartridge on the air inlet side. The container contains 14 such cartridges, which provides at least 28 days of autonomous operation. The command to replace the cartridge depends on the operating time or the degree of contamination, which is measured by the difference in air pressure at the inlet and outlet of the cartridge. The measurement range of ^{131}I volumetric activity is from 10^{-1} to 10^{5} Bqm^{-3}. The software allows to set the values of two programmable thresholds according to the volumetric activity, and also constantly monitors the state of the components of the monitor.

MOBILE AEROSOL MONITOR FOR ALPHA AND BETA EMITTING RADIONUCLIDES

The mobile monitor of alpha-beta aerosols, fig. 4, is designed as a compact version of the aerosol monitor and is used for rapid assessment of a radiation situation, where it is difficult to install a stationary monitor. In accordance with paragraph 4 of Standard IEC 60761-2, it is classified as an alpha spectroscopy monitor with a movable filter-sampler and simultaneous measurements. The mobile monitor of alpha-beta aerosols, if necessary, allows additional connection of

Figure 4. The mobile alpha-beta aerosol monitor

external GDRM, which turns it into a complete mobile analogue of the aerosol monitor.

The mobile monitor is mounted on a two-wheeled transport trolley for easy transportation and operation. The alpha-beta radionuclide measurement channel is identical to the corresponding channel in the automated aerosol radioactivity monitor and retains all its characteristics and capabilities. The capacity of the cassette with filter belt provides autonomous operation for about 90 days. Automatic filter replacement is performed at the end of the measurement time or according to the degree of contamination of the filter.

Since a lead shield is not used in the mobile monitor to save weight, compensation for background gamma radiation is carried out using an additional detector separated from the main one by a beta radiation opaque screen. The readings of the additional, background detector are subtracted in real time from the readings of the main detector.

The separation of long-lived (or so-called "artificial") and natural (daughter products of radon and thoron) radionuclides for both alpha and beta regions of interest (ROI) in the energy spectrum is implemented algorithmically. The algorithm also provides the ability to measure the concentration of radon and thoron in ambient air.

Monitoring and processing of measurements are carried out by the control unit, which is a compact fanless Advantech TPC-651T system with a 5.7-inch LCD display VGA TFT and an integrated Intel® Atom™ processor with low power consumption. The functions of the software include operations control and checking the operability of all monitor units, monitoring, and processing of measurement results. The measurement results are recorded and stored in the internal data storage (SDD/HDD) and can be transmitted to the remote-control center via USB, LAN, or RS485 interfaces.

The system has automatic self-diagnostics. All events, alarms, and/or failures are recorded and described in a log file indicating the date and time of occurrence, and then transmitted to the remote-control center along with monitor status messages and measurement data. This feature enables efficient remote maintenance of the monitor, and accurate diagnosis of a problem, thereby reducing monitor downtime and associated costs. Easy-to-use built-in testing procedures and graphical interfaces make it easy to perform configuration, diagnostics, and calibration procedures on site.

WATER MONITOR DEVELOPMENT

According to IAEA recommendations [1], if the controlled territory is located on an island or on the shore of a body of water where radioactive contamination may occur, automatic monitoring of the aquatic environment should be provided. We have developed a monitor for monitoring aquatic environments on the basis of the SrI_2(Eu) spectrometer. It is made in an IP68 titanium cylindrical housing for working underwater at a depth of up to 25 m. The design of the spectrometer allows its installation both on the wall of a pier in ports and on buoys to control radiation pollution of the water area away from the coastline.

Tests have shown that the developed monitor provides an MDA better than 0.5 BqL^{-1} for the ^{137}Cs with a measurement time of 1 hour. Identification of radionuclides is implemented in accordance with a library list, which includes: ^{144}Ce, ^{132}Te, ^{143}Ce, ^{131}I, ^{140}Ba, ^{134}Cs, ^{137}Cs, ^{95}Zr, ^{54}Mn, ^{136}Cs, ^{60}Co, ^{140}La. The radionuclide library can be configured for specific requirements.

CONCLUSION

As can be seen from the above, the alpha, beta and gamma radiation monitors developed in this work in accordance with the recommendations provide environmental monitoring both in terms of dose rate values in real time and in the activity of specific radionuclides. Closing all the tasks of monitoring the territories, the developed monitors are the basis for the creation of modern radiation early warning systems for radiation accidents.

AUTHORS CONTRIBUTIONS

The development and testing process of the device based on scintillation crystal was made by I. A. Krainukovs. The development of circuit engineering was made by V. S. Litvinsky. A. N. Vlasenko executes the ideology for all monitors structure. V. V. Gostilo has made the total supervision and general concept of the manuscript.

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Received on July 22, 2022
Accepted on January 17, 2023

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РАЗВОЈ НУКЛЕАРНИХ МОНИТОРА ЗРАЧЕЊА ЗА СИСТЕМЕ РАНОГ УПОЗОРАВАЊА НА ЗРАЧЕЊЕ

Приказани су резултати развоја савремених прецизних монитора алфа, бета и гама зрачења за постављање система раног упозоравања на радиоактивну контаминацију у атмосфери и брзу процену насталих претњи. За развој користе се пропорционални бројачи, сцинтилациони SrI_2(Eu) кристали и полупроводници Si, CdZnTe и HPGe детектори. Дизајнирани монитори дају информације како о вредностима дозе у реалном времену, тако и о активности одређених радионуклида. Софтвер контролише режим мерења и дијагностикује стање самих монитора.

Кључне речи: систем раног упозоравања на зрачење, радиоактивни аеросол, мониторинг животне средине, праћење зрачења воде, еквивалент амбијенталне дозе