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C. TSABARIS, I. THANOS

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C. TSABARIS [1] and I. THANOS [1]

[1] Hellenic Centre for Marine Research,
P.O. Box 712, 19013 Anavissos, Greece

e-mail tsabaris@ncmr.gr

Abstract

We describe a set up and an application for an autonomously working, radioactivity sensing instrument, usable in seawater and river environments. The system is based on a NaI scintillator with the appropriate specifications for use in the marine environment and for real time acquisition. It is simple, stable for long-term monitoring, and of low consumption. Many tests were carried out for the linearity and the stability of the electronics. The investigation of energy resolution and energy calibration of the sensor was performed in the laboratory using various reference point radioactive sources. The system was also deployed in a water tank in order to measure background radiation in the water and low volumetric activity of ^{137}Cs (17 Bq/m^3). Appropriate software identifies qualitatively the low level ^{137}Cs contribution to the measured γ-ray spectrum.

Keywords: Radioactivity; Underwater sensors; Monitoring systems; Warning systems.

Introduction

The development of an autonomous measuring system for radioactivity in the marine environment is today of important scientific priority for the marine sciences and especially for the Operational Oceanography (FLEMMING, N. C., 1995).

A state of the art development of the entire sensor and buoy platform should include:

- Innovative sensor selection
- High reliability and longevity of the measuring apparatus
- On line data transmission
- Long interval periods between two successive maintenances

The radiometric techniques are based on the measurement of the gamma ray emitters (radionuclides) as those have been produced from natural constitutes as well as from anthropogenic radionuclide activities. The development of instruments for measuring radioactivity in the marine environment was focused in the past mainly on the improvement of the sensor construction and to the suitable techniques for the identification of various radiations.

The application of the Real-Time Gamma Spectroscopy method has many difficulties due

to the set up of the whole system (sensor and communication) and to the poor energy resolution of the systems with low consumption. On the other hand this method has a lot of advantages compared to the widely used method (FLOROU, H. *et al.*, 1994 and PAPUCCI, C. *et al.* 1999) of sampling and performing the measurement with a good resolution static system (off line method). The requirements of a good monitoring system are low power consumption, low cost, high efficiency, good energy resolution and robust construction for longevity. These requirements are satisfied by a NaI scintillator, apart from its energy resolution, which is about 20 times lower compared to a HpGe detector (POVINEC, P. P. *et al.*, 1995 and OSVATH, I. *et al.*, 2001).

HPGe detectors have been used for such measurements with a maximum measuring time interval of 2 hours, due to the overheating of the crystal. Using cryogenic sensors this problem can be solved, but the power consumption of such a system is very high (~ 500W). It cannot be used in the field without a proper power supply. Recently, the problem of the power consumption seemed to be solved by a Japanese group (KOBAYASHI, Y. *et al.*, 1998), with a HPGe system. However its efficiency (about 22%) combined with its considerable weight (~ 45 kg), makes the system inconvenient for operational use.

On the contrary, the NaI crystals have high efficiency, good stability for long runs and low cost. This type of sensor can thus be attached to an oceanographic floating system for monitoring and alarm purposes. The floater can operate under severe weather conditions and especially under extreme winds and waves. The only disadvantage of the system is the energy resolution (7% at 662keV), which is relatively low compared to the Germanium detectors, so that the various contributions of gamma rays may overlap and deform the measured gamma-ray spectrum.

Materials and Methods

A new system for underwater measurements and continuous monitoring has been developed for acquiring radioactivity produced from gamma ray emitters in the energy range from threshold until 3000 keV. The system consists of a 3"x3" NaI detection crystal, with a built in photomultiplier tube and preamplifier. A separate power supply, together with an electronics card for data acquisition and storage is used. The power consumption of the whole system is relatively low (~ 2W). The influence of the operating temperature (between 0 and +50˚C) to the gain shift of the detector is compensated automatically.

A watertight cylindrical casing has been designed, which houses the above mentioned sensor unit. The following specifications were defined for the casing, prior to design:

- Capability to offer free of error continuous sensor functionality up to 100 meter water depth. (Testing depth of 200 meter).
- Corresponding plug in watertight cabling system for real time data transmission. Redundant power transmission to/from the sea surface.
- A lifespan for the casing in the deep sea of at least 5 years.
- Minimum gamma ray absorption through the casing.

In order to investigate the total absorption of the gamma rays into the material, a systematic study has been carried out for five candidate materials for the casing (Al, Fe, Acetal, POM and PVC). In the requested energetic region, acetal has the lowest value of the total mass attenuation coefficient multiplied by the density of the material (see Fig. 1). The thermoplastic material 'Acetal' (commercially known as Celcon, Delrin etc.) was finally chosen. The material is machineable and has an excellent resistance in the sea environment, as well as good mechanical strength properties. The wall thickness of the

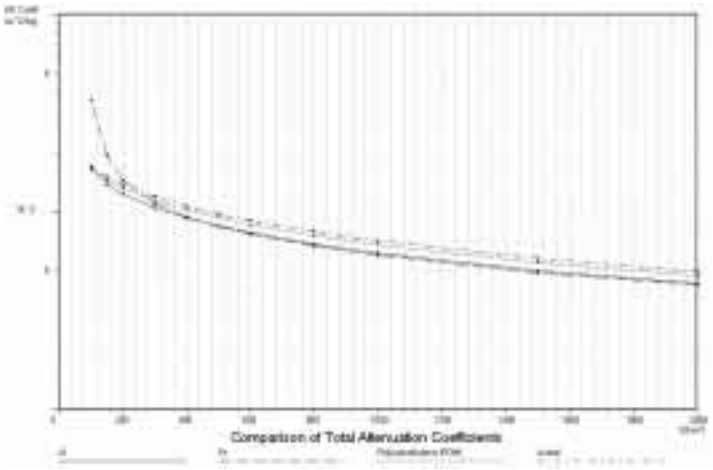

Fig. 1: Total attenuation coefficients for five materials (Al, Fe, Acetal, POM, PVC).

cylindrical casing was calculated using Roark's thick wall formulas (YOUNG, W.C., 1989). In order to take into account the creeping behavior of the thermoplastic material, additional calculations were performed using the approved 'Under Pressure' housing design software package from Deep-Sea Company (Under Pressure 2001). A conservative working strength of 0.9 Ksi was assumed for Acetal. Similar calculations were performed for enclosure's cup and the bottom geometry. Two radial and one axial Buna N (nitrile) O-rings were used in order to achieve a very good sealing mechanism despite the frequent reopening of the casing (at least during the testing phase).

The calculation of O-rings' parameters (diameter, thickness, material, hardness, groove dimensions, maximum gap etc.) was performed, following the suggestions of the Seal Design Guide from Apple Rubber Products Inc. (APPLE 1989) The enclosure's 3D artistic picture can be seen in Figure 2. For easy mounting of the sensorial, the unit (sensor

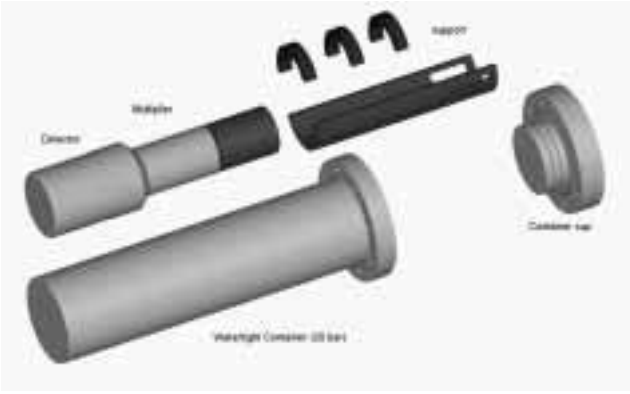

Fig. 2: The watertight cylindrical casing, which houses the sensor, the photomultiplier and the appropriate sled.

+ photomultiplier) is mounted on a support (sled). Furthermore, the sled is permanently fixed with bolts, on the casing's cup. A watertight connector- and cabling system was designed according to the specifications mentioned above. The wiring diagram is shown in Figure 3. The custom-designed cabling system was then ordered for construction by an UK Company. It is functional up to 700 meters of water depth.

In order to use this system for continuous monitoring, the sensor has been energy calibrated and tested for its stability to temperature variations and its energy resolution. Particularly, using five reference sources (emission of eight gamma rays), the energy and energy resolution calibration in the air environment has been performed. In addition, underwater measurements of the detector efficiency and absolute calibration have also been performed, by using a calibration tank of 5.5m^3 volume, filled with water. The sensor was mounted in the middle of the tank in order to be surrounded by one meter of water, which is enough to imitate the real marine environment for energies less than 700 keV, due to the high attenuation of the γ-rays in the water. An electric pump was placed at the bottom of the tank in order to circulate the water to avoid sedimentation, to mix the water with the appropriate reference radionuclide (^{137}Cs) and to get homogenous conditions.

Then the system will be placed on a floating measuring system and with the appropriate facilities, the measured data will be transmitted to the operational centre (HCMR). The communication can be achieved via Satellite and/or mobile telephony network (NITTIS, K. *et al.,* 2000).

Results and Discussion

The sensor was first energy calibrated (out of the tank) by using reference point sources and then it was placed in the tank filled with drinking water. Background spectra were systematically collected in order to investigate variation of the natural radioactivity and test for temperature stability. A typical spectrum is presented in Figure 4, where the photopeaks from the decay series of ^{238}U are clearly seen. A detailed study has been carried out for the gas loss of ^{222}Rn (mother nucleus of the measured daughter nuclides ^{214}Pb and ^{214}Bi), which causes reduction of the background

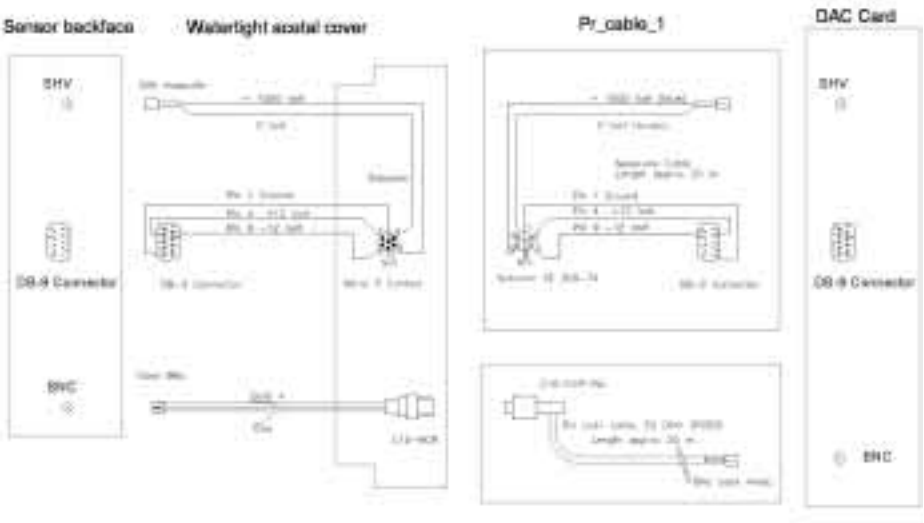

Fig. 3: Cabling system between the Detector housing and the sea surface apparatus..

counts during the first month (the half life is 3.825 days).

The γ-rays used to perform energy calibration of the system were the 1461 keV of ^{40}K with an elemental abundance 0.012% in natural potassium and the 2615 keV of ^{208}Tl. The linearity of the system was checked by the emission of other contributions, like ^{214}Bi and ^{214}Pb. The used electronics offer a very low non linear amplification coefficient of the order 10^{-9}. After the first month the volumetric activity of ^{222}Rn was stable in the tank and several spectra were recorded and found to be identical, indicating also the homogeneity of the water. Then a low concentration of ^{137}Cs, 17Bq/m^3 was mixed in the water with 65% HNO_3 0.005N. One of the acquired spectra is presented in Figure 5 (the measuring time was 1 day). The 662keV transition of ^{137}Cs is sitting on the high background produced both by the natural background and the Compton scattering of the 1461 keV γ-ray of ^{40}K and the other high energy photopeaks.

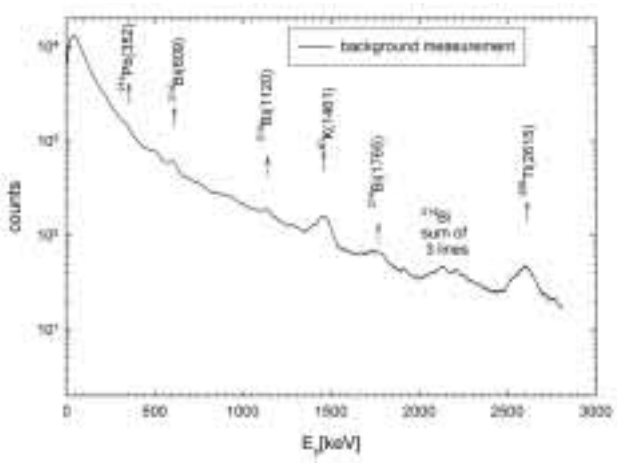

Fig. 4: Background spectrum as acquired with the developed system (1 day). The energy and origin of the individual photopeaks are indicated in the figure.

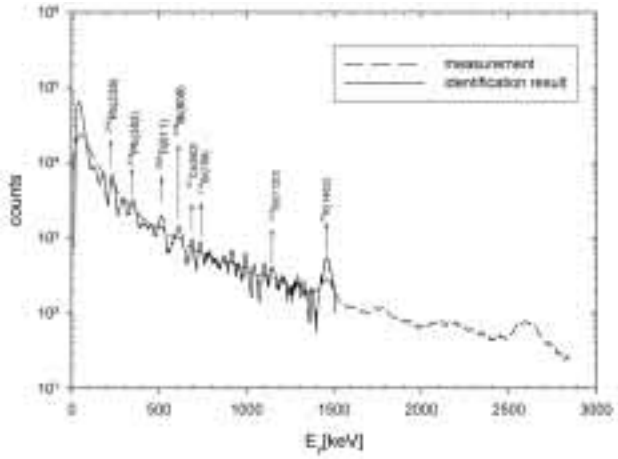

Fig. 5: Gamma ray spectrum recorded in the laboratory (1 day) by mixing ^{137}Cs in drinking water (dashed line). The solid line represents the identification result of the various radioisotope contributions.

According to the technique described in literature (BRUTSCHER, J., *et al.*, 2001), suitable software was developed for this application in order to identify qualitatively the various gamma ray contributions in the spectrum. This algorithm is very effective, when the statistics are low and the peak shape poor. The algorithm results are shown in Fig. 5 within the energy range from threshold to 1500 keV (due to low statistics of the measurement at higher energies the calculated curve is not plotted above 1500 keV). The system is optimized for ^{137}Cs, the characteristic peak of which can be clearly seen at 662 keV in Figure 5. Further measurements, in combination with 'Monte Carlo' simulation, will lead to a more precise estimation of the particular peaks. Specifically, the quantitative estimation of various contributions should be the next step of this investigation.

Conclusions

A new apparatus, sensing radioactivity for use in underwater marine environment, is developed aiming in the near future, its integration on a real-time data-forwarding sea surface buoy. This conforms to the further idea of building an operational and low cost buoy network for monitoring the radioactivity levels in open sea. The device has been under continuous testing, during the last three months, immersed in a large water tank. Due to the quality of stabilization of the electronic unit, no voltage drifts were observed during this measuring period. Frequent energy calibrations were indeed performed, to confirm this stability.

The developed hardware and software for underwater gamma ray spectrometry has been tested and calibrated in the laboratory for its stability and its sensitivity for ^{137}Cs. The device could observe clearly the given input of 17Bq/m^3 of ^{137}Cs concentration in the water, within a period of one day. This observation is enhanced qualitatively by running the developed software. The derived sensitivity (17Bq/m^3 of ^{137}Cs within one day) is much higher compared with a similar system (RADAM III) used in the POSEIDON network (TSABARIS, C. *et al.,* 2002).

The system will be installed in future on a floating measuring system for the surveillance and monitoring of gamma radiation in the marine environment. In addition, such a system could be used as an early alarm system for possible water pollution and natural gamma ray emitters.

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