IMPROVEMENT OF THE MINIMUM DETECTABLE ACTIVITY OF A FREE RELEASE MONITOR FOR SMALL ARTICLES

by

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This paper presents results of the development of a small-sized free release monitor designed for the release of materials, various hand tools, equipment and instruments of nuclear enterprises and laboratories staff that weight up to 50 kg, from radiation control. To increase the registration sensitivity of controlled radionuclides, 12 scintillation units based on a 3" 3" sized NaI (Tl) crystal were used as gamma-radiation detectors. Volume of the measuring chamber of the monitor amounted to 200 L, the chosen thickness of the low-background shielding was 50 mm. The values of the minimum detectable activity of the designed monitor for the point sources ^{123}I, ^{131}I, ^{99m}Tc, ^{18}F were better than 100 Bq with measurement time not exceeding 60 seconds.

Key words: free release monitor, minimal detectable activity, registration efficiency, scintillation gamma detector, radionuclide waste

INTRODUCTION

In the course of its life, any nuclear facility (nuclear power plants, fuel cycle processing plants, nuclear laboratory, medical organizations, *etc.*), in addition to its main product, generates various production waste (work clothes, shoes, tools, computer equipment, materials related to the work of personnel and *etc.*). In addition to activated or contaminated materials that must be evacuated and disposed of as radioactive waste, a large amount of non-radioactive materials can either be recycled or released into free circulation [1]. The purpose of free release monitoring is to reduce the amount of radioactive waste requiring disposal. This task requires the development of procedures and criteria for such release, along with the development of equipment, to ensure highly effective control of contamination of objects and materials at all stages.

As a rule, free release monitors (FRM) designed for these purposes represent a measuring chamber with the volume up to 400 L, where the measured objects are placed, surrounded by passive shielding protection for screening from external background. To measure the activity of gamma-emitting radionuclides in a FRM, gamma-ray detectors based on plastic scintillation materials are usually used [2-4]. The low resolution of such detectors actually makes it possible to measure only ^{137}Cs and ^{60}Co by the radiometric method in windows.

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The existing design of modern FRM is contingent on two main factors. The first is the standards for release from radiation control established by the IAEA [5, 6], as well as by regional regulatory bodies. For example, the EU has adopted the Euroatom directive [7] and the recommendations of the European Commission [8, 9] which also establish norms for release from regulatory control, and somewhat differ from the IAEA standards. The second factor is the size and throughput (kg or tons per hour) of controlled objects.

In some cases, for example, in radiological centres and nuclear medicine clinics, it is often necessary to perform measurements associated with the release from regulatory control of materials and instruments not only for relatively long-lived radionuclides, such as ^{137}Cs and ^{60}Co, but also for short-lived ones, such as ^{131}I, ^{99m}Tc, ^{177}Lu, ^{123}I, ^{18}F, *etc.* [10, 11]. Typically, the mass of the measured objects in such applications does not exceed 50 kg.

This task requires not only a high registration efficiency of the controlling monitors, but also energy resolution sufficient for the minimum detectable activity (MDA) of the FRM to allow monitoring of samples with high throughput. For example, the directive [7] implies the MDA of such FRM should not be worse than 100 Bq for ^{137}Cs and ^{60}Co and not worse than 500 Bq for ^{131}I. The measurement time in practice, however, should not exceed a few minutes in order to ensure high throughput. To solve this problem, the use of plastic scintillators is difficult because of the low energy resolution and, as a consequence, low MDA.

This work is devoted to the results of the development of a small-sized highly-sensitive FRM designed to release materials, various hand tools, equipment and instruments for personnel of nuclear facilities and laboratories weighing up to 50 kg, from radiation control. The main purpose of the paper was to study the possibility of improving the MDA for controlled radionuclides in comparison with the existing analogues [2-4] and practical demonstration of improved MDA characteristics in the developed FRM.

INCREASING THE EFFICIENCY OF RADIONUCLIDES REGISTRATION

For the development of FRM for small-sized objects with improved MDA characteristics, the guidelines of the directive [7] were taken as a basis. The MDA of radionuclides in a particular device is always determined by the number and sensitivity of the detectors used, their energy resolution, low-background shielding, and the measurement time. To increase the registration efficiency and energy resolution of the developed device, instead of the commonly used plastic scintillators [2-4], we used 12 smart detection units (SDU) based on a NaI (Tl) 3" 3" crystal (with a Hamamatsu R6233 photomultiplier) with a built-in digital MCA [12] and a high-voltage power supply. The energy resolution of each SDU at the 662 keV line was not worse than 7 %. To connect the SDU to a PC, a standard industrial RS485 interface with a half-duplex scheme was used, which allows simultaneous start/stop of spectrum acquisition on all 12 SDU. Thus, the developed FRM is actually a 12-channel spectrometer.

The general view of the designed FRM is shown in fig. 1, and its functional block diagram is shown in fig. 2. The FRM is a monolithic box construction with dimensions 800 mm 800 mm 1300 mm based on an aluminium profile frame. Inside the frame, there is a 500 mm 500 mm 800 mm measuring chamber made of stainless-steel sheets with a door for loading the inspected samples inside. To ensure that the entire area of the measuring chamber is covered with sensitive detectors maximally evenly, two SDU were placed on each side wall, so that the centres of the detector crystals lie in the centre of the upper and lower halves of the wall, respectively.

To reduce the influence of gamma background on the FRM measurement results, the measuring chamber is sheathed on the outside with lead plates

Figure 1. General view of the designed FRM

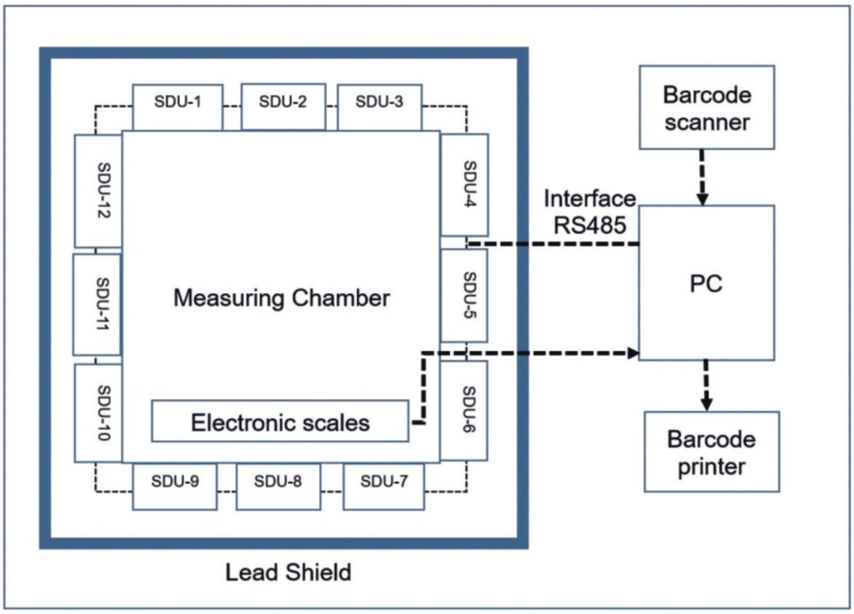

Figure 2. The FRM functional flow diagram

(including the door), forming a continuous low-background lead shield 50 mm thick. Additionally, the FRM is equipped with a barcode scanner and a printer. The weight of the loaded samples is determined by electronic scales built into the chamber. The total mass of the device with the lead shielding was 2800 kg.

The FRM software and operation algorithm

To control the developed FRM, a specialized GammaPro [12, 13] software was created that performs the acquisition and accumulation of spectra, creates/edits a working library of nuclides, efficiency calibration, makes analysis of the spectra and report output.

The operation algorithm of the developed FRM is quite simple. Rectangular containers with tools or materials weighting up to 50 kg, are placed into the measuring chamber. The weight value, which is determined by the scales built into the chamber, is automatically transferred to the GammaPro software for further calculations. A 12-channel gamma spectrometer acquires gamma radiation spectra of objects placed in the camera. The FRM control software analyses the acquired spectra for the content, activity and MDA of ^{131}I, ^{99m}Tc, ^{177}Lu, ^{123}I, ^{18}F, ^{137}Cs, ^{60}Co and other nuclides. After calculating, the program gives the user a report and a decision on the release of the measured objects from radiation control. If the activity of the nuclides exceeds the norm for release, then the report indicates the date when the activity due to decay will be below critical. Based on the analysis results, a bar code is formed, which can be printed and glued to the sample. The barcode scanner is used to read the barcode glued on the measured object and display the information encrypted in it, in a special information window.

CURVE OF FRM REGISTRATION EFFICIENCY

The total curve of registration efficiency from all 12 FRM detectors was calculated by the Monte Carlo method using the MCC-MT program [14]. In the calculation model, it was assumed that the point source is located in the centre of the measuring chamber. The calculation results in the range of 50-2500 keV are shown in fig. 3. As can be seen from the graph, the registration efficiency curve has a classical form, typical for detectors based on NaI(Tl). In this case, the maximum FRM registration efficiency falls within the range of 100-500 keV, which includes all the energy lines of the short-lived isotopes ^{131}I, ^{99m}Tc, ^{177}Lu, ^{123}I, ^{18}F.

Research of MDA of the designed FRM

The MDA is an important characteristic of any spectrometric system. For the level of confidence p = = 95 %, MDA can be represented as [15]

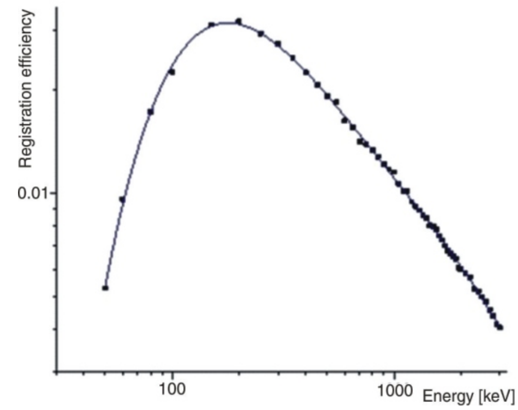

Figure 3. Registration efficiency curve of the developed FRM for a point source located in the centre of the measuring chamber

$$MDA = \frac{2.7 + 4.65(B_{bg})^{1/2}}{\varepsilon I T}$$

where ε is the registration efficiency for a given energy, I – the line intensity, T – the measurement time, and B_{bg} – the number of pulses within the range of the peak minus the number of pulses in the area of the peak.

The MDA was calculated from the background spectrum for the most intense lines of ^{131}I, ^{99m}Tc, ^{177}Lu, ^{123}I, ^{18}F, ^{137}Cs, ^{60}Co nuclides. The background spectrum is shown in fig. 4. The measurement time was 30 minutes. From the background spectrum, the count rates were determined in the corresponding areas of peaks of the nuclides in question.

The results of calculating the MDA of the designed FRM for various radionuclides are presented in tab. 1. As can be seen from the table, the FRM, due to a significantly higher resolution of the applied detectors, makes it possible to measure the activities of not only ^{137}Cs and ^{60}Co, but also short-lived *medical* radionuclides. For a measurement time of 90 seconds this FRM provides MDA values better than 100 Bq for all studied nuclides, except for ^{177}Lu. The higher

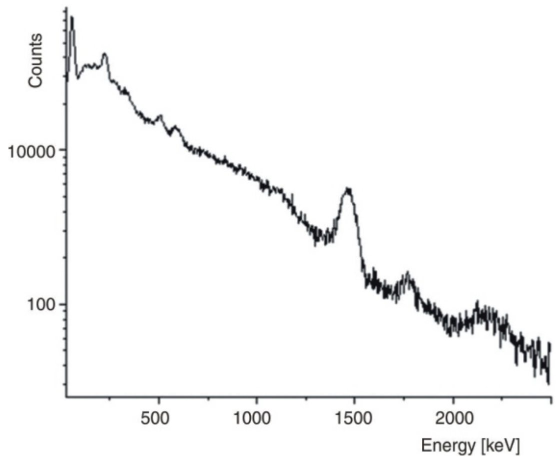

Figure 4. Background spectrum of the designed FRM, acquisition time 61755 seconds

Table 1. The MDA of various radionuclides for the designed FRM

Nuclide	MDA [Bq]	
	T = 60 s	T = 90 s
^{131}I (364.5 keV)	95	75
^{99m}Tc (140 keV)	67.4	55
^{177}Lu (208.3 keV)	685	555
^{123}I (159 keV)	74	58
^{18}F (511 keV)	49	38.5
^{137}Cs (661.8 keV)	121	98
^{60}Co (1332 keV)	108	87

MDA values for ^{177}Lu are due to the fact that its most intense line at 208.3 keV is superimposed on the rather intense line at 238 keV (^{232}Th), which is present in the background spectrum.

Analysis of the characteristics obtained in relation to the existing Directives [5-9] shows that the developed FRM meets all the existing standards for gamma-emitting nuclides in terms of radiation clearance and release from radiation control.

CONCLUSION

As the results of the work showed, the MDA of the developed FRM quite allows it to be used to release from regulatory control of materials and instruments in laboratories and centres of nuclear medicine in accordance with the EU regulations. The values of the minimum detectable activity of the developed monitor for precision sources of short-lived radionuclides ^{123}I, ^{131}I, ^{99m}Tc, ^{18}F are better than 100 Bq with a measurement time not exceeding 60 seconds. At the same time, the throughput of FRM, referred to the standards specifically for ^{137}Cs and ^{60}Co, is about 1500 kgh^{-1} or about 30 samples per hour. In the case of analysis only for short-lived radionuclides, the throughput of the developed FRM can be several times higher, depending on the density of the materials under study.

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AUTHORS' CONTRIBUTIONS

General concept and design of FRM was developed by S. S. Pohuliai as well as the analysis of all the results. All the measurements and simulations in the MCC-MT software were made by I. A. Krainukovs.

REFERENCES

[1] Klein, M., Moers, S., The Free Release of Dismantled Materials, The Practical Case of the BR3 Reactor, *Appl. Radiat. Isot., 53* (2000), 1-2, pp. 323-329
[2] ***, Mirion Technologies, Cronos-4 and Cronos-11 Gamma Object/Tool Monitors https://www.mirion.com/products/cronos-4-11-gamma-object-tool-monitors
[3] ***, VF Nuclear, MCM-300 Tools and Materials Contamination Monitor https://www.vfnuclear.com/en/products/tools-and-materials-contamination-monitor-mcm-300
[4] ***, Thermofisher Scientific, SAM-12 Small Article Monitor https://www.thermofisher.com/order/catalog/product/SAM12L6C1D2L#/SAM12L6C1D2L
[5] ***, IAEA Safety Standards Series No. WS-G-5.1 Release of Sites from Regulatory Control on Termination of Practices
[6] ***, Radioactivity Measurements at Regulatory Release Levels OECD 2006 NEA No. 6186
[7] ***, European Society of Radiology (ESR) Summary of the European Directive 2013/59/Euratom: essentials for health professionals in radiology // Insights Imagin, 2015, 6:411-417
[8] ***, European Commission, Recommended Radiological Protection Criteria for the Recycling of Metals from the Dismantling of Nuclear Installations, Radiation Protection No. 89, Luxembourg, 1998
[9] ***, European Commission, Recommended Radiological Protection Criteria for the Clearance of Buildings and Building Rubble from the Dismantling of Nuclear Installations, Recommendations of the Group of Experts Set Up Under the Terms of Article 31of the Euratom Treaty, RP 113, 2000
[10] Macdonald, J., Gibson, C. J., Calibration of Radioactive Contamination Monitors for Practical Use in Hospitals, *Nuclear Medicine Communications, 23* (2002), June, pp. 573-580
[11] Klett, A., *et al.*, A New Contamination Monitor with Scintillation Detection. 2005 (IEEE Nuclear Science Symposium Conference), Record N 32-4
[12] Finkel, F., *et al.*, Performance Stabilisation of Scintillation Spectrometer for Aerosol Monitors, *Nucl Technol Radiat, 34* (2019), 1, pp. 72-78
[13] Nurgalejev, R., *et al.*, Spectrometric Performance of a HPGe Semi-Planar Detector with Large Diameter, NIM A 985, 2021, 164712
[14] Finkel, F. V., *et al.*, Application of the Monte Carlo Method for Spectrometer Calibration to Determine

the Surface Activity of Radionuclides Deposited on the Ground, *Physics of Atomic Nuclei, 79* (2016), 9-10, pp. 1424-1428

[15] Currie, L. A., Limits for Qualitative Detection and Quantitative Determination, Application to Radiochemistry, *Anal. Chem., 40* (1968), 586, ACS Publications

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ПОБОЉШАЊЕ МИНИМАЛНЕ ДЕТЕКТАБИЛНЕ АКТИВНОСТИ МОНИТОРА КОНТАМИНАЦИЈЕ ЗА ПРЕДМЕТЕ МАЊИХ ДИМЕНЗИЈА

У раду су приказани резултати развоја монитора контаминације мале величине, пројектованог за потребе ослобађања од радијационе контроле материјала, различитих ручних алата, опреме и инструмената из нуклеарних постројења и лабораторија, тежине до 50 кг. Ради повећања осетљивости детекције радионуклида мерењем гама зрачења коришћено је 12 сцинтилационих сонди са NaI(Tl) димензије 3 3 . Запремина коју монитор може да мери износи 200 литара, а заштитом од зрачења дебљине 50 mm обезбеђује се низак фон. Добијене вредности минималне детектабилне активности пројектованог монитора за тачкасте изворе ^{123}I, ^{131}I, ^{99m}Tc, ^{18}F биле су мање од 100 Bq за време мерења које није било дуже од 60 секунди.

Кључне речи: монитор контаминације, минимална детектабилна активност, ефикасност детекције, сцинтилациони детектор гама зрачења, радиоактивни отпад