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Automated spectra analysis of in situ radioactivity measurements in the marine environment using NaI(Tl) detector

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HIGHLIGHTS

- The detector developed and its calibration is given.
- Simulation method for marine radioactivity measurement is discussed.
- Detection response matrix is established.
- TImproved Richardson-Lucy deconvolution method is proposed for spectrum analysis.
- Continue research method is discussed.

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ABSTRACT

The NaI(Tl) detector has become the main way of research and application of in-situ radioactivity measurement in the marine environment due to many advantages on detection efficiency, power consumption, cost and applicability. But for the poor energy resolution and there is a high background in the low energy region of the spectrum measured mainly originating from the Compton scattering of natural radionuclides such as ^{40}K with high concentrations in seawater, it is difficult and also an interesting topic for NaI(Tl) detector to identificate the radionuclides and calculate them in seawater by analyzing the spectra measured. In this paper, an in situ NaI(Tl) detector developed for the marine environment was energy, resolution and efficiency calibrated. The detection response matrix was calculated by taking all the responsible processes and interactions of gamma rays in water as well as in the detector into account using Monte Carlo simulation method. And then an improved Richardson-Lucy (R-L) deconvolution algorithm was proposed to reconstruct the gamma spectrum measured in the seawater to remove as efficiently as possible the background counts into the corresponding photopeaks. The original spectrum was transformed from poor energy resolution to the corresponding deconvolution spectrum of the high energy resolution with several isolated photopeaks. The experiments of synthetic spectrum and measured spectrum both showed promising results for the radionuclide qualitative and quantitative analysis.

1. Introduction

In situ radioactivity measurement in the marine environment are mainly performed using two detection systems based on the gamma spectrometry method: HPGe and NaI(Tl). Despite its superior resolution, HPGe system exhibits several limitations for autonomous and continuous operation, concerning high power consumption, lower detection efficiency and poor resistance for vibration and swing. The NaI (Tl) detector better suits marine monitoring due to its low consumption, low cost, high efficiency and fair applicability. Various efforts towards the direction of NaI(Tl) detector for the in-situ radioactivity measurement in the marine environment have been published in the last few years especially after the Fukushima nuclear accident, the main point being the better calibration, excellent detection performance and automatic analysis of gamma spectra (Caffrey et al., 2012; Eleftheriou et al., 2013; Sartini et al., 2011; Thornton et al., 2013; Tsabaris and Prospathopoulos, 2011; Zhang et al., 2015). However, for the poor energy resolution, the spectrum measured by NaI(Tl) detector has often

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overlap and interference between the components with similar energy. This makes it difficult to identify the few artificial radionuclides with similar energy such as ^{137}Cs and ^{134}Cs and further to calculate them. There is also a high background in the low energy region of the spectrum measured by NaI(Tl) detector mainly originating from the Compton scattering of natural radionuclides such as ^{40}K with high concentrations in seawater. This makes it difficult to identify the few artificial radionuclides with low concentrations in seawater and further to extract the characteristic parameters for calculation them. The algorithm for automated analysis of in situ NaI(Tl) gamma spectra in the marine environment is very important.

One purpose of the gamma spectrum analysis is to find and identify the radionuclides in seawater. The previous study focused on the spectra smoothing and peak detection (Robin et al., 2011; Yamada and Takano, 2014), which is important but difficult for NaI(Tl) detector because of the poor energy resolution. There are also some limitations in the application of the methods used for resolving overlapped spectra. For example, the standard spectra of many radionuclides have to be measured in the seawater or established before using spectrum stripping method. It is critical but difficult for the fitting method to determine the initial parameters through the experiments.

Another purpose of the gamma spectrum analysis is to determine the activity of radionuclides in seawater. The classic method has developed from counting to function fitting, which is simple and fast, and the main concern is to determine the relative edges and net area (Robin et al., 2011; Tsabaris, 2011; Yamada and Takano, 2014). Some methods such as Fourier transform, SNIP and high order function filtering have been used to eliminate influence of measurement background, which also need to detect the peak and determine the relative edges (Miroslav, 2009; Shi et al., 2018; Zhang et al., 2011). Neural network and other intelligent algorithms have been also used for gamma spectrum analysis, which need a large number of learning samples in the same measurement conditions (Bobin et al., 2016). It is difficult for these methods to be used for the automatic spectra analysis of in situ measurements in the marine environment with a high natural radioactivity background. The inverse matrix method transforms the spectral lines of low energy resolution into high energy resolution lines by solving the equations, but the problem is how to establish the accurate response matrix and how to solve the complex equations (Bare and Tondeur, 2011; Miroslav and Vladislav, 2011). This method is suitable for the quantitative analysis of the gamma spectrum in the seawater, but the energy spectrum of a certain number of single energy photons needed cannot be obtained by experiments as the measurement on the ground. There were already several literatures about the response function of NaI(Tl) detector when it is used in the marine environment in recent years (Bare and Tondeur, 2011; Vlachos and Tsabaris, 2005; Wang et al., 2015). The further involved response matrix of the measurement spectrum in the seawater and the quantitative analysis are still the important research topics.

In this paper, we present the calculation and result of the response matrix of NaI(Tl) detector for in situ radioactivity measurement in the marine environment using Monte Carlo simulation method. Then the spectrum reconstruction from low energy resolution to high energy resolution is studied for better qualitative and quantitative analysis of radionuclides in the seawater. All research and experiments are based on the detector developed by ourselves.

2. Materials and methods

2.1. Detector and its calibration

The in situ NaI(Tl) detector developed for the marine environment, consisting of a 3 in. × 3 in. NaI(Tl) crystal (made in China with the energy resolution less than 7.0% at 662 keV) connected with a photomultiplier (HAMAMATSU CR109) and integrated electronic circuits with density of 3.667 g cm^{-3}, is packaged in a watertight cylindrical case made of polyamides. The detector, as shown in Fig. 1, is designed to offer continuous operation up to 200 m water depth considering the water proof, the pressure resistance, the anti-corrosion and the minimum gamma ray absorption.

Fig. 1. In situ NaI(Tl) detector developed for the marine environment.

The detector was energy calibrated in the laboratory using five reference radioactive point-sources ^{137}Cs, ^{60}Co, ^{241}Am, ^{152}Eu and ^{133}Ba, which were placed in fixed geometry and the distance from the detector. The energy calibration, as shown in Fig. 2, becomes with the use of linear function:

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

where E is the energy of the specific gamma ray (in keV), ch is the number of channel. The fitted parameters a and b are both not varied during the detector operation. The energy resolution as a function of gamma ray energy is depicted in Fig. 3, which follows the equation:

$$FWHM = e + f \times \sqrt{c \times E + d \times E^2} \tag{2}$$

where the parameters c, d, e and f are all obtained experimentally by fitting the $FWHM$ experimental values, which are also not changed during the detector operation.

2.2. Field measurements

The detector has been used for the radioactivity measurement of seawater near Eight Gap Qingdao port for several times. It was suspended by a crane and positioned 3 m below the seawater level in order to eliminate the measurement interferences of the seabed and cosmic radiation, where the seawater is four to eight meters in depth. The detector was connected to a notebook for the continuous measurement and data collection. Fig. 4 is a typical spectrum in the seawater there for a measuring period of 24 h.

2.3. Simulation model

For detection efficiency, response function and response matrix, a simulation model was established using Monte Carlo N-Particle (MCNP) Transport Code based on the NaI(Tl) detector developed. Taking the NaI(Tl) crystal in the detector as the center of the sphere, the field measurement was made into an infinite source of seawater, with the standard composition and with density of 1.025 g cm^{-3}, with uniform distribution of natural and artificial radioactive nuclides. The gamma photons in the seawater were mainly calculated before entering the detector by considering the interactions with the various atoms in the seawater, maybe changing energy due to Compton scattering or

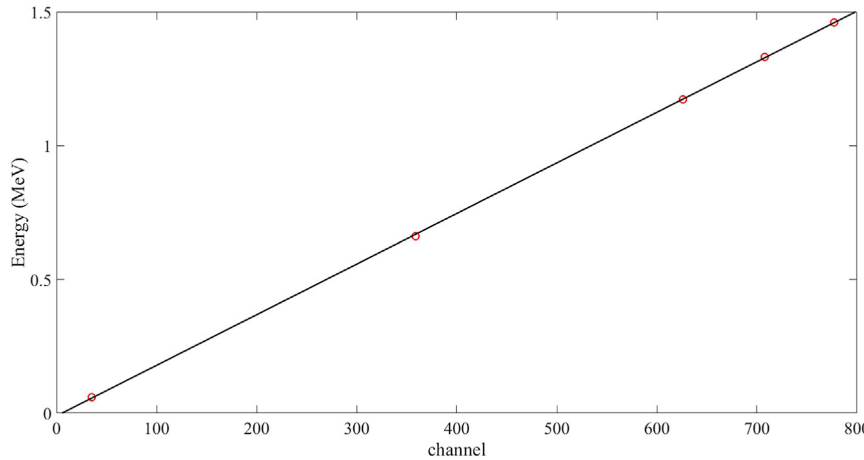

Fig. 2. Energy calibration curve of the detector by means of a set of radioactive point sources. The dots are experimental values. The solid line is a linear fit through the experimental points.

electron pair effect, maybe disappearing because of the photoelectric effect. After entering the detector, the gamma photons were mainly calculated by considering some attenuations due to various metal structures in the detector and some interactions with atoms in the crystal due to the Compton scattering, photoelectric and electron-pair effects happened. To simulate the measurement more really, the attenuation of gamma ray due to the polyamides case around the crystal is considered in the model. The model geometries are shown in Fig. 5. The calculation mode is mode P and the number of particles for the simulation is 10^8.

3. Results and discuss

3.1. Response matrix

To minimize computer time and achieve the best possible resolution and accuracy at the same time, the energy step in the simulation experiments is set to 20 keV to calculate the amount of energy in the range from 0 to 3 MeV. The deposition of the single energy photon in the detector and the distribution of each line in different channels were calculated. The model of in situ radioactivity measurement in the seawater, verified by the detection efficiency (Zhang et al., 2015), was used in the experiments.

Considering the statistical fluctuation in the gamma spectra and some influences such as the electronic noise in the measurement, the line spectra obtained by simulation were Gaussian pulses broadened. The response functions, with the same characteristics as the actual measurement in the sea water, were simulated using MCNP Transport Code by inputting the energy resolution through the GEB card. In order to test the validity of the simulated results, the calculated response function for 1460 keV (^{40}K) was compared with the gamma ray spectrum acquired in the field (Vlachos and Tsabaris, 2005). In Fig. 6, the thick line shows the spectrum measured in the seawater using NaI(Tl) detector developed. The activity of ^{40}K in the seawater of 1.185×10^4

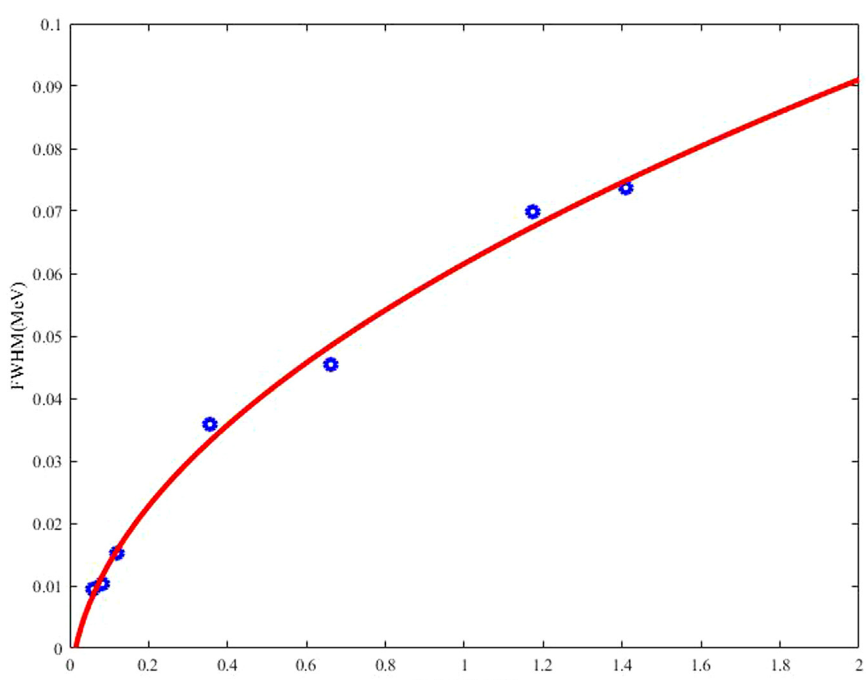

Fig. 3. Energy resolution calibration curve of the detector by means of a set of radioactive point sources. The dots are experimental values. The solid line is a fit through the experimental points.

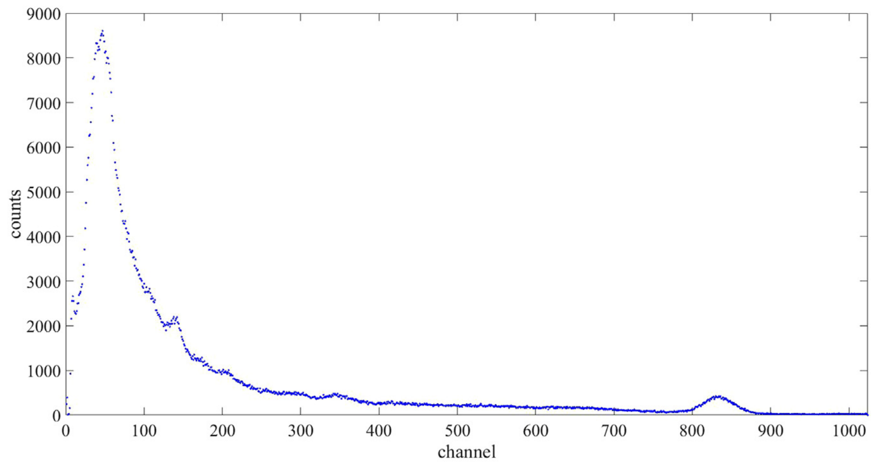

Fig. 4. Spectrum in the seawater for a measuring period of 24 h.

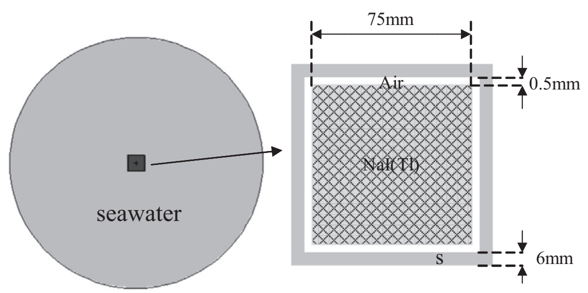

Fig. 5. Measurement simulation model of NaI(Tl) detector immersed in seawater.

Bq m^{-3} can be easily calculated. The thin line shows the simulated spectrum contains only the contribution of ^{40}K in the seawater. It was scaled on the coefficient corresponding to an activity of ^{40}K in the seawater of 1.20×10^4 Bq m^{-3} in order to fit the measured spectrum at the photopeak located at 1460 keV. The agreement in the photopeak of ^{40}K validates the simulation result concerning the quantitative estimation in seawater. Certainly, the measured spectrum contains not only ^{40}K but also other natural radionuclides such as U and Th series present in the seawater. There are higher background counts in the low energy region mainly originating from the Compton scattering of natural radionuclides.

The response functions were arranged on the energy step. The resulting response matrix of the seawater measurement was constructed as shown in Fig. 7. This is a lower triangular matrix, order of which is N. From the part of the response functions in Fig. 7, the components of the gamma spectrum, such as photopeak, Compton edge, backscattering and so on, can be clearly distinguished.

3.2. Spectral unfolding

The spectrum measured in the seawater $y(x)$ can be approximately described as the sum of several Gauss peaks:

$$y(x) = A_1 e^{-\frac{(x-a_1)^2}{2\sigma_1^2}} + A_2 e^{-\frac{(x-a_2)^2}{2\sigma_2^2}} + \ldots\ldots + A_n e^{-\frac{(x-a_n)^2}{2\sigma_n^2}} \quad (3)$$

where a_i is the position of the i-th peak, A_i is the amplitude of the i-th peak. n is the number of the peak, σ is the width of the peak. The measurement in the seawater can be considered as a signal modulation system. The relationship between the measuring spectrum in the seawater and the response function of the detector is expressed as the sum of discrete convolution:

$$\sum_k h(k, j) f(j) = y(k) \quad (4)$$

where $f(j)$ is the spectral distribution of radioactive sources in seawater. $h(k, j)$ is the element in the simulated response matrix H, k is the row number and j is the column number. For such an ill-posed problem, to avoid the large error of the traditional inversion algorithm, the discrete data obtained by the actual measurement can be iterative solved by using R-L algorithm:

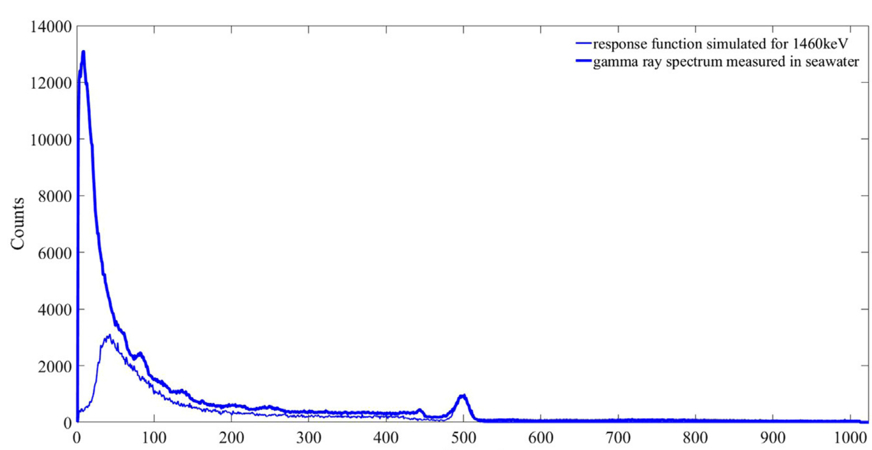

Fig. 6. Simulated spectrum for 1460 keV (^{40}K) gamma ray contribution in the seawater in comparison with the spectrum measured in the field using NaI(Tl) detector developed. The measuring time is 34 h.

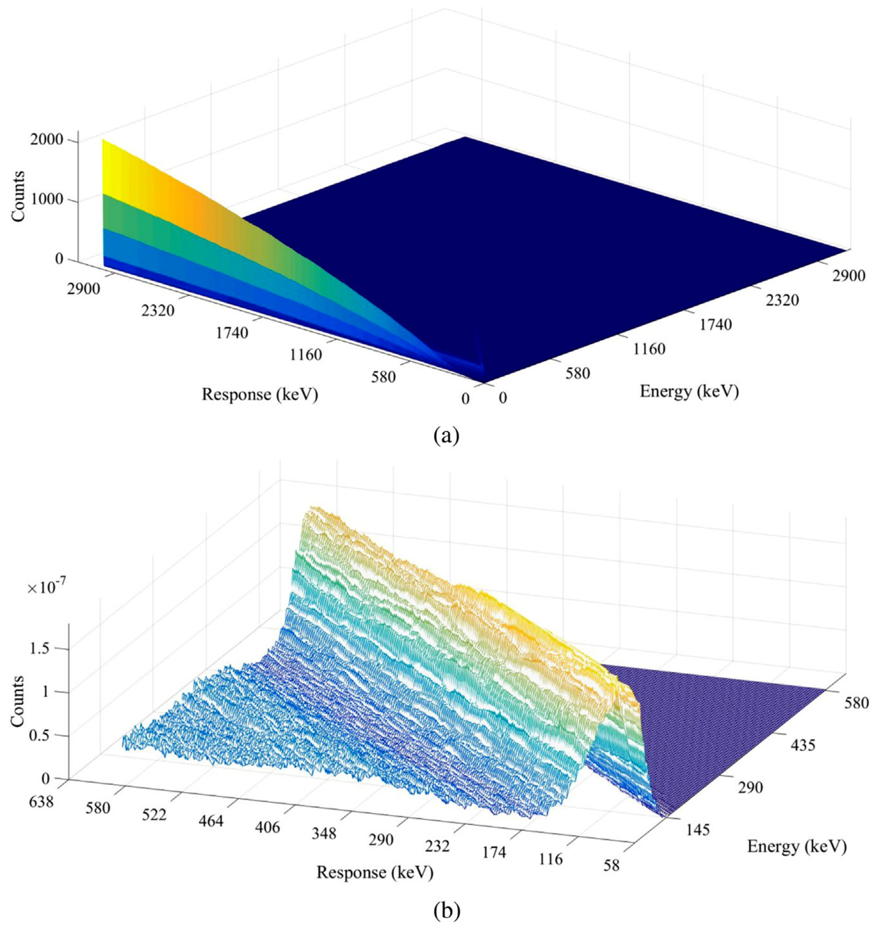

Fig. 7. The simulated response matrix. (a). the whole matrix. (b) the part of simulated response functions.

$$f^{(n+1)}(j) = f^{(n)}(j)\sum_{k=0}^{N-1} h\left(k, j\right)\frac{y(k)}{\sum_{i=0}^{N-1} h(k, i)f^{(n)}(i)}, j = 0, 1, 2, \ldots, N-1 \tag{5}$$

To avoid the premature of the regularized deconvolution R-L algorithm, the spectral lines are iterative reconstructed by introducing the reinforcement factors on the basis of R-L algorithm in order to better simulate the spectra in the in situ measurement. Since the exponent of Gauss function is still a Gauss function, the formula (3) can be changed into the following by introducing the exponent function in each gamma spectrum:

$$y^p(x) = \left(\sum_{i=1}^{n} A_i e^{-\frac{(x-a_i)^2}{2\sigma_i^2}}\right)^p = \sum_{i=1}^{n} A_i{}^p e^{-\frac{(x-a_i)^2}{2\left(\frac{\sigma_i^2}{p}\right)}} + \sum R \tag{6}$$

where $\sum R$ is the cross product of each term. For gamma spectrum analysis, the higher the energy resolution of the detector, the better the radionuclide identification is. It can be seen that, when $p > 1$, the peak width changes to $\sigma/\sqrt{p}$, which is equal to $1/\sqrt{p}$ of the original peak. The ratio of maximum and minimum amplitude is changed from original $A_{\max}/A_{\min}$ to $(A_{\max}/A_{\min})^p$ by introducing the index of p. If the value of p is too large, the smaller peak may be suppressed and then disappear in the deconvolution spectrum. It is certainly not desirable. In the experiment, p is set to the optimal value in the range of [1,2]. There were no false peaks occurred in the reverse modulation spectrum by increasing the number of iterations for $\sum R$ in formula (6).

3.3. Data analysis

For NaI(Tl) detector with the poor resolution, how to detect and identify the possible radionuclides in the seawater, especially the close energy peaks, is one of the important tasks of marine in-situ monitoring. It is also the premise for accurate detection of target nuclide concentration. To test the spectrum analysis algorithm proposed, the following experiments were carried out. The synthetic spectrum was established by extracting the several characteristic spectral lines for ^{137}Cs (661.66 keV)、^{134}Cs (605.795 keV)、^{40}K (1460.8 keV) and ^{60}Co (1173.24 keV and 1332.50 keV) from the response functions and then added in accordance with the attenuation and the different gain. The Gauss noise equivalent to one percent of the maximum value of the spectral line was added. The thin line in the lower part of Fig. 8 is one of the synthetic spectra in the experiment. Because the nuclides of ^{137}Cs and ^{134}Cs have the close photopeak energy, and ^{60}Co has two close photopeaks, so there are some characteristic peaks overlap in the synthetic spectrum. The improved R-L algorithm was applied to the experimental spectrum and then the corresponding deconvolution spectrum was acquired, as shown in Fig. 8 above. It can be seen that the background continuum was very effectively removed from experimental spectrum. Five photopeaks were apparent separated and thus easy identified. The reconstructed spectrum was calculated from the deconvolution spectrum using the convolution algorithm and shown in the lower part of Fig. 8 with the original experimental spectrum for comparison.

To evaluate the spectral unfolding method, the spectrum in the seawater near Eight Gap Qingdao port for a measuring period of 24 h was used for the activity calculation of ^{40}K. The corresponding

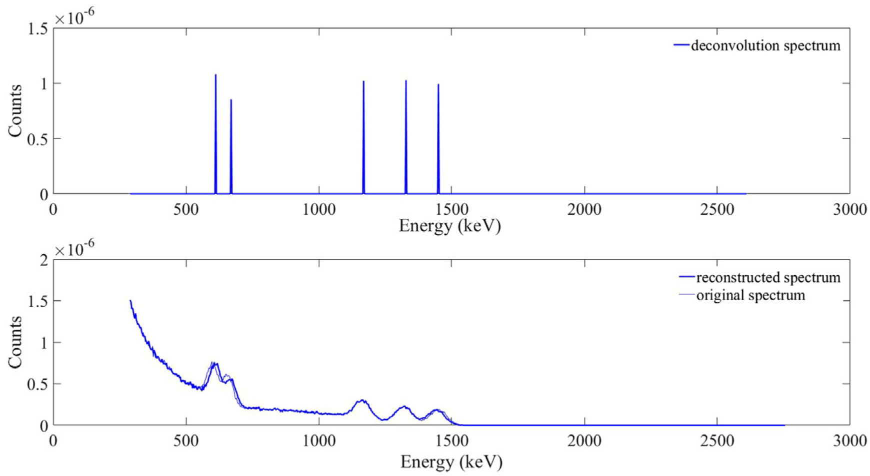

Fig. 8. Effect of the deconvolution method applied to the experimental spectrum for the radionuclide identification. The deconvolution spectrum for better radionuclide identification is shown in the upper part of the figure and below, the before-and-after experimental spectra are shown. Thin and thick lines show the original spectrum and reconstructed spectrum after deconvolution, respectively.

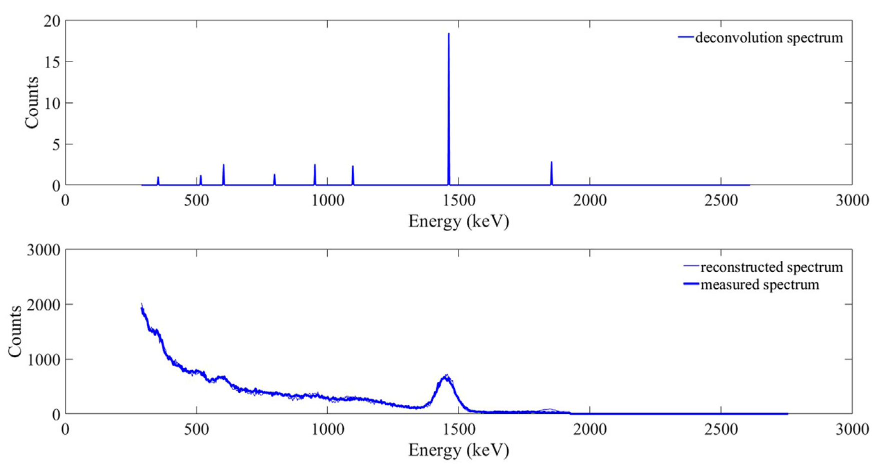

Fig. 9. Effect of the deconvolution method applied to the measured spectrum for the quantitative detection. The deconvolution spectrum for better detection is shown in the upper part of the figure and below, the before-and-after spectra in seawater are shown. Thick and thin lines show the original measured spectrum and reconstructed spectrum after deconvolution, respectively.

deconvolution spectrum is shown in Fig. 9 above obtained by using the improved R-L algorithm. It's easier for the deconvolution spectrum to detect the activity of radionuclides in the seawater because the background continuum was very effectively removed from the measurement spectrum. The marine detection efficiency value of ^{40}K obtained in situ measurement is 1.7×10^{-4} cps $(\mathrm{Bq\,m^{-3}})^{-1}$ for the detector we developed (Zhang et al., 2015). The activity of ^{40}K, *c*, is calculated according to the following equation:

$$c = \frac{N}{\varepsilon I t} \tag{7}$$

where *N* is the number of counts under the peak, ε is the marine detection efficiency (cps $(\mathrm{Bq\,m^{-3}})^{-1}$), *I* is the emission probability, *t* is the acquisition time (in seconds). From the deconvolution spectrum, the activity of ^{40}K was calculated to be $1.26 \times 10^4\ \mathrm{Bq\,m^{-3}}$. The activity of ^{40}K was measured to be $1.185 \times 10^4\ \mathrm{Bq\,m^{-3}}$ in the laboratory by processing and then analyzing the seawater samples collected in the Eight Gap Qingdao port. The error of two measurement methods was about $7.5 \times 10^2\ \mathrm{Bq\,m^{-3}}$ and the relative error is 5.9%. The result of the deconvolution spectrum is acceptable for the quantitative detection in the seawater. The reconstructed spectrum was also calculated from the deconvolution spectrum using the convolution algorithm and shown in the lower part of Fig. 9, which is close to the spectrum measured in the sea water.

4. Conclusions

Due to its good efficiency, wide working temperature ranges, low consumption and low cost, NaI(Tl) detector has been common used and researched as an underwater sensing system for monitoring radioactivity in the marine environment. But in view of the high background spectrum in the low energy region mainly originating from the Compton scattering of natural radionuclides such as ^{40}K with high concentrations in seawater, it is difficult for NaI(Tl) detector, with the poor energy resolution, to identify the possible radionuclides with the close energy photopeaks or several possible radionuclides with little activity in the seawater, let alone the quantitative calculation. The gamma spectrum analysis is seen to be an interesting technique. In this paper, a NaI(Tl) detector developed for the marine environment was energy and resolution calibrated using five reference radioactive point-sources. It was also efficiency calibrated using Monte Carlo simulation and experiments. Its response matrix was calculated by taking all the responsible processes and interactions of gamma rays in water as well as in the detector into account using Monte Carlo simulation method. An improved R-L algorithm proposed was applied to the gamma spectrum to get a corresponding deconvolution spectrum with good energy resolution, which made the radionuclide identification and calculation easier. The analysis of the synthetic spectrum simulated in the experiment and actual spectrum measured in the marine environment both showed promising results for the spectrum unfolding method as well as for the qualitative and quantitative estimation of radionuclides in the seawater.

To apply the spectrum analysis method proposed to the automatic software of NaI(Tl) detector for the radioactivity monitoring in the marine environment, there are still some problems to be further explored. How to set the optimal parameters such as iteration times in deconvolution under constraint conditions. How to determine the true and false peaks from some weak peaks appearing in the deconvolution spectrum. To examine and improve the spectrum analysis method, some experiments are to be carried out by putting some radioactive standard solution into the seawater tank.

Acknowledgements

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