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Gamma-spectrometer for water areas and bottom sediments radiation monitoring

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Abstract

In order to solve the problem of continuous or periodic monitoring of water areas affected by radioactive contamination in the result of scheduled emissions in nuclear power plants or in the result of emergency situations in nuclear fuel cycle plants we need to develop measurement instruments with advanced mathematics and program support to assess the level of radioactive contamination with required accuracy. The aim of theoretical research was to optimize detection device construction, estimate spectrometer metrological parameters in given measurement geometries, and determine effective position of detection device in the process of *in situ* measurements.

This device consists of spectrometric scintillation probe packed into sealed container (detection device) based on NaI(T1) crystal of Ø 63 × 63 mm or Ø 63 × 160 mm size, cable reel with deep-sea cable and a tablet PC for data processing and displaying. The container withstands static hydraulic pressure up to 5 MPa and can be used for measurements at depths of 500 m maximum. Probe measures energy distribution of gamma-radiation with energy from 70 keV to 3000 keV. The implemented three-dimensional system for detection device position and orientation determination allows automatic operation of the device (without operator) for water areas or bottom sediment scanning. The spectrometer can output measurement results with three-dimensional geographical coordinates as index maps of distribution with necessary resolution and accuracy. Monte Carlo models of spectrometer and controlled objects are developed in order to determine the detector response functions to given radionuclides in given measurement geometries without use of expensive standard measures of activity.

Multifunction gamma-spectrometer for *in situ* radiation monitoring of water areas and bottom sediments was developed and constructed. In the result of theoretical researches the response functions have been calculated in the form of theoretical spectra of monitored radionuclides in definite measuring geometries. The results of mathematical modeling of the gamma-emitting transfer process allowed to estimate effective position of detection device for *in situ* measurements of specific activity radionuclides ^{134}Cs and ^{137}Cs in bottom sediments.

Keywords: submersible gamma-spectrometer, detection efficiency, geometry of measuring, *in situ* measuring.

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Гамма-спектрометр для радиационного мониторинга акваторий и донных отложений

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Задачи постоянного или периодического мониторинга водоемов, подвергшихся радиоактивному загрязнению в результате штатных выбросов АЭС или в результате возникновения нештатных ситуаций на предприятиях топливного ядерного цикла, приводят к необходимости разработки соответствующих средств измерений с современным математическим и программным обеспечением, позволяющих оценить уровень радиоактивных загрязнений с заданной точностью. Цель теоретических исследований заключалась в оптимизации конструктива устройства детектирования, определении метрологических параметров спектрометра в заданных геометриях измерения, определении эффективного положения устройства детектирования спектрометра в процессе *in situ* измерений удельной активности радионуклидов ^{134}Cs и ^{137}Cs в донных отложениях с использованием разработанных Монте-Карло моделей: устройства детектирования, воды и донных отложений.

Спектрометр представляет собой многофункциональный прибор, состоящий из размещаемого в герметичном контейнере спектрометрического сцинтилляционного блока детектирования с кристаллом NaI(T1) размерами Ø 63 × 63 мм или Ø 63 × 160 мм, вьюшки с глубоководным кабелем и планшетного компьютера для обработки и отображения информации. Контейнер устойчив к статическому гидравлическому давлению до 5 МПа, что позволяет проводить измерения на глубинах до 500 м. Устройство детектирования позволяет измерять энергетическое распределение импульсов гамма-излучения с энергией от 70 до 3000 кэВ. Реализованная система определения положения устройства детектирования в пространстве позволяет использовать спектрометр в автоматическом режиме (без участия оператора) для сканирования водной акватории и донных отложений. Результаты измерения заданной величины с трехмерными географическими координатами могут быть оперативно представлены в виде карт-схем распределения с необходимой дискретностью и точностью. Для определения функций отклика детектора к заданным радионуклидам в требуемых геометриях измерения без использования физических дорогостоящих стандартных мер активности разработаны Монте-Карло модели спектрометра и объектов контроля.

Для радиационного контроля водной среды и донных отложений методом *in situ* разработан и изготовлен многофункциональный портативный гамма-спектрометр. В результате теоретических исследований были рассчитаны функции отклика спектрометра к контролируемым радионуклидам в заданных геометриях измерения. Результаты математического моделирования процесса переноса гамма-излучения позволили определить эффективную позицию устройства детектирования в процессе *in situ* измерений активности радионуклидов ^{134}Cs и ^{137}Cs в донных отложениях.

Ключевые слова: погружной гамма-спектрометр, эффективность регистрации, геометрия измерения, *in situ* измерения**.**

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Introduction

Water systems are an important part of the environment and are also exposed to radiation pollution as the result of accidents at nuclear fuel cycle enterprises.

Radiation monitoring of the environment, in particular, water areas and sediments, is a prerequisite for ensuring radiation safety during normal operation of nuclear power plants, as well as in the case of emergency situations at nuclear fuel cycle enterprises. Due to technological requirements nuclear power plants are located near large bodies of water (seas, lakes, large rivers) for heat removal and core cooling [1]. Events at Fukushima nuclear power plant have showed that radioactive substances fell not only on fruit and rice fields, but also in ponds and ocean, thus water pollution monitoring represents an urgent and important task [2, 3]. Significant amount of radionuclides which accumulates at the bottom as a result of sedimentation, are absorbed by fish and plants, and may eventually get into the human body [4]. Sediments radiation control is the same important task along with water radiation control.

Activity of controlled radionuclides is usually estimated by the method of representative sampling with transportation to laboratory, preparation and measurement in stationary gamma-spectrometers. Despite attempts to reduce measurement result uncertainty associated with sample selection and preparation activities for radiation control of sediments, this technique has some major disadvantages associated primarily with a high probability of random and systematic errors. Due to difficulties with obtaining of quality bottom samples and time-consuming measurement, there emerges a need for activity measurement of controlled radionuclides by *in situ* [5].

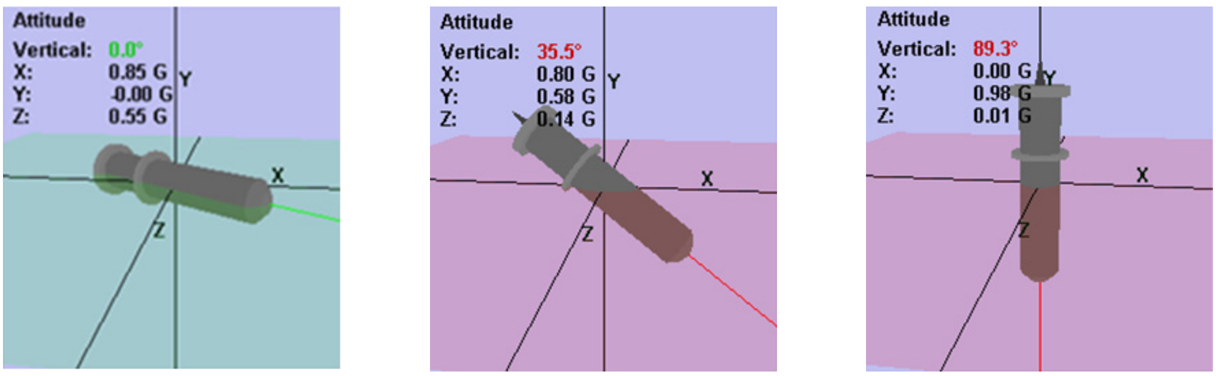
Specialists in many countries are developing and manufacturing submersible gamma-spectrometers for water radiation monitoring [2, 5–11]. Theoretical and experimental researches presented in their works have been carried out to optimize and improve metrology and design parameters of equipment, aimed primarily for monitoring and water radiation control. At the same time, events at Fukushima nuclear power plants in 2011 have showed that determination of controlled radionuclides' activity in sediments is still a high priority task [3]. Portable and lightweight submersible spectrometers with in situ measurement functionality allows for rapid estimation of specific activity of controlled radionuclides in water and bottom sediments with required accuracy [2, 5].

The aim of theoretical researches have consisted in optimization of detection device construction, definition of metrological parameters of spectrometer in special measuring geometries, estimation effective position of detection device to measure specific activity radionuclides ^{134}Cs and ^{137}Cs in bottom sediments by in situ, using Monte-Carlo models of detection device, water and bottom sediments.

Materials and methods

Submersible spectrometer [12] (hereinafter spectrometer) is a modular instrument, consisting of temperature and impact resistant watertight stainless steel container (detection device) with scintillator probe based on NaI(Tl) detector of Ø 63×63 mm or Ø 63×160 mm size inside, deep water cable reel and tablet computer.

Spectrometer withstands static hydraulic pressure up to 5 MPa, which is equivalent to immersion to 500-meter depth. The position of detection device is determined by a position sensor (gyroscope), which is used to identify the moment when the de-



a *b* *c*

Figure 1 – Possible positions of detection device in water: *a* – horizontal position; *b* – intermediate position; *c* – vertical position

tection device contacts the bottom, and for its proper positioning (Figure 1).

Additionally, spectrometer is equipped with a pressure sensor to determine the depth of immersion and humidity sensor to control its integrity. Communication with detection device is carried out by means of cable. Data from detection device are sent to tablet computer and processed by special application software. The values of energy distribution of gamma-radiation pulses, result of radionuclides identification, specific activities of controlled radionuclide or ambient dose equivalent rate of gamma-radiation are displayed on tablet computer screen.

Energy range of detected gamma-radiation, in which the energy distribution is measured, is from 70 to 3000 keV. Relative energy resolution for gamma-radiation of ^{137}Cs (661.7 keV) does not exceed 8 %. Measurement range of gamma-radiation dose rate is from 0.01 to 100 µSv/h.

Spectrometer has internal systems for continuous automatic LED stabilization of energy scale and digital temperature compensation of measuring path. Operating temperature range of spectrometer is from –20 °C to +50 °C.

Numerical Monte-Carlo simulation is a generally recognized solution for verification, calibration, design optimization, and definition of spectrometers' metrological parameters and basic provisions of measurement procedures, when reference materials are not available or cannot be created [13–17].

MCNP software (Monte-Carlo N-Particle Transport, Los Alamos National Labaratory, USA) of version 4A has been used for Monte-Carlo simulation [18]. At the primary stage, the detection device verification has been carried out with standard spectrometric gamma sources containing gamma-emitting radionuclides ^{134}Cs and ^{137}Cs. In the course of experiment and Monte-Carlo simulation the sources were located at a distance of 5 cm from lateral surface of container with detection device. Amplitude deviation of full absorption peak (FAP) of gamma-radiation with energy of 661.7 keV for ^{137}Cs as well as 597.4 keV and 796.4 keV for ^{134}Cs at the level of 2–3 % showed a high degree of spectrometer's compliance with Monte-Carlo model.

Determination of detection device's response function to radionuclides ^{134}Cs and ^{137}Cs in given measurement geometry has been carried out using Monte-Carlo model of water in 4π geometry and of sediment in 2π geometry with specified radionuclide uniformly distributed in specified volume of water or sediment.

For determination of detection efficiency for specified value of gamma-radiation energy the effective volume of water has been taken into account, which is determined by the value of effective radius – radius of contaminated sphere generating more than 90 % of spectrometer response function [19]. Here the tolerance of spectrometer readings for water measurement, which volume is determined by effective radius, is within the range from 5 to 10 % relative to spectrometer readings for measurement of sphere with conventionally infinite radius containing gamma-photons uniformly distributed over its volume.

The results of Monte-Carlo simulation in 4π measurement geometry in the form of calculated theoretical spectra allowed to determine the relation of detection efficiency to the energy of gamma-radiation for spectrometer based on NaI(Tl) scintillation crystal of Ø 63 × 63 mm and Ø 63 × 160 mm size (Figure 2), which is used for calculation of specific activity of most dose-forming radionuclides [20].

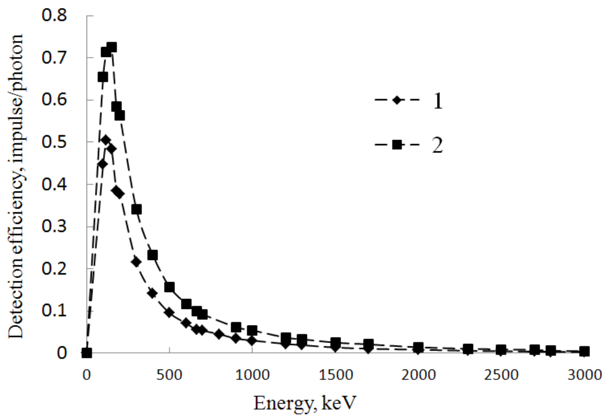
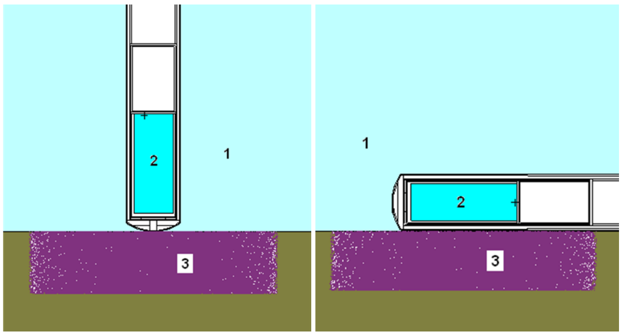


Figure 2 – The relation of detection efficiency to gamma-radiation energy in 4π measurement geometry based on NaI(Tl) scintillation detector: 1 – Ø 63 × 63 mm size; 2 – and Ø 63 × 160 mm size

One of the most important spectrometer's parameters in any geometry is sensitivity to measured radionuclide or relation of detection efficiency to energy of gamma-radiation. Spectrometer's detection limit depends on this metrology parameter along with mathematical algorithm of processing of gamma-impulse energy distribution. In order to set requirements for sediment radiation control methods and find optimal detection device position relative to the object of control in which the spectrometer demonstrates maximum sensitivity to controlled radionuclides. Monte-Carlo model of detection device and contaminated bottom sediments with contaminated layer thickness D = 10 cm has been used (Figure 3).



a *b*

Figure 3 – Monte-Carlo model of detection device and contaminated bottom sediments in in situ measurement geometry: *a* – Vertical; *b* – Horizontal. 1 – Seabed layer of water; 2 – NaI(Tl) detector of 63 × 160 mm size; 3 – Bottom sediments

The density of Monte-Carlo sediment model has been taken as 1.3 g/cm3. See Table [19] for elemental composition of sediments used in simulation of gamma-radiation transfer in 2π measurement geometry.

Table

Elemental composition of sediments

Chemical element	W/w, %
Si	24.32
Al	5.07
Fe	1.04
Ca	0.31
Mg	0.62
K	1.21
Na	1.08
O	62.18
P	0.03
S	0.07
C	3.53
H	0.38
N	0.19

Figure 4 shows the dependence of integral spectrometer response from the radius of sediment site contaminated by ^{137}Cs radionuclide for detection device based on NaI(Tl) scintillation crystals of Ø 63 × 63 mm and Ø 63 × 160 mm size when placing the detection device vertically and horizontally relative to the source of gamma-radiation.

Seabed layer of water has a significant impact on detector response function during in situ measurements of sediments. Seabed water layer diffuses and weakens gamma-radiation emitted from the source (sediment layer contaminated by radionuclides) in detector direction. Scattered gamma-photons make additional contribution to low-energy part of spectrometer response function. This effect increases when detection device is used according to the scheme shown in Figure 4*a* with detection device based on scintillation crystal, which length exceeds its diameter. In this case integral response in the form of response function increases, resulting in reduced detection limit, while significant increase of scattered component introduces additional error into determination of thickness of contaminated sediment layer. Such factors as small effective area of gamma-radiation source relative to the size of the scintillation crystal, lower value of scattered component of instrument spectrum relative to response in FAP, higher sensitivity of measurement by side spectrometer's surface demonstrate the advantages of measurement of contaminated sediments according to scheme shown in Figure 4*b*. Also, fast and accurate determination of measurement start (Horizontal position on the bottom) allows reduction in time required to carry out measurements at check point.

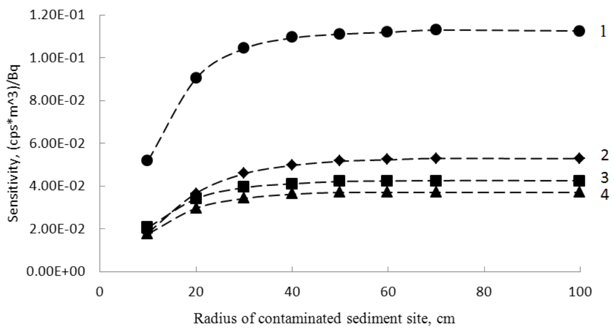


Figure 4 – Dependence of integral spectrometer response from the size of contaminated sediment site: 1 – horizontal position of detection device based on NaI(Tl) crystal of Ø 63 × 160 mm size; 2 – vertical position of detection device based on NaI(Tl) crystal of Ø 63 × 160 mm size; 3 – horizontal position of detection device based on NaI(Tl) crystal of Ø 63 × 63 mm size; 4 – vertical position of detection device based on NaI(Tl) crystal of Ø 63 × 63 mm size

Conclusion

The peculiarity of the conducted researches is in the application of Monte-Carlo simulation method in the achievement of the objectives.

Monte-Carlo models of detection devices, water and sediments have allowed determination of necessary spectrometer response function in the form of spectra of controlled radionuclides in required measurement geometries.

The results of theoretical research allowed determination of metrological parameters of detection devices based on NaI(Tl) scintillation crystals of Ø 63 × 63 mm and Ø 63 × 160 mm size in basic measurement geometries (2π and 4π) and establishing a set of rules and procedures for specific activity measuring of controlled radionuclides without sampling and preparation of water and sediment samples.

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