Development of on-Line Monitoring Device for Marine Radioactive Pollution

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Abstract: On-site measurements are indispensable to swiftly and precisely evaluate the magnitude of radioactive contamination triggered by a nuclear accident or terrorist attack in the ocean. Consequently, an on-line monitoring device for marine radioactive pollution has been developed. A plastic scintillator was used as the detector of the device, and a pump was used to extract the seawater around the measuring region, ultimately enabling continuous on-line measurement. The device measures sea water for 10 minutes, and the minimum detectable activities of ^{137}Cs and ^{60}Co are 1.7 Bq/L and 2.1 Bq/L, respectively. It can promptly and efficiently the detect events of excessive radioactivity in wastewater and give an alarm signal. The device will be critical for monitoring radioactive pollution in the maritime region around nuclear power plants, and it has the benefits of a quick measuring time, a high monitoring frequency, and a high degree of real-time monitoring, allowing for the early identification of accidents and the implementation of emergency strategies.

1 INTRODUCTION

Developing nuclear power is an excellent technique to alleviate environmental problems and optimize the energy structure. In the process of nuclear power development, the safety of nuclear reactors is very important. Once a nuclear leakage accident occurs, it will have a great impact on the environment (Arvela et al. 1990; Lee et al. 2017; Fereshteh et al. 2021). In 2011, a serious nuclear accident occurred at the Fukushima nuclear power plant in Japan, resulting in the release of huge quantities of radionuclide into the environment (Kinohita et al. 2011; Nakano and Povinec, 2012). As a nearby territory, China immediately carried out a vast number of radioactive monitoring projects. Traditional marine radioactivity monitoring consists of collecting samples of seawater, biological organisms, and sediments from critical sites 1 to 4 times per year, which are then transported back to the appropriate laboratory for processing and analysis (Vlastou et al. 2006; Tsabaris 2008). Using γ-ray spectrometer to measure samples in laboratory is a time consuming method, which requires chemical pretreatment of the measured samples. This non real-time and discontinuous method is difficult to effectively monitor the marine radioactive pollution, and it is even more difficult to realize the early warning of marine radioactive pollution.

On April 13th, 2021, the Japanese government officially agreed to discharge sewage from the Fukushima Daiichi Nuclear Power Plant into the ocean at a cabinet meeting that day. Since then, Japan intends to discharge 1.25 million tons of nuclear sewage into the sea, with the goal of starting to discharge in two years. While TEPCO has claimed that after treatment, most of the radionuclides in the nuclear sewage except ^{3}H can be removed. However, monitoring data reveal that there are radioactive material residues such as ^{14}C, ^{60}Co and ^{90}Sr that are difficult to completely remove in the nuclear sewage after "filtration". β radionuclides, such as ^{3}H, ^{14}C and ^{90}Sr, cannot be monitored online by γ spectrometry, hence the question of how to monitor β radionuclides in the water near nuclear power plants in real-time and precisely has emerged as an essential one. Furthermore, with the development of nuclear power, the construction of real-time monitoring network for radioactivity in sea areas around nuclear power plant has also been put on the agenda. It will be a trend to develop a radiation monitoring network along the whole coast with online monitoring, real-time early

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warning and other functions (Wedekind et al. 1999; Tsabaris 2005; Katsumi and Pavel, 2020).

2 INSTRUMENT DESIGN

The designed online monitoring device for marine radioactive pollution is mainly composed of a detector, a sampling chamber, a lead chamber and signal processing unit (see Figure 1). To complete the continuous measurement and indication of seawater, the device is required to have the following characteristics: 1) high sensitivity is required of the detector; 2) a superior shield layer and fewer background count; 3) cleanliness and anti-fouling should be priorities while designing the sampling chamber; 4) easy to manufacture and reasonable cost; 5) stable, dependable performance over an extended period of time, as well as simple maintenance.

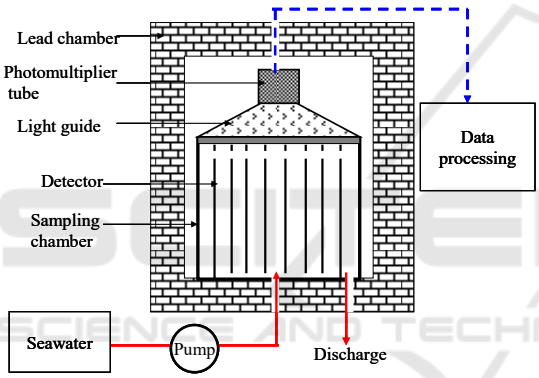

Figure 1: Structure diagram of the online monitoring device for marine radioactive pollution.

2.1 Detector Selection

Plastic scintillator is a kind of organic scintillator that may be used to detect α particles, β particles, γ rays, neutrons and fission fragments (Suffian et al. 2020; Kagami et al. 2020). It is widely used in the measurement of some low-level radioactive or liquid samples. Plastic scintillator has several benefits, including its ease of production, low cost, great mechanical strength, ability to be molded into a variety of forms, stable performance, strong radiation resistance, and high corrosion resistance. Plastic Scintillators are thus perfect for β radionuclide monitoring in the marine environment. To detect radioactivity in seawater in real time, the plastic scintillator is built as a nested mode of circular rings, with the outermost detector separating the sampling chamber into an inner and an outer portion (see Figure 2). Seawater enters from the bottom of the detector, and there are some overflow holes with a diameter of 5 mm on the top of each circle ring detector. When the pumped seawater exceeds the overflow hole, it will gradually fill the whole sampling chamber through the overflow hole, and finally flow out of the sampling chamber through the water outlet.

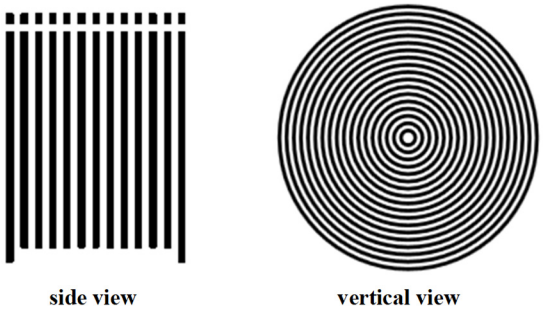

Figure 2: Structure of the plastic scintillator detector.

2.2 Design of Lead Chamber

The concentration of radionuclides in the measured seawater is low most of the time. To reduce the influence of background, the typical low-background lead chamber is used as shield. The designed lead chamber is a cylinder with a size of Φ 40×50 cm, the thickness of lead is 5 cm. Since natural lead materials contain ^{210}Pb and other radionuclides, a 2-mm copper layer was added to the inside of the lead chamber to shield the radiation of radioisotopes of lead.

2.3 Design of Sampling Chamber

The sampling chamber is positioned within the lead chamber, and its size is Φ 25×30 cm. Its main function is to continuously collect seawater into the sampling chamber. The sampling vessel is composed of ABS plastic that is resistant to impact, heat, and low temperatures. The bottom is equipped with an input and an outlet for water, enabling the continuous collection and discharge of seawater. The inner wall of the sampling chamber (light-transmitting at the top) is covered with aluminum foil to isolate the interference of the outside world to the detector and the photomultiplier tube, and reflect the fluorescence produced by the interaction of radiation with the detector back to the photomultiplier tubes.

2.4 Data Processing Unit

The data processing unit consists of signal discrimination, filtering shaping, and counting display, which is mainly used to process the nuclear signal output from the detector and display the results.

Firstly, the nuclear signals are filtered by signal discrimination, then it is turned into a Gaussian waveform and noise interference is minimized. In the counting display module, the nuclear signals are classified and counted after filtering, and the result are displayed.

3 TECHNICAL INDICATORS OF THE INSTRUMENT

3.1 Minimum Detectable Activity

One of the key performance indicators of the measuring equipment is the minimum detectable activity (MDA). It refers to the smallest amount of radioactivity in a sample or medium that may be detected with a particular degree of confidence (Choi et al. 2019). In general, the smaller the MDA in the same time, the better the detection performance of the instrument. However, MDA characterizes the qualitative analysis ability of the system for the existence of nuclides, which cannot characterize the quantitative measurement accuracy of the system for nuclide activity. At the 95% confidence level, MDA can be expressed as:

$$MDA = \frac{2.71 + 4.65\sqrt{B}}{t \cdot \varepsilon} \quad (1)$$

where B is the background count; t is the live time of counting; ε is the detection efficiency. The change trend of MDA of ^{137}Cs and ^{60}Co in seawater with measurement time by this device is shown in Figure 3.

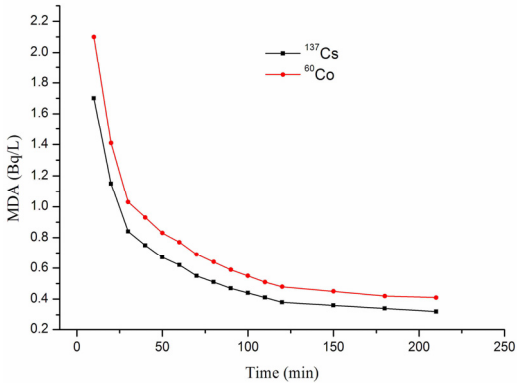

Figure 3: The change trend of MDA of ^{137}Cs and ^{60}Co in seawater with measurement time by the designed device.

The seawater containing ^{137}Cs and ^{60}Co with an activity concentration of 5 Bq/L were measured respectively in the laboratory, and the technical indicators when the device was used as the "activity detection" function were obtained: the MDA of ^{137}Cs and ^{60}Co were 1.7 Bq/L and 2.1 Bq/L respectively after 10 minutes of measurement. For the seawater containing ^{137}Cs and ^{60}Co with an activity concentration of 5 Bq/L, the statistical uncertainty of measurement for 10 minutes is less than 10%.

3.2 Monitoring Performance of the Instrument

The MDA test results indicate that the device is capable of detecting β radionuclide contamination events in a short time frame. As a real-time monitoring device, the measurement results must be provided continuously in a short time. The shorter the measurement interval, the more precisely the data may be represented throughout time. In the case of real-time monitoring, after deducting the background, the count of the device is less and obeys the Poisson distribution. The false alarm rate and the missing alarm rate of the monitoring equipment are the two most crucial and contradictory technical indicators. According to the calibration results of the detection efficiency of the device, based on the Poisson distribution law, we have formulated the exceeding threshold and compiled the real-time monitoring program.

The measured results show that when the activity concentration of ^{137}Cs is 5 Bq/L, the total count rate of the monitoring device is 2.7 cps, or an average of 162 counts per minute. This means that the mathematical expectation for the device to count in two minutes is 324. According to Poisson distribution, the corresponding probability density was calculated with 324 as the expected value. For setting different exceeding threshold y (the alarm will be given if the counts detected in any 2 minutes is $\geq y$), "probability of reaching the threshold" is equal to the curve area on the right side of the medium threshold.

Set the monitoring cycle of the designed device to 2 minutes. When the count of two adjacent cycles is greater than or equal to the exceeding threshold, the device will automatically give an alarm signal. By setting the threshold value in this way, the number of false alarms per year can be controlled below 0.1. For radionuclides with activity concentration greater than 5 Bq/L in seawater, the missing alarm rate is less than 1% after 20 minutes of measurement.

To test the performance of the device, seawater containing ^{137}Cs with an activity concentration of 5 Bq/L was measured and monitored continuously for 100 cycles. The monitoring results were analyzed, and it was found that 60 cycles sent out alarm signals,

with an average delay of 4.52 minutes and a maximum delay of 7 minutes. Count 100 cycles, and the results are shown in Figure 4. It can be seen from Figure 4 that the count obtained by the device is in good agreement with the Poisson distribution, and it is effective to use the Poisson distribution law to design the alarm program.

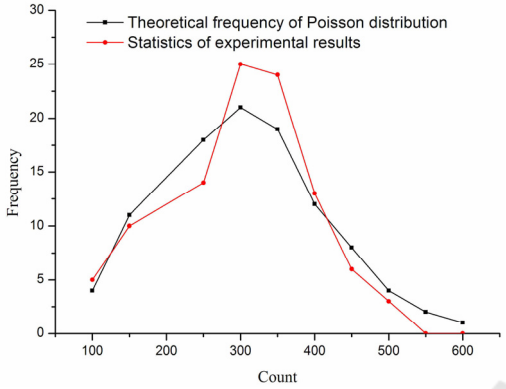

Figure 4: Comparison of counting statistics of 100 cycles with the theoretical Value of Poisson distribution.

4 CONCLUSIONS

The designed online monitoring device for radioactive contamination in seawater has a high degree of sensitivity, can provide information on radioactive contamination in seawater in a short period of time, and has the function of rapid measurement of radioactive activity concentration in seawater. In the "detection" aspect, the plastic scintillators with a low price, stable performance, good radiation resistance and corrosion resistance. Combined with the 14.7 L sampling chamber in the lead chamber, excellent MDA was obtained by reducing the background. Taking ^{137}Cs as an example, MDA measured for 10 minutes can reach 1.7 Bq/L.

As for the "Alarm" feature, we have adopted the optimized design of inlet and outlet waterways, so that the seawater in the sampling chamber can be updated quickly. We have also developed a set of "short time and less counting" design method based on Poisson distribution to determine the alarm threshold and minimize the false alarm rate and missing alarm rate within a reasonable and acceptable detection time. For seawater containing ^{137}Cs with an activity concentration of 5 Bq/L, it takes only 4.52 minutes on average to give an alarm, and the probability of missing an alarm after 20 minutes of monitoring is less than 1%.

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