Preliminary Study on the Use of NaI(Tl) detector for the Marine Environmental Monitoring

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1. Introduction

The continuous monitoring of natural radioactivity is playing a fundamental role in radiation protection, even providing the pollution warning. After the 2011 Fukushima accident, marine pollution has become a significant concern. The traditional method of measuring in situ sampling with high purity germanium detectors takes long counting times to get the quantitative results. Now that the rapid measurements are required for natural radioactivity monitoring, NaI(Tl) detectors have been commonly used with high detection efficiencies.

Notwithstanding the wide utilization of NaI(Tl) detectors, there is still lack of the practical method of efficiency calibration for radioactive sources in certain types. The detection efficiency is usually calibrated by Certified Reference Materials (CRM) but these are not always available process for every type. Therefore, Monte Carlo has been used to describe the dimension and characteristics of detector and the radiation sources.

In this present work, the Monte Carlo N-Particle transport code (MCNP) was used to simulate 3 x 3 inch NaI(Tl) detector and source types. The CRM volume source in 450 mL Marinelli beaker was measured to obtain the parameters in the Gaussian option to generate spectra of detector responses. In addition, the detection efficiency of measurement and simulation were evaluated and compared.

2. Materials and Methods

A 3 x 3 inch NaI(Tl) detector, model 905-4 (Ortec-Ametek) was used for gamma ray measurement. The simulation model was performed using Monte Carlo simulation to calculate the detection efficiency of NaI(Tl) detector.

2.1 Monte Carlo simulation

The experimental setup for the calibration of the NaI detector described in Fig.1 was modeled with MCNP code. The NaI crystal density was $3.6667 \text{ g}\cdot\text{cm}^{-3}$, the MgO reflector density was 2 g $\cdot\text{cm}^{-3}$ and the aluminum housing of the NaI/PM set density was 2.702 g $\cdot\text{cm}^{-3}$. The photomultiplier (PM) tube is separated from the NaI crystal by a 3 mm thick glass window having an effective density of 2.2 g $\cdot\text{cm}^{-3}$ [1]. To simulate the measurement more accurate, glass housing and copper wires of PM tube were considered in the model

geometry. The geometry information of the volume source, CRM in 450 mL Marinelli beaker, was selected as discussed in Kang et al. (2016) [2]. The schematic diagram of 450mL Marinelli beaker is shown in Fig.1 [1-3].



Fig. 1 MCNP geometry of the 450 mL Marinelli beaker and NaI(Tl) detector.

The calculation was performed by mode p, tracking photons. To simulate the statistical broadening of the photopeak, the Gaussian Energy Broadening (GEB) option were used [4]. In MCNP code, GEB is defined by the Full Width at Half Maximum (FWHM):

$$FWHM = a + b\sqrt{E + cE^2}$$
(1)

where E is the energy of the particle and constants a, b and c are derived from the measured photopeak, of which units are MeV, $MeV^{1/2}$ and 1/MeV, respectively.

2.2 Measurement

CRM volume source contained in 450 mL Marinelli beaker was prepared by Korea Research Institute of Standards and Science (KRISS) and measured on top of the detector head. The radionuclides were contained in Agar and listed in table 1. The measurement was performed in a cylindrical lead shield to eliminate the interferences of background radiation. The standard disk sources (¹³⁷Cs, ⁶⁰Co and ²²Na) were used as isotropic point source to evaluate the FWHM of CRM

volume source. The point sources were placed at a distance of 10 cm from the detector surface.

| Nuclide | Half life (Day) | Gamma ray | | Activity |
|-------------------|--------------------|-----------------|------------------|----------|
| | | Energy (MeV) | Intensity (%) | (Bq) |
| ²⁴¹ Am | 158004 | 0.05954 | 35.92 | 1448 |
| ⁵⁷ Co | 271.81 | 0.12206 | 85.49 | 298 |
| ¹³⁹ Ce | 137.641 | 0.16586 | 79.90 | 373 |
| ¹¹³ Sn | 115.09 | 0.39170 | 64.97 | 689 |
| ⁸⁵ Sr | 64.850 | 0.51401 | 98.5 | 1155 |
| ¹³⁷ Cs | 10976 | 0.66166 | 84.99 | 695 |
| ⁶⁰ Co | 1925.23 | 1.17323 | 99.85 | 817 |
| | | 1.33249 | 99.9826 | |
| ⁸⁸ Y | 106.63 | 0.89804 | 93.7 | 1777 |
| | | 1.83605 | 99.346 | |

Table 1. The specification of Certified Reference Materials

3. Results

3.1 Energy resolution

To evaluate the energy resolution of the NaI(Tl) detector for different types of sources, sources given in table 1 were respectively measured. The full width half maximum of two types of sources at various gamma-ray energies is shown in Fig. 2. The solid line is a fitting curve of FWHM derived from the CRM volume source measurement. The comparison of the volume and point source values on the FWHM of ¹³⁷Cs, ⁶⁰Co and ²²Na (1.275 MeV) are also given in Fig.2. According to the agreement, the parameters in the GEB option of simulation (a: -0.01392, b: 0.06968, c: -0.05862) were extracted from fitted equation.



Fig. 2 FWHM values measured in different types of sources. The solid line is a fitting curve of volume source values.

The MCNP simulated and the measured energy spectra of CRM volume source of the NaI(Tl) detector can be presented as a function of counts and illustrated as a spectrum graph shown in Fig.3. The parameters of GEB option derived from the fitting curve in Fig. 2 were used for this work.



Fig. 3 Comparison between simulated and experimental spectra of the NaI(Tl) detector in the case of CRM volume source.

3.2 Detection efficiency

The detection efficiency is defined by number of counts, activity, intensity of photons, and volume of the area containing particle source. The geometrical model of CRM source in 450mL Marinelli beaker is shown in Fig.1 simulated as experimental setup. A comparison between simulation and measurement detection efficiency of CRM volume source case is shown in Fig. 4. The solid line is a fitting curve of measured data in which the dots of simulated values follow fairly closely.



Fig. 4 Comparison of detection efficiency between simulation and measurement.

For the marine environment, the calculation of the NaI(Tl) detector efficiency for gamma energy depends strongly on the volume of water mass [6]. The detection efficiency for 137 Cs (0.66166 MeV) diluted in various radius of spherical seawater surrounding the NaI(Tl) detector was simulated by MCNP code and shown in Fig. 5 similar with the results presented by Bagatelas et al. (2010) and Zhang et al. (2015) [5,6]. The horizontal coordinate is the detection radius and the solid line is an exponential fitting curve of the simulated values. As shown in Fig. 5, the larger detection radius in seawater,

the larger detection efficiency, of which saturated value was 0.3198 cps/(Bq/L). Based on the results, the effective detection radius was 80 cm, of which detection efficiency was 0.3189 cps/(Bq/L) at the 99.7% of saturated value.



Fig. 5 The detection efficiency for ¹³⁷Cs in the seawater simulated by MCNP.

4. Conclusions

To determine the efficiency of radioactivity measurement for different environment, in the present work, a Monte Carlo simulation was performed using MCNP code. The MCNP code produced reliable gamma-ray spectra of CRM volume source compared with measured results. The NaI(Tl) detector's detection efficiencies of ¹³⁷Cs (0.66166 MeV) in CRM volume source measurement and simulation are 0.08273 cps/(Bq/L) and 0.08273 cps/(Bq/L), respectively. The simulated value on the marine detection efficiency of ¹³⁷Cs was 0.3189 cps/(Bq/L) at detection radius 80 cm.

For evaluating Minimum Detectable Activity (MDA) of NaI(Tl) detector as a function of gamma-ray energy, measurement of marine environment is imperative. Therefore, marine experiments will be performed to provide calculation of MDA and comparison with simulation results.

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