

The remote-controlled spectrometric system for searching and characterization of high-level radioactive waste

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Abstract - In 2003, the work on environmental rehabilitation of the former coastal technical base of the Russian Navy in Gremikha town, with technical and financial support of the State Atomic Energy Corporation “Rosatom” and the French Alternative Energies and Atomic Energy Commission had begun. In 2017, the task of searching, characterization and packaging in transport containers for export to the long-term storage of high-level waste containing ^{152}Eu was set as part of this program. These wastes were presented as fragments of reactor control and safety system rods of first generation nuclear submarine reactors and were placed in temporary storage containers mixed with other radioactive waste and construction garbage on the territory of the Gremikha branch of the North-West Center “SevRAO”.

To accomplish the task, the spectrometric system “Gamma-Pioneer” was developed and made at National Research Center “Kurchatov Institute”. The system consist of a collimated spectrometric gamma-ray CdZnTe detector 500 mm³ volume, a dosimeter for continuous measurement of ambient equivalent dose rate at the instrument location and a color video camera with zoom lens. The device is placed in a protective frame and installed as an attachment implements on robot Brokk-90. Communication between the measuring system and the control computer is carried out using USB extender and 100 meters cable. The video camera is connected to the control computer via Ethernet.

To search for the most active sources of gamma radiation, an equivalent dose rate calculation program using the results of measured instrumental spectra was developed. The characterization of the extracted fragments was carried out by activity evaluation of the main radionuclides polluting the samples under investigation. The paper describes design and operating principles of the system, as well as methods for the equivalent dose rate calculation and the activity evaluation of the fragments of reactor control and safety system rods based on spectral measurements obtained using the developed radiometric system

Keywords—radiation detection, gamma spectrometry, radioactive waste, detectors.

I. INTRODUCTION

Environmental rehabilitation of the former coastal technical base of Russian Navy in Gremikha town (today, this is Gremikha branch of the North-West Center “SevRAO” branch of FSUE “RosRAO”, the closed city Ostrovnoy) is currently one of the largest projects addressed to solve the “nuclear legacy” problems in the North-West region of Russia. The works began in 2003 under the Global Partnership G-8 program with the financial and technical support of Atomic Energy Commission of France and the Ministry of Atomic Energy (Rosatom) [1].

At the first stage, the arrangements to improve the radiation situation, modernize the infrastructure and ensure

the personal safety were implement. Specialists of NRC “Kurchatov institute”, NIKIET, VNIPIET and other conducted a comprehensive engineering and radiation survey of buildings, structures, territory and coastal waters to obtain initial data. In conditions of an increased radiation situation, in building 19 and in the open site for storage of spent nuclear fuel (SNF) and solid radioactive waste (SRW), remote identification of γ -radiation sources was carried out. Fragments of SNF were located in the open site. In concrete containers and in two packages with radioactive waste highly active sources with ^{152}Eu lines in spectra were placed. This suggest that the founded sources are fragments of reactor control and safety system rods of first generation nuclear submarine reactor [2].

In the period from 2008 to 2013, more than 800 SNF assemblies were removed from the temporary storage site to Production Association “Mayak” for reprocessing [3]. From 2013, Gremikha branch became the first facility among the former coastal technical bases, freed from nuclear fuel of submarine’s water-cooled reactors.

In 2017, with the financial support of the State Corporation Rosatom, works aimed at solving the task of handling high-level radioactive waste (HLW) accumulated during the operation of nuclear submarines, including the preparation and removal of SRW and HLW began. The main objects of search were reactor control and safety system rods stored in abnormal conditions in temporary packages and concrete containers in building 19 and in the open area of temporary storage of SRW. For the searching and sorting of radioactive waste by category and activity, NRC “Kurchatov institute” developed the spectrometric system “Gamma-Pioneer”.

II. SPECTROMETRIC SYSTEM “GAMMA-PIONEER”

A. System designated purpose

Spectrometric system “Gamma-Pioneer” is designed to search for and identify highly active objects in conditions of intense gamma radiation and conducting surveys and scouting of contaminated premises. Using the CdZnTe detector allows not only determine the radionuclide composition of contamination, but also to calculate the dose rate generated by objects in the field of view of the system.

B. System composition

Spectrometric system “Gamma-Pioneer” consist of (Fig.1):

- The measuring part, which is mounted on the boom of the Brokk-90 robot using a standard mount;
- Control computer for managing the system and processing results;

C. Protective case with installed inside power supply unit and USB extender.

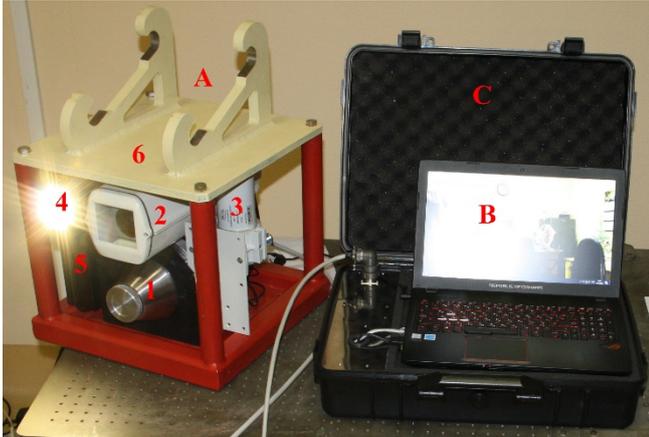


Fig.1. Photo of the “Gamma-Pioneer” system with a control computer and a protective case.

The connection between the control computer and the measuring system is carried out using a USB extender and a twisted pair cable 100 meters length.

The measuring part of system consist of (Fig.1):

1. Collimator with internally mounted spectrometer μ SPEC500 [4];
2. Color video camera with a trans focal lens;
3. Gamma radiation detector DBG-S11D for measuring the dose rate at the point of the system location [5];
4. LED floodlight;
5. Waterproof box with USB port extender and power supply, installed inside;
6. Protective frame that serves to protect the measuring system’s components from damage due to accidental impact.

The collimator of the measuring system is made of lead and placed in a stainless steel body. To eliminate the influence of the characteristic radiation of lead on the measurement result, the micro spectrometer is placed in a brass case with a wall thickness of 2 mm, and only then is installed in a collimator. Also, to reduce the influence of the characteristic radiation of lead, and in addition, to change the collimation angle and to reduce the flux of gamma quanta on the detector surface, two interchangeable brass inserts into collimator with an internal diameter of 10 mm and 5 mm were made.

When examining contaminated objects under conditions of strong radiation fields, it may be necessary to take into account background radiation. For this, a lead plug covering the collimator hole at the time of the background spectrum collection, was made.

To reduce the number of gamma quanta, determined by scattered radiation, and falling on to the detector, a filter consisting of two glued together round plates of lead and brass, is used. If necessary, the filter is installed in front of the collimator hole with a brass plate inwardly.

In operation, the measuring part of system, installed on robot Brokk-90, scans the inspected object in order to find the most active fragments (in our case, control and safety system rods have been). The video image of the object under examination, as well as gamma spectrum, the dose rate values,

measured by collimated and “opened” detectors are displayed on the screen of the control computer. Next, another robot extracts the fragment of interest to clarify its radionuclide composition, measure activity and package it into a transport container.

III. THE METHOD OF DOSE RATE CALCULATION

The result of the registration of radiation by spectrometric detector is the instrumental spectrum $N(E)$. This information is sufficient to estimate the dose rate generated by the registered radiation, and the formula for calculating the equivalent dose rate can be represented as follow [6]:

$$\dot{H} = A_{calibr} * \sum_{i=0}^N \frac{N_i * K_{ei}}{t}, \quad (1)$$

where A_{calibr} – calibration coefficient; N_i – number of pulses in channel i ; K_{ei} – weighting coefficient for channel i ; N – number of channels in spectra; t – lifetime in seconds.

The weighting coefficient K_{ei} allows taking into account the energy dependence of the registration efficiency of a spectrometric detector. This dependence is calculated based on the data obtained by the Monte-Carlo method, and can be written in the form of a six-order polynomial:

$$K_{ei} = \sum_{j=0}^6 a_j E_i^j, \quad (2)$$

where a_j – polynomial coefficients; E_i – energy of channel i , calculated by formula:

$$\begin{aligned} E_i &= (i - a_c) / b_c, \\ a_c &= N_1 - b_c * E_1, \\ b_c &= (N_2 - N_1) / \Delta E, \\ \Delta E &= E_2 - E_1, \end{aligned}$$

N_1 and N_2 – the channel numbers of the instrumental spectrum corresponding to the positions of the total absorption peaks of gamma quanta with energy E_1 and E_2 . In this case, photo peaks ^{137}Cs (661.657keV) и ^{60}Co (1332.501keV) were used for the energy calibration of the spectrometer.

To determine the calibration coefficient A_{calibr} , a ^{137}Cs source with a known activity of $1.5 * 10^{10}$ Bq was used. The source was installed at a distance of 10 to 100 centimeters from spectrometer’s detector, placed in collimator of system, in steps of five centimeters. Measurements of the dose rate generated by the source at the detector’s location were carried out for different distances, as well as for different collimator variants. Collimators with an internal diameter of 5 mm and 10 mm were used. In addition, measurements were carried out with a lead-brass filter installed in front of the collimator hole, and without it. The theoretical dependence of the dose rate generated by the source at the detector’s location on the distance between them was calculated for all four options. According to the measurement results and theoretical data, calibration coefficients were calculated. Further, they were taken into account in the program for calculating the dose rate of radiation registered by the spectrometric detector.

IV. THE METHOD FOR CALCULATING THE ACTIVITY OF RADIONUCLIDES

The radionuclides of ^{152}Eu and ^{154}Eu contained in reactor control and safety system rods have complex emission spectra consisting of a large number of lines with different quantum yield for decay.

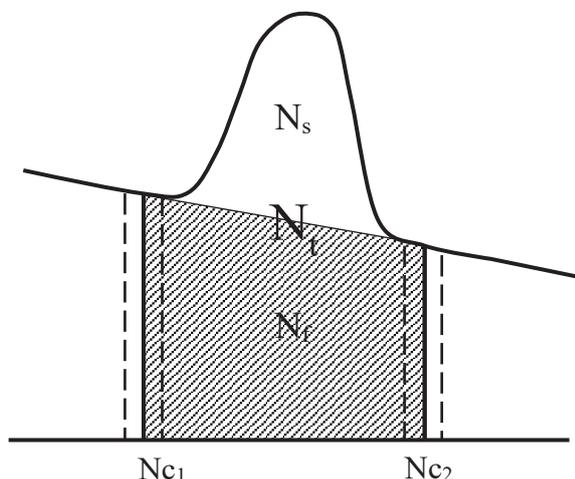


Fig.2. Graphic representation of the peak of the radionuclide in spectrum.

It has known that for a specified measurement geometry, the activity of a radionuclide is proportional to counting rate in the total absorption peak, i.e. equal to the ratio of the useful signal to the live time of measurement multiplied by the calibration coefficient:

$$A = C_{calibr} * \frac{N_s}{T}, \quad (3)$$

where N_s – useful signal, C_{calibr} – calibrating coefficient in Bq/(impulse/s), T – life time of measurement in seconds.

The useful signal is equal to the difference of the total signal area in selected range (the number of registered impulses in the total absorption peak) and the background signal:

$$N_s = N_t - N_f, \quad (4)$$

where N_t – number of registered impulses in the total absorption peak in selected range, N_f – background signal.

To estimate the calibration coefficients, a mathematical model of the radiometric system, in which it was assumed that the source of gamma radiation corresponded to the size, material and radionuclide composition of the object of study (reactor control and safety system rods), was built.

Since it was supposed to measure the objects at different distances, the calibration coefficient was presented in form of a polynomial:

$$C_{calibr} = \sum_{i=0}^2 b_i x^i, \quad (5)$$

where x – distance between the object and the detector in meters; b_i – polynomial coefficients.

The determination of the number of pulses in the total absorption peak minus the background substrate was carried out in standard way (according to formula (4)). The total

number of pulses in the range where the peak is located, was determined by the sum:

$$N_t = \sum_{i=N_{c1}}^{N_{c2}} N_i, \quad (6)$$

and the background signal was calculated using the trapezoid area formula (Fig.2):

$$N_f = (a + b) * c, \quad (7)$$

$$c \equiv (N_{c2} - N_{c1} + 1) / 2,$$

$$a = (N_{j-1} + N_j + N_{j+1}) / 3, \text{ where } j=N_{c1},$$

$$b = (N_{k-1} + N_k + N_{k+1}) / 3, \text{ where } k=N_{c2},$$

N_{c1} and N_{c2} – numbers of the initial and final channels of the selected range.

In radiometric measurements, when the activity of radionuclides is estimated by the counting rate at the total absorption peak, it is also important to know the statistical error of this quantity:

$$\Delta N_s = 2 * \sqrt{N_t + c^2 * (N_a + N_b)} \quad (8)$$

$$N_a = \frac{1}{9} \sum_{i=N_{c1-1}}^{N_{c1+1}} N_i,$$

$$N_b = \frac{1}{9} \sum_{i=N_{c2-1}}^{N_{c2+1}} N_i.$$

In the case when the statistical error of measurement is greater than the useful signal ($N_s < \Delta N_s$) or the useful signal is less than zero, the activity is considered equal to zero.

V. THE RESULTS OF SPECTROMETRIC SYSTEM USING

In the summer of 2018, the spectrometric system “Gamma-Pioneer” installed on the Brokk-90 robot was used to search and characterize high-level waste containing ^{152}Eu , housed in two storage facilities, placed on the territory of branch Gremikha.



Fig.3. The location of the equipment during the measurements of the rods activity in the process of sorting radioactive waste. 1 – measuring part of system “Gamma-Pioneer”, 2 – Brokk-90 robot, 3 – Brokk-160 robot, 4 – rod fragment, 5 – inspected container, 6 – construction waste container, 7 – low level waste container. 8 – transport container for high level waste.

In the first storage, the waste was located in two metal containers. It was necessary to find highly active fragments of control and safety system rods among the medium and low

active waste and construction debris contained in containers. The Brokk-160 robot was used to extract the contents from the inspected container.

Figure 3 shows the disposition of the equipment during the measurement of activity of one of the rods fragments found in the container. An example of the instrumental spectrum is presented in Figure 4.

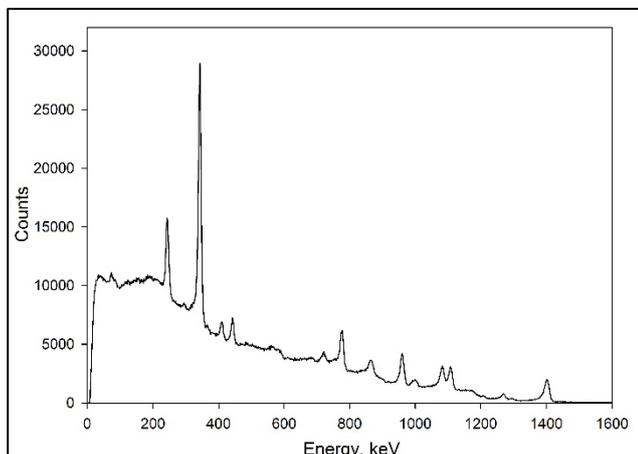


Fig.4. An example of an instrumental spectrum of radiation of control and safety rod.

As a result, twenty-three fragments of control and safety system rods with a total activity of $3.55 \cdot 10^{12}$ Bq were found and placed in a transport container. The results of the activity evaluation of radionuclides ^{152}Eu , ^{154}Eu , ^{60}Co и ^{137}Cs , contained in the rods loaded in the transport container are presented in Table I.

TABLE I.

$A(^{152}\text{Eu}),$ Bq	$A(^{154}\text{Eu}),$ Bq	$A(^{60}\text{Co}),$ Bq	$A(^{137}\text{Cs}),$ Bq	A_{Σ}, Bq
$3.17 \cdot 10^{12}$	$3.57 \cdot 10^{11}$	$2.25 \cdot 10^{10}$	0.00	$3.55 \cdot 10^{12}$

The purpose of the work in the second storage was the extraction of high-level waste in the form of fragments of control and safety system rods from three containers, which, according to the results of previous surveys, contained the most active radioactive waste and made a significant contribution to the dose rate at the site. Preliminary spectrometric measurements showed that these containers contain ^{152}Eu .

As result, twenty-two rods fragments with total activity of $9.96 \cdot 10^{12}$ Bq were loaded into the second transport container. The results of the activity evaluation of radionuclides ^{152}Eu , ^{154}Eu , ^{60}Co и ^{137}Cs , contained in the rods loaded in the transport container are presented in Table II.

TABLE II.

$A(^{152}\text{Eu}),$ Bq	$A(^{154}\text{Eu}),$ Bq	$A(^{60}\text{Co}),$ Bq	$A(^{137}\text{Cs}),$ Bq	A_{Σ}, Bq
$8.83 \cdot 10^{12}$	$9.14 \cdot 10^{11}$	$2.16 \cdot 10^{10}$	0.00	$9.96 \cdot 10^{12}$

VI. CONCLUSIONS

Spectrometric system “Gamma-Pioneer” can be effectively used for the exploration and survey of objects in conditions of high-levels of radioactive contamination. It is especially necessary during emergency abnormal operations

at HLRW and SNF storage facilities, decommissioning of nuclear facilities, searching for lost sources (for example RITEGs) and emergency response.

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