Comparison of underwater in situ gamma-ray spectrometers with NaI:Tl and $SrI_2:Eu^{2+}$ scintillators

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Based on experiments in a thermal chamber, temperature models for the detector energy scale were refined, relating the position of the photopeak in the spectrum with the energy of the registered gamma quantum depending on the temperature of the detectors and the high voltage of the photomultiplier. The detection efficiencies were determined from previously measured spectra of certified aqueous solutions of radionuclides in a water tank: the obtained experimental values were compared with the theoretical findings of Monte Carlo simulations of the detectors. Based on the analysis of spectra obtained in the results of field trials of the submersible two-detector in situ gamma spectrometer with NaI:Tl and $SrI_2:Eu^{2+}$ scintillators in an experimental lake, the activity concentrations of dissolved natural radionuclides and the minimally detectable activities of ^{137}Cs were estimated.

Keywords: autonomous underwater gamma-ray spectrometer, marine radioactivity monitoring, detector calibration, scintillation response, Monte Carlo simulations.

Сравнение подводных in situ гамма-спектрометров со сцинтилляторами NaI:Tl и $SrI_2:Eu^{2+}$

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На основе экспериментов в термокамере уточнены температурные модели для шкалы энергии детектора, связывающие положение фотопика в спектре с энергией регистрируемого гамма-кванта в зависимости от температуры детекторов и высокого напряжения ФЭУ. Эффективность детектирования определяется по ранее измеренным спектрам сертифицированных водных растворов радионуклидов в резервуаре с водой: полученные экспериментальные значения сравниваются с теоретическими результатами моделирования детекторов методом Монте-Карло. На основании анализа спектров, полученных в результате полевых испытаний погружного двухдетекторного *in situ* гамма-спектрометра со сцинтилляторами NaI:Tl и $SrI_2:Eu^2$ в опытном водоёме, оценены объемные активности растворенных естественных радионуклидов и минимально детектируемые активности ^{137}Cs.

Ключевые слова: автономный подводный гамма-спектрометр, радиационный мониторинг водной среды, калибровка детектора, сцинтилляционный отклик, моделирование Монте-Карло.

Introduction

Underwater in situ gamma spectrometers are designed to measure the activity concentration of dissolved radionuclides directly at the dive site. NaI:Tl-scintillator based detectors are the most common choice for in situ gamma-radiation spectroscopy in water environment since they combine good detection efficiency, high light yield, stable performance, wide operating temperature range and low price [1, 2]. Due to the relatively low energy resolution of NaI:Tl detectors which leads to a possible overlap of

close spectral lines, the development of in-situ detectors based on scintillation crystals with better resolution has become topical in recent years. The $SrI_2:Eu^{2+}$ material has been found to possess exceptional scintillation properties including extremely high light yield, excellent energy resolution and proportional response [3]. Their disadvantages include high cost and not fully developed manufacturing process. In this work, a comparative study of submersible in situ NaI:Tl and $SrI_2:Eu^{2+}$ gamma spectrometers is performed, and, in particular, an analysis of joint calibration experiments and numerical modeling of the detector response using the Monte Carlo method are carried out.

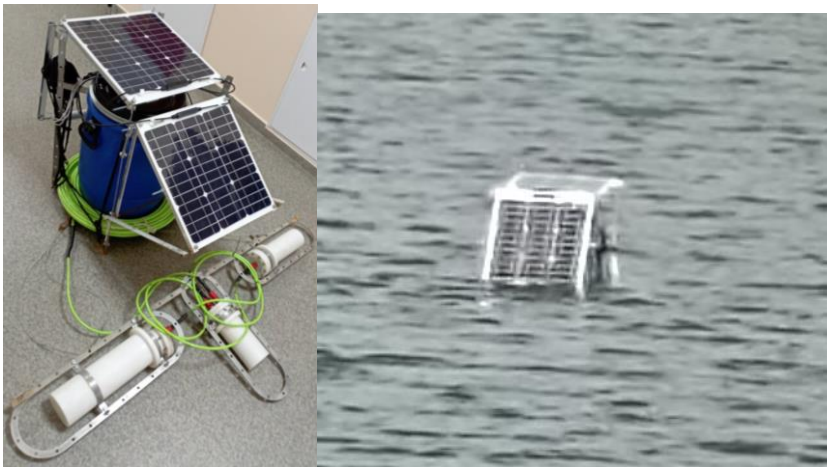

Fig. 1. The two-detector gamma-ray spectrometer (left panel), and the installed gamma spectrometer in an experimental lake (right panel)

1. Results

Comparative characterisation of two detectors based on 3" × 3" NaI:Tl crystals and of two detectors based on 1,5" × 1,5" $SrI_2:Eu^{2+}$ crystals with different resolution is carried out, as well as field tests of a two-detector submersible in-situ gamma-ray spectrometer with NaI:Tl and $SrI_2:Eu^{2+}$ scintillation detectors. Hereafter, the NaI:Tl detector studied in this paper will be referred to as "NaI-2" and the better resolution NaI:Tl detector from [2] studied earlier as "NaI-1". The names "SrI2-1" and "SrI2-2" will refer to the worse and better resolution $SrI_2:Eu^{2+}$ detectors, respectively. Figure 1 shows a photo of the two-detector gamma-ray spectrometer (a surface module: a buoy with solar panels, a battery and transmitting electronics; and a submersible module: two detector units and a microcomputer control unit), and a photo of the installed gamma spectrometer in an experimental lake. The gamma-ray spectra obtained in this field trials are presented in fig. 2. The following calibrations are required for the operation of radionuclide identification programs: 1) calibration of the energy scale of the detector (the dependence of the position of the photopeak in the spectral histogram on the energy of the absorbed gamma quantum); 2) calibration of the energy resolution of the detector (dependence of the width of the detected line on the energy); 3) calibration of the detection efficiency. These calibrations are discussed below. The main difference between the spectrum of fresh water (detector "NaI-2") from the experimental lake (fig. 2, left panel) compared to the similar spectrum of sea water from [2] (detector "NaI-1") is a two orders of magnitude lower activity concentration ^{40}K:

$n_a[^{40}K]$ = 0.48 Bq/L, which is associated with a significantly lower salinity. Analysis of the spectra in fig. 2 gives the following levels of daily minimum detectable activity of the ^{137}Cs radionuclide for each of the detectors and experimental conditions: MDA_{24}[NaI-1, Sea] = 0.039 Bq/L, MDA_{24}[NaI-2, Lake] = 0.030 Bq/L and MDA_{24}[SrI2-1, Lake] = 0.12Bq/L.

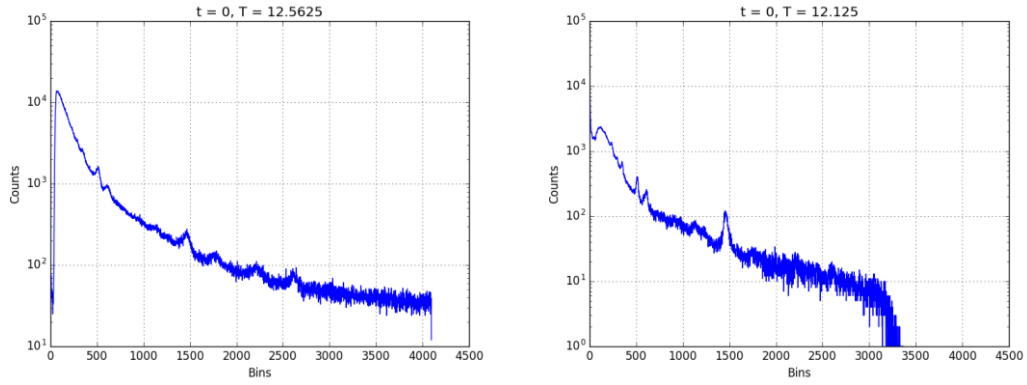

Fig. 2. The gamma-ray spectra for "NaI-2" (left panel) and for "SrI2-1" (right panel) detectors in the experimental lake

The first calibration experiment is measurements of the calibration gamma spectra of ^{152}Eu radionuclide with the gamma spectrometers placed in a temperature chamber in the temperature range from –10 °C to +50 °C both at a constant high voltage, as well as with the active high voltage control stabilizing the position of the photopeaks. While the temperature is slowly changing, the accumulated spectra are saved every 10 minutes, in addition, several time responses of the scintillator to gamma-quantum detection events are additionally saved. To obtain energy scale calibrations, the following set of well-resolved ^{152}Eu reference photopeaks (with gamma quanta energies E_i) is used: E_i = [121.8, 344.3, 1408, 964.1, 778.9, 244.7] keV. The brightness of the scintillation response changes with changes in the temperature of the crystal, which leads to a shift of photopeaks in the spectral histogram of the detector. To compensate for this shift, one approach we use is to adjust the high voltage (HV) of the PMT based on software feedback so that the position (P_i) of the selected photopeak of the calibration source (here the ^{152}Eu photopeak with an energy of 1408 keV) remains constant [4] (fig. 3). Another possibility – similar experiments in a thermal chamber for three detectors with constant high voltage – is also discussed in detail in [4]. Given the single-exponential approximation of the scintillator time response, it is easy to obtain the following more sophisticated temperature model, which relates the temperature dependence of the linear gain coefficient of the detector energy scale calibration $a'_1(T)$ to the temperature dependence of the decay times (the scintillation pulse shape factor) $\tau_1(T)$, the integration time (the digital processing parameter of the multichannel analyzer) t_{int}, and high voltage of the photomultiplier HV(T):

$$a'_1(T) = \phi_a(T)\tau_1(T)\left(1-\exp\left(-t_{int}/\tau_1(T)\right)\right)\mathrm{HV}(T)^n$$

where the coefficient $a'_1(T)$ is from: $P_i(T) = a'_0(T) + a'_1(T)\, E_i + ...$, and the temperature dependences $a'_1(T)$ and $\tau_1(T)$ (as well as HV(T)) are represented by fitting polynomials obtained based on the analysis of the above experiments.

The energy resolution was determined from experiments with radionuclides ^{139}Ce, ^{137}Cs, ^{40}K, ^{88}Y, ^{241}Am (59,5keV), ^{208}Tl (2614keV) by measuring the Gaussian widths of the corresponding photopeaks. The resolution of each detector on the ^{137}Cs line, $R = W_E/E$ (W_E – the width in keV) is as follows: R[^{137}Cs, NaI-2] = 7,5 %, R[^{137}Cs, SrI2-1] = 4,2 %, R[^{137}Cs, SrI2-2] = 3,0 %, and for comparison R[^{137}Cs, NaI-1] = 6,5 %.

To obtain the absolute value of (volumetric) detection efficiency, verified solutions of radionuclides ^{139}Ce (166 keV), ^{137}Cs (662 keV), ^{40}K (1461 keV) and ^{88}Y (898 keV, 1836 keV) with a known activity concentration are sequentially poured into a water tank with a volume of 8m^3. For each detector, based on calculated areas of photopeaks, the absolute values of the detection efficiency for the energies of gamma quanta corresponding to these photopeaks are estimated: $\varepsilon_{exp} = N / (n_a\, I_\gamma\, t_{acc})$, where ε_{exp} is the detection efficiency, n_a is the activity concentration, N is the photopeak area, t_{acc} is the accumulation time, I_γ is the emission probability. The detection efficiency determined from these spectra for the energies of gamma rays emitted by reference radionuclides, as well as a fitting curve ($\varepsilon_{fit}(E)$ – the efficiency versus the energy) are presented in [4]. The detection efficiency for the "SrI2-1" detector turn out to be two times lower compared to the efficiency of the "SrI2-2" detector.

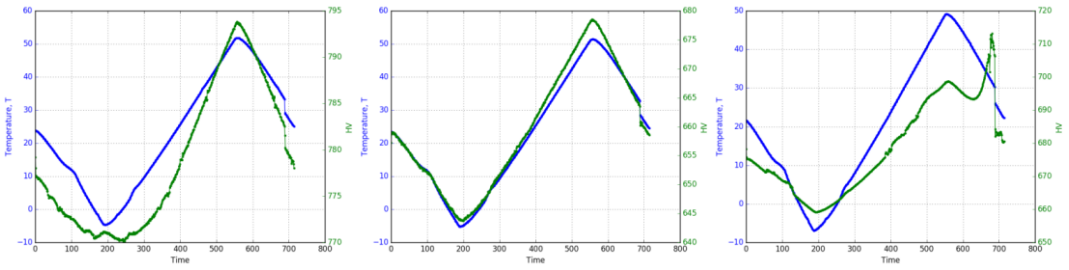

Fig. 3. Thermal chamber nonlinear gain control. Temperature (T) and high voltage (HV) vs time for detectors: "NaI-2" (left), "SrI2-1" (middle), and "SrI2-2" (right). The time unit is 10 min

The obtained experimental efficiency values are compared with the theoretical results of Monte Carlo (MC) simulation using the free software "Geant4", taking into account the materials and geometry of the detectors, as well as the geometry of the water tank. Monte Carlo simulation models include cylindrical 3" × 3" NaI:Tl or 1,5" × 1,5" SrI_2:Eu^{2+} scintillation crystals coated with a PTFE reflector on the bottom and side surfaces and placed in copper or aluminum containers, cylindrical glass and aluminum layers for PMT and MCA, mu-metal magnetic shields, and cylindrical POM external cases with a wall thickness of 10 mm. Gamma quanta with a certain energy and random initial directions and positions are generated uniformly throughout the entire volume of the water tank with a detector immersed in it. Convolution of the spectra with the detector instrument function (with Gaussian of resolution calibration) was not applied at this stage. The detection efficiency determined from MC simulations (blue dots) and experiments (red stars) as a function of gamma-ray energy is given in the fig. 4 for NaI (left panel) and SrI_2 (right panel) detectors.

The following natural radionuclides (and their photopeaks) were identified in the obtained gamma spectra: e^+e^- annihilation [510.8 keV], ^{40}K[1460.8 keV], ^{208}Tl[2614.5 keV], ^{214}Bi[609.3, 1764.5, 1120.3, 2204.2, 934.1 keV], ^{214}Pb[295.2, 351.9 keV], ^{212}Pb[238.6 keV], ^{228}Ac[794.9 keV]. In particular, the estimated activity concentration of the dissolved radionuclide ^{40}K was $n_a[^{40}\text{K}] = 0.48(0.02)$ Bq/l (based on measurements on the "NaI-2" detector). Activity concentrations of other natural radionuclides: $n_a[^{208}\text{Tl}] = 0.024(0.003)$ Bq/l, $n_a[^{214}\text{Bi}] = 0.13(0.01)$ Bq/l, $n_a[^{214}\text{Pb}] = 0.17(0.02)$ Bq/l, $n_a[^{212}\text{Pb}] = 0.075(0.003)$ Bq/l.

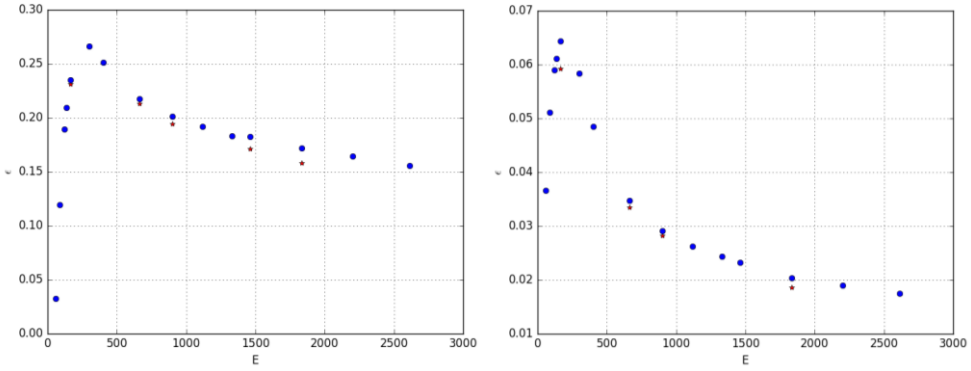

Fig. 4. The detection efficiency determined from MC simulations (blue dots) and experiments (red stars) versus the energy for NaI (left) and SrI_2 (right) detectors

Conclusion

A comparative study of submersible in situ NaI:Tl and $SrI_2:Eu^{2+}$ gamma spectrometers was performed. In particular, an analysis of joint calibration experiments, field tests in an experimental lake, and numerical modeling of the detector response using the Monte Carlo method were carried out. Based on the analysis of the obtained spectra and detector calibrations, the activity concentrations of dissolved radionuclides, the intrinsic detector's activity for ^{40}K radionuclide, and the minimally detectable activities of ^{137}Cs were estimated. Volumetric detection efficiencies of NaI:Tl and $SrI_2:Eu^{2+}$ detectors were obtained both experimentally and based on Monte Carlo simulations.

Acknowledgments

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