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Development and deployment of an autonomous sensor for the in-situ radioactivity measurement in the marine environment

Yingying Zhang*, Bingwei Wu, Dongyan Liu, Ying Zhang, Yan Cheng

Institute of Oceanographic Instrumentation, Qilu University of Technology (Shandong Academy of Sciences), Shandong Provincial Key Laboratory of Ocean Environmental Monitoring Technology, National Engineering and Technological Research Center of Marine Monitoring Equipment, No 7 Miaoling Road, 266061 Qingdao, China

HIGHLIGHTS

- An autonomous sensor for marine radioactive monitoring is given.
- Calibration of energy, resolution and efficiency are analyzed.
- Monte Carlo simulation and new electronic design are presented.
- Field measurements in Qingdao offshore are discussed.
- Minimum detectable activity is compared with similar systems.

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ABSTRACT

An autonomous sensor for the in-situ radioactivity measurement is developed , based on NaI(Tl) crystal, to detect the activity concentrations of natural and anthropogenic radionuclides in seawater. Its electronics module, integral enclosure and spectrum analysis were presented. The energy, resolution and efficiency were calibrated. Monte Carlo simulation and the minimum detection estimations were realized. The sensor was tested in the water tank and then successfully deployed for the continuous monitoring in the marine environment in Qingdao offshore for the performance test, background measurement and environmental survey. Some results were deduced from the gamma ray spectra and discussed in comparison with those from the laboratory and literatures.

1. Introduction

Due to their superior performance of high detection efficiency for gamma rays and wide temperature ranges, less power and lower cost, NaI(Tl) scintillation crystal detector has always been the common method for in situ radioactivity measurements. IAEA-MEL used both high-efficiency NaI(Tl) and high-resolution HPGe spectrometry to investigate contamination with anthropogenic radionuclides in a variety of marine environments (Povinec et al., 1996; Osvath and Povinec, 2001). An underwater system “RADAM” which comprises a 3″ × 3″ NaI(Tl) ruggedized crystal with 7% resolution and a 512-channel analyzer was used in many marine applications like continuous measurements with buoy operation (Osvath et al., 2005; Tsabaris, 2008; Vlachos and Tsabaris, 2005). The limit of detection for ^{137}Cs in water is 19 Bq m^{-3} for a 24-h spectrum integration time. A detection system named “KATERINA” using NaI(Tl) as a crystal was developed and applied for acquiring radon daughters (^{214}Pb and ^{214}Bi) in a region where submarine groundwater discharge exists in the coastal zone (Tsabaris et al., 2008). The marine volume efficiency for ^{137}Cs is 1.8×10^{-4} cps (Bq m^{-3})$^{-1}$ and for ^{40}K is 1.32×10^{-4} cps (Bq m^{-3})$^{-1}$.

The research and applications of NaI(Tl) scintillation crystal detector in the marine environment have again aroused people's concern after the Fukushima accident (Ahmadi et al., 2011; Caffrey et al., 2012; Dulai et al., 2016; Kalfas et al., 2016; Sartini et al., 2011; Tsabaris and Prospathopoulos, 2011; Zeng et al., 2013; Zhang et al., 2015). A lot of effort has been made during the last years in simulating the measured spectra, improving the nuclide detection and system performance. The marine detection efficiency of “KATERINA” has been updated as 2.1×10^{-4} cps (Bq m^{-3})$^{-1}$ for ^{137}Cs in Bagatelas et al. (2010).

In China, the radioactivity monitoring of the marine environment has been using the method of in situ sampling and then analyzing in the laboratory, which always takes two or three days to get the quantitative results through the complicated handling procedures. There is a need to develop an autonomous sensor for the in situ, continuous and long-term

* Corresponding author.
E-mail address: triciayyz@163.com (Y. Zhang).

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monitoring of radioactivity in the seawater.

A lot of effort has been made during the last years in developing an autonomous sensor for the in-situ radioactivity measurement in the marine environment. The methods and results of our research are described in this paper. A Na(Tl) detector and the digital multi-channel pulse signal amplitude analysis was used in the new sensor design. The sensor's packaging technology and materials and the automatic spectrum analysis were also discussed. The autonomous sensor has been energy calibrated with the standard sources and also efficiency calibrated using the Monte Carlo simulation approach. In order to test the accuracy and reliability of the measurement of the sensor developed, the water tank experiments and some field measurements at sea had been already conducted.

2. Materials and methods

2.1. Sensor description

The autonomous sensor new developed for the in-situ radioactivity measurement in the marine environment is composed of a Na(Tl) scintillation crystal detector, the power supply, the modules of digital multi-channel pulse signal amplitude analysis, communication and power supply, and the integral enclosure and software. As shown in Fig. 1, the detector, consisting of a 3 in. × 3 in. NaI(Tl) crystal (customized in China with the energy resolution less than 7.0% at 662 keV) connected with a photomultiplier with low potassium (HAMAMATSU CR109) and a pre-amplification circuit board, is packaged in a watertight housing. The NaI(Tl) crystal emits fluoresce when exposed to gamma rays in the seawater, which is collected by the photomultiplier and then converted into the impulse voltage. Some optically coupled materials such as the silicone oil had been used to improve the photoelectric conversion efficiency. The weak voltage signal, proportional to the energy of the gamma ray, is amplified and shaped and then analyzed according to its amplitude. The gamma spectrum of the seawater is thus generated. The autonomous sensor new developed is connected via CAN bus communication protocol to a PC with the software for on-line data acquisition. The software also provides the data storage, display, processing and some parameters setting of the sensor operation. Some nuclides in the seawater need to be identified and then analyzed in quantity. The power supply provides the high voltage (400–500 V) for the photomultiplier and some different amplitude of voltages (1.5 V, 3.3 V and 5 V) for other electronic devices in the sensor. The specifications of the sensor are given in Table 1.

2.2. Digital multi-channel pulse amplitude analysis

The design method of a Field Programmable Gate Array (FPGA) in combination with a high speed Analog to Digital Converter (ADC) is used for data acquisition and processing in the autonomous sensor. In Fig. 2, the Micro programmed Control Unit (MCU) is used for accepting and executing demands from the upper computer, and also for regulating the high voltage and recording the spectrum. The 12-bits ADC, with high speed and high precision, is in charge of sampling the preprocessed data from the detector and then converting them to the digital data under the control of FPGA. Based on double buffers for data process and transmission in FPGA, a series of operations including the high voltage regulation, gain adjustment, pulse acquisition, pulse

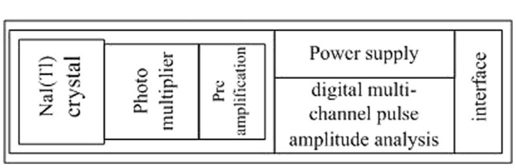

Fig. 1. Components in autonomous sensor.

Table 1
The specifications of autonomous sensor new developed.

Sensor type	3″x3" NaI(Tl)
Energy Range	Adjustable (with maximum value of 3000 keV)
Energy Resolution (661.6 keV)	< 7%
Minimum detectable activity (661.6 keV)	~ 18 Bq m^{-3} in 24 h
Spectroscopy	1024 channels
Operating temperature	− 5 °C to + 50 °C
Operating voltage	9–18 V DC
Consumption	~ 2.0 W
Communication protocols	RS232, USB, CANBUS
Enclosure	Shape “Cylinder”, Φ115 × 470 mm material Polyamides

processing, amplitude analysis in 1024 channels and communication with the upper computer are all conducted all in real time for the in-situ radioactivity measurement in the marine environment. The pulse processing designed is most complex. Some digital methods such as the trapezoidal filtering, pulse shape discriminating; baseline recovering and pulse separating are used.

2.3. Integral enclosure

The Na(Tl) scintillation crystal detector was aluminum packaged as shown in Fig. 3. Some materials such as silicone oil were used to improve the photoelectric conversion efficiency in the detector. The autonomous sensor new developed was packed in a watertight cylindrical enclosure made of polyamides, which was designed to offer continuous operation of the sensor up to 200 m water depth considering the water proof, the pressure resistance, the anti-corrosion and the minimum gamma ray absorption analyzed by Monte-Carlo simulation. The sensor was integrated enclosured with screw-thread and silicone rubber, and O-rings were designed to improve high sealing performance. A four-core watertight joint was used for the power supply and also for the communication between the sensor and the computer based on CAN bus.

3. Experiments and results

3.1. Energy calibration

The autonomous sensor for the in-situ radioactivity measurement in the marine environment was energy calibrated in the laboratory using five reference radioactive point-sources ^{137}Cs, ^{60}Co, ^{241}Am, ^{152}Eu and ^{133}Ba (similar works in Tsabaris et al., 2008). The point sources were placed a set distance of 25 cm from the sensor. As shown in Fig. 4, the energy calibration becomes with the use of linear function as follows corresponding to 95% confidence level:

$$E = a + b \times ch \tag{1}$$

where E is the energy of the specific gamma ray (in keV), ch is the number of channel, a, and b are both fitted parameters. This calibration has been integrated into the spectrum analysis to process data and identify the nuclide. The energy calibration factor b and the offset a are not varied during the sensor operation. The standard deviation of the fitting is 0.9999, so the linear function of the energy of gamma rays and the channel address is very well.

The energy resolution calibration was measured to specify the baseline of the full width at half maximum (*FWHM*, in keV) at various energies at the same time. As shown in Fig. 5, corresponding to 95% confidence level, the energy resolution as a function of the gamma ray energy is:

$$FWHM = a + b \times \sqrt{c \times E + d \times E^2} \tag{2}$$

where the parameters a, b, c and d are all obtained experimentally by