Calibration Method and Experiment of in-Situ Radioactivity Measurement in Seawater Based on Monte Carlo Simulation

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Abstract. Some research on the in-situ radioactivity measurement in seawater and development of underwater spectrometer using NaI(Tl) scintillation crystal are being carried out for the automatic and continuous monitoring in the marine environment. The gamma spectrometer must be efficiency calibrated for the detection of radionuclides in the seawater. For the problem of traditional experiments, a simulation model of in-situ radioactivity measurement in the seawater was established by using Monte Carlo statistical method. The model simulates the in-situ measurement environment of seawater and the characteristics of the underwater spectrometer developed by ourselves. By calculating the interaction between gamma-ray photons emitted by radionuclides in seawater and various atoms in seawater and the internal structures of the spectrometer, the marine detection efficiency of the underwater spectrometer developed was calculated and verified by the field experiments. The results show that this kind of non-experimental calibration method is effective and feasible for the underwater spectrometer and further research and experiments will continue.

1. Introduction

The radioactivity measurement in seawater has been using the method of in-situ sampling and returning to the laboratory for processing and analysis. Using the traditional method, it is impossible to monitor the marine radioactivity environment effectively, and even more difficult to supply the pollution warning timely. It is necessary to carry out research on the in-situ monitoring technology of marine radioactivity, which can guide sampling, and provide the important technical support for the continuous radioactivity monitoring in the marine environment, as well as efficient response to the emergency accidents.

The spectrometer using NaI(Tl) scintillation crystal is always the main method of studying the in-situ radioactivity monitoring in the marine environment, because of high detection efficiency, wider temperature range, stable performance, low power consumption and low cost. The Fukushima accident has once again attracted the attention of international maritime countries to this technology, and related research is still continuing [1-8].

The efficiency calibration of the spectrometer is very important for the quantitative determination of radionuclides in the seawater. But it is very difficult for the traditional method using some standard sources because few radionuclides can be dissolved in the seawater, and more importantly, the large
volumes of polluted seawater. Therefore, this paper studies a kind of non-experimental method for the efficiency calibration and also carries out the experiment to test the method.

2. Materials and method

2.1. Simulation model

The simulation model of in-situ radioactivity measurement in seawater is mainly focused on the geometric structure of the spectrometer and the description of the marine environment, which is used for calculation of the interaction between gamma ray and atoms in seawater and the absorption of gamma ray in various structures in the spectrometer. A simulation model of NaI (Tl) underwater spectrometer was established by using Monte Carlo software according to the structure, material and size of the instrument developed by ourselves. As shown in figure 1, the 7.5cm x 7.5cm Na (Tl) crystal is connected to the photomultiplier tube. The photomultiplier tube is connected with preamplifier, high voltage and multi-channel processing circuit in turn. The spectrometer is encapsulated in a polyamide shell with a thickness of 0.6 cm.

![Figure 1. Structure diagram of spectrometer](image)

The in-situ measurement model is shown in figure 2 based on the composition and density of the seawater in the off-shore area of Qingdao. Taking the crystal as the spherical center, the in-situ measurement model is modelled as an infinite source of seawater with natural and artificial radionuclides evenly distributed. Before gamma photons enter the spectrometer, the interaction between gamma photons and various atoms in the seawater is calculated in the model. The gamma photons may change energy due to Compton scattering or electron response, or may disappear due to the photoelectric effect. After gamma photons enter the spectrometer, the model mainly calculates the photoelectric response, Compton scattering and electron pair effect of photons and atoms in the crystal, and also the attenuation of gamma photons in various geometric structures of the spectrometer.

![Figure 2. Measurement model illustrated in seawater](image)

2.2. Calculation and analysis method
The gamma photons near the spectrometer contribute more counting to the measurement in the seawater. According to the some research results, the underwater spectrometer using Na(Tl) crystal has different effective detection distances for different radionuclides in the seawater, so corresponding to the different effective detection sources in the model.

The radionuclides follow the different decay laws of gamma rays in the seawater, so the marine detection efficiency of the certain radionuclide is related to the volume of seawater and the energy of the radionuclide. Assuming that the radionuclides of known activity is evenly distributed in the source of seawater, with the radius \( r = 10, 20, \ldots \), the energy deposition of photons and their secondary electrons in the spectrometer is recorded by using the pulse height counting \( f_8 \) in the simulation model. The pulse height generated by the energy deposition was then calculated as the detection efficiency of the particle in the specific channel. The radionuclides calculated and gamma-ray characteristics are shown in Table 1. The pulse height generated by the energy deposition is taken as the detection efficiency of the spectrometer for particles. We define that the intensity of gamma rays in the seawater decreases to 0.1% of the initial intensity, and the corresponding detection distance is the marine detection distance. As shown in figure 3, the marine detector distance increases with the energy of the gamma rays [6]. To calculate the marine detection efficiency by simulation method, in situ measurement in the seawater is simulated only considering the gamma rays equally distributed within the marine detection distance in the seawater. Define \( V \) is the marine detection volume. Using \( E_v = E \cdot V \) as the volume detection efficiency of seawater, the saturated volume detection efficiency of the underwater spectrometer for the different radionuclides in seawater is calculated and then the efficiency calibration curve is fitted. The minimum detectable activity \( MDA \) of the spectrometer for the radionuclides in seawater (in 95% confidence level) is calculated by the following formula:

\[
MDA = \frac{2.71 + 4.65 \sqrt{B}}{\epsilon_r \cdot I_r \cdot t}
\]

where \( t \) is the measuring time, \( I_r \) is the branching ratio of the radionuclides and \( B \) is the background count of the photopeak.

| Radionuclide | Energy (keV) | \( I_r \) (%) | Radionuclide | Energy (keV) | \( I_r \) (%) |
|-------------|-------------|--------------|-------------|-------------|--------------|
| \(^{109}\)Cd | 88          | 3.7          | \(^{214}\)Bi | 1764        | 15.8         |
| \(^{57}\)Co | 122         | 85.5         | \(^{137}\)Cs | 661.6       | 85           |
| \(^{99m}\)Te | 140.5       | 89.1         | \(^{54}\)Mn  | 834.8       | 100          |
| \(^{214}\)Pb | 295         | 19           | \(^{65}\)Zn  | 1115.5      | 51           |
|              | 352         | 37           | \(^{60}\)Co  | 1173.2      | 100          |
| \(^{131}\)I | 364.5       | 81           |             | 1332.5      | 100          |
| \(^{22}\)Na | 511         | 180          | \(^{40}\)K   | 1461        | 11           |
|              | 1274.5      | 100          |             |             |              |
Figure 3. Marine detector distance of the underwater spectrometer developed for the different gamma rays energy

3. Calibration result and experiment

Based on the detection efficiency of the different radionuclide in the seawater simulated, the marine detection efficiency curve for the underwater spectrometer developed using Na(Tl) crystal is acquired and shown in Fig.4. The data points in the figure are the simulated values of the marine detection efficiency for the different gamma ray energies. The simulation errors of little than plus or minus three percent correspond to statistical uncertainty. The solid line represents a fitting curve of the simulated values and can be described in the empirical expression:

$$\varepsilon_{\nu} = \frac{a \times E^b}{c + E^d}$$  \hspace{1cm} (2)

where $E$ is the specific energy of the certain radionuclide in keV, and $a$, $b$, $c$ and $d$ are all fitted parameters. The resulting error (2-3%) was included in the total uncertainty determination of the efficiency by using the fitting curve.

Figure 4. Marine detection efficiency curve of underwater spectrometer developed
To verify the marine detection efficiency calculated by simulation, some field experiments have been carried out by using the underwater spectrometer developed. Before the in situ measurement experiments in the seawater, the underwater spectrometer developed has been energy calibrated by using some radioactive standard sources such as $^{137}$Cs, $^{60}$Co, $^{40}$K and $^{54}$Mn in the laboratory. The background spectrum of seawater obtained by continuous cumulative measurement for 24 hours is shown in figure 5. Some seawater was collected and brought back to the laboratory. The activity of $^{40}$K in the seawater sample was measured of $1.078 \times 10^4$ Bq m$^{-3}$ [7]. The marine detection efficiency of the underwater spectrometer developed for $^{40}$K can be calculated by the following formula:

$$\varepsilon_v = \frac{cps}{c I}$$

(3)

where $cps$ is the counting rate of the photopeak, $c$ is the activity concentration of the radionuclide in Bq m$^{-3}$. The marine detection efficiency value of $^{40}$K obtained in field experiment is expressed by a star point in Fig.4. It is clear seen that the marine detection efficiency of $^{40}$K from experiment is very near to that from simulation, which showed promising results for the simulation code as well as for the quantitative estimation of radionuclides in seawater.

![Figure 5. Background spectrum of seawater for 24 hours](image)

**4. Conclusion and discussion**

A non-experimental calibration method for underwater spectrometer using Na(Tl) crystal is established by simulating the measurement environment and the detection process of gamma photons in seawater using Monte Carlo statistical calculation method in this paper. The experiment result showed promising results for the quantitative estimation of radionuclides in the seawater. The more quantitative experiments of the radionuclides should be carried out to fully verify the feasibility and accuracy of this kind of non-experimental calibration method. A certain size of seawater tank can be designed and developed, and then the underwater spectrometer is placed in the central position of the tank. However, at present, due to the problem of large-scale contaminated seawater treatment, the specific experiments have not been completed. In the next work, the construction of the standard water tank and some relevant quantitative measurement experiments of radionuclides in the seawater will continue in the next work.

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