



An in-situ gamma-ray spectrometer for the deep ocean

C. Tsabaris*, E.G. Androutakaki, S. Alexakis, D.L. Patiris

Hellenic Centre for Marine Research, Institute of Oceanography, P.O. Box 712, GR-19013 Anavyssos, Greece



HIGHLIGHTS

- The KATERINA system is upgraded for quantitative measurements at the deep ocean.
- MC simulations using MCNP5 reproduced experimental energy spectra and efficiency.

ARTICLE INFO

Keywords:

Subsea detectors
Marine radioactivity
Monte Carlo simulations
Deep ocean

ABSTRACT

A low resolution subsea gamma-ray spectrometer is developed for monitoring radioactivity in the deep ocean. The detection system provides quantitative results combining experimental and simulated data. The system is tested for pressures up to 456×10^5 Pa and is demonstrated in the deep sea South of Crete (Greece) at a depth of 2700 m. The minimum detectable activity for ^{214}Bi considering a 7200 s measurement, was found 0.3 Bq/L due to the extremely low background contribution at high energies.

1. Introduction

The sustainable protection and management of the ocean require a comprehensive understanding of the processes and conditions that affect the status of the marine environment. At the same time, observing and understanding changes, hazards and opportunities in the ocean is crucial to guide international actions, optimize governments' policies and industrial strategies. This understanding is strongly dependent on observations and monitoring of key parameters at a wide range of space- and time-scales, such as radioactive gases and fluids. The activity concentration of natural radionuclides varies considerably due to natural processes arising from the atmosphere (e.g. rainfall) and/or from the ocean floor (e.g. gas/fluid emanation). Furthermore, the anthropogenic radionuclides are present in the sea due to nuclear reactors, accidents and waste management plants.

Deep Ocean is a repository of valuable new knowledge on unexplored scientific phenomena, natural hazards as well as energy and sources opportunities. Due to the gap of long-term observation and technologically advanced systems, the research and exploitation of deep sea environments is still in immature and undeveloped stage. The knowledge concerning geophysical processes in seismic faults, volcanoes, sediment slides, benthic storms, hydrothermal vents and phenomena related to climate change such as the emanation of greenhouse gases from the oceanic crust is still limited and scarce. Furthermore, the measurement of natural radioactivity has been used in a qualitative and

a quantitative manner in mineral exploration (natural radioactivity in mineral exploration and processing (De Meijer et al., 1997)). One of the natural emanating gases from the ocean floor is radon, which its daughters (^{214}Bi , ^{214}Pb) are identified via the detection of the corresponding emitted gamma-rays. Radon is a colorless and non-chemically reactive gas found in the earth crust and it is continuously formed during the radioactive decay of uranium (^{238}U), which is contained within the earth's substrata.

Cesium isotopes are monitored in the water column, due to their applications as radiotracers, even though in many cases, they exhibit low concentrations (Tsabaris et al., 2015). Especially in areas affected from past nuclear accidents (Chernobyl in 1986 and Fukushima in 2011), buried nuclear waste (Gwynn et al., 2016) and nuclear weapons tests (carried out in the 1950s and 1960s) Cesium concentrations should be monitored also at the deep part of the ocean. Recently, a lot of effort has been made to quantify Cesium isotopes at the sediment of the ocean floor close to Fukushima area at depths up to 500 m (Thornton et al., 2013).

In situ detection systems for long-term measurements of marine radioactivity are scarce due to the requirements for their applicability (e.g. low power consumption, robust and tolerant in high pressures). Low resolution crystals (such as NaI (Tl)) are the most common crystals for long term measurements due to their low cost. Such crystals are used the last years in many marine applications like continuous measurements with buoy operation (Aakenes, 1995; Soukissian et al., 1999;

* Correspondence to: Hellenic Centre for Marine Research, P.O. Box 712, GR-19013 Anavyssos, Greece.

E-mail address: tsabaris@hcmr.gr (C. Tsabaris).

<https://doi.org/10.1016/j.apradiso.2018.08.024>

Received 15 March 2018; Received in revised form 29 June 2018; Accepted 30 August 2018

Available online 10 September 2018

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Wedekind et al., 1999; Tsabaris and Ballas, 2005; Osvath et al., 2005; Li et al., 2016), seabed mapping (Maučec et al., 2004; Osvath and Povinec, 2001) and in situ measurements of gamma ray emitters in seawater (Povinec et al., 1996; van Put et al., 2004; Tsabaris et al., 2005a; Vlastou et al., 2006; Tsabaris et al., 2008; Bagatelas et al., 2010; Caffrey et al., 2012). A lot of effort has been also made the last years for quantitative measurement of radionuclides present in the sediment using low resolution crystal in a mobile or stationary mode (Androulakaki et al., 2016a, 2015; Kofuji, 2015; Thornton et al., 2013). HpGe detectors were also used in aquatic environments and on the seabed (Povinec et al., 1996), but the cooling of the crystal and the high power consumption of commercial detectors could not make them applicable for long-term measurements in the marine environment. The sediment radionuclide mapping was first realised by the IAEA group at Monaco using in-situ methods (Osvath and Povinec, 2001). Such applications took place at several marine areas with radiological interest (Povinec et al., 1999; Povinec et al., 1997; Osvath and Povinec, 2001).

In addition, a lot of effort has been made for developing software for the simulation of the response function of low resolution detection systems in order to provide an efficient method of measurement. Various methods have been developed (Vlastou et al., 2006; Vlachos and Tsabaris, 2005; Bagatelas et al., 2010; Androulakaki et al., 2016b) for the simulation of NaI(Tl) spectrometers performance, taking into account the photon interactions in the sediment or seawater (including also interactions with the crystal and the enclosure material). As concerns the spectra analysis using the KATERINA system (Tsabaris et al., 2008) a new software package SPECTRW (Kalfas et al., 2016) has been developed for peak de-convolution. Wavelet practices (Tsabaris and Prospathopoulos, 2011) and a FSA technique (Androulakaki et al., 2016b) have been also developed for analysis of short-time acquisition spectra.

The background values for radon at the seawater are very low. However, deployments of gamma-ray spectrometers at the deep sea are requested mainly for monitoring the radon progenies that may be present in areas where gas/fluids emissions take place at the ocean floor (submarine springs, mud volcanoes, cold seeps, hydrothermal vents). Such detection systems are applied the last years mainly for monitoring radon progenies at submarine groundwater discharges (Povinec et al., 2001, 2006a, 2008, 2006b; Tsabaris et al., 2010, 2012). The detection limit of the DUS gamma-ray spectrometer (Povinec et al., 2006b) for radon measurements at deep seawater masses is reported ~ 0.05 Bq/L while the minimum detectable activity for the radon progeny measurements using KATERINA at the shallow water masses was ~ 0.03 Bq/L. Furthermore, the background values of ^{40}K are almost constant and easily measured using a conductivity meter. However, combined measurements of ^{40}K using gamma-ray spectrometers and conductivity meters may provide significant information for the total (including particulate) and dissolved potassium concentrations in the aforementioned areas.

At surface waters the values of radon progenies are enhanced mainly due to rainfalls and can reach activity concentration values up to 1.3 Bq/m³ (Tsabaris and Ballas, 2005; Tsabaris et al., 2008). At deep waters the activity concentration of radon is enhanced (up to 5.3 Bq/m³) in areas where submarine water discharges or thermal springs are present (Povinec et al., 2001; Povinec et al., 2006; Povinec et al., 2008; Tsabaris et al., 2012). The baseline information for radon gas concentration at the deep sea is very poor. Past results demonstrated that the ^{222}Rn concentration using lab methods from various hydrothermal vents is 50–200 times greater than the ^{222}Rn concentration in ambient rise crest waters (Dymond et al., 1983). These concentrations can be detected by in-situ subsea gamma-ray spectrometers. Nevertheless, the baseline information for the deep ocean (and especially in the Earth's hadal zones) is very poor.

Current available networks and sensors are dedicated for operations at intermediate sea levels, limiting their application for routine operational monitoring and especially in the deep ocean. The last years very

few documented works are made for radioactivity monitoring at the deep ocean (Li et al., 2016; Zhang et al., 2015; Thornton et al., 2013; Sartini et al., 2011).

In the present work, a new autonomous system named “KATERINA-D” is presented, which can be applied for radioactivity measurements in a variety of natural marine processes (e.g radioactive gas and fluid emission) or for detection of potential sunk objects at the ocean floor. The detection system was tested for stability and linearity and was calibrated for measurements in the deep ocean. The system was deployed in an area close to south of Crete (Greece).

2. Materials and methods

2.1. Detection system

The system “KATERINA-D” consists of a $3'' \times 3''$ NaI detection crystal, connected with a photomultiplier tube, preamplifier, amplifier and power supply, together with a multichannel analyzer for data acquisition and storage. The electronic modules are especially constructed to fit inside the detector housing (85×600 mm) and the power consumption is low (~ 0.8 W) and continuously controlled. The stability of the gain calibration is checked and restored using the photopeak at 1461 keV of ^{40}K . The specifications of the subsea detection system are given in Table 1.

A watertight cylindrical enclosure has been designed consisting of stainless steel, which houses the above-mentioned NaI crystal together with the appropriate digital electronics and modules. The enclosure was designed to offer continuous operation up to 4500 m water depth. The selection criteria for the appropriate enclosure material were based on minimizing gamma ray absorption and maximizing the pressure tolerance. The following candidate materials were studied: borosilicate glass, Titanium, carbon fiber and stainless steel. Acetal was also considered as an enclosure material for deployments up to the intermediate water masses. The optimal wall thickness of the cylindrical enclosure was calculated using Roark's thick wall formulas according to a previous study (Tsabaris and Thanos, 2004). In order to take into account the creeping behavior of the material, additional calculations were performed using a commercial software package for the housing design (Tsabaris and Thanos, 2004). Two radial and one axial Buna N (nitrite) O-rings were used to achieve safe sealing despite the frequent re-opening of the detection system.

The calculation of O-rings' parameters (diameter, thickness, groove dimensions, maximum gap) was performed, following the suggestions of the Seal Design Guide from Apple Rubber Products Inc. The unit (detector + photomultiplier) is mounted on a support (sled). The sled is furthermore, permanently fixed with bolts, on the enclosure's cup. Two

Table 1

The specifications of the detection system KATERINA-D.

Sensor type	$3'' \times 3''$ NaI(Tl)
Energy Range	Adjustable maximum and minimum energy of detection (with maximum value of 3000 keV)
ADC	Successive approximation 10 bit
High Voltage	Internally controlled 300 – 1200 V
Spectroscopy	Adjustable 256 , 512 or 1024 , 2048 , 4096 channels
Amplification: Pole Zero Cancellation, Base Line Restoration	Adjustable externally using a software
Energy Resolution (140.5 keV)	10%
Energy Resolution (661.6 keV)	7%
Operating temperature	-5°C to $+50^\circ\text{C}$
Consumption	0.7 – 0.8 W
Output	Time, date, cps, spectrum, dead time.
Communication protocols	RS232, USB, Ethernet
Enclosure	Shape “Cylinder”, material Stainless Steel

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