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Preliminary study on the detection efficiency and estimation of minimum detectable activity for a NaI(Tl)-based seawater monitoring system

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ABSTRACT

To monitor radioactivity levels in seawater Korea Institute of Nuclear Safety has installed and been operating 18 NaI(Tl)-based gamma detectors around the Korean peninsula. This study was conducted to estimate the detector efficiency and MDA of ^{137}Cs in seawater for measurement situations. For this purpose, experiments in the air and a water tank, and Monte Carlo simulations were performed using a seawater radioactivity monitor system with 3 in. × 3 in. NaI(Tl) scintillation detector.

In the geometry reliability assessment using certified reference materials in a disc source, the validity of simulations was obtained by comparing measurement and Monte Carlo simulation results. The FWHM of the seawater radioactivity monitor were obtained from the results of the water tank measurement for applying a Gaussian Energy Broadening (GEB) option to Monte Carlo N-Particle (MCNP) radiation transport code. In addition, the detection efficiency of ^{40}K in the water tank was measured and compared with the Monte Carlo simulation results in order to estimate the MDA and the detection efficiency of the seawater radioactivity monitoring system. For the based condition of water tank, ^{40}K concentration in water tank was controlled to 10.13±0.18 Bq/L, similar to that of real marine.

In laboratory water tank experiments, the detection efficiency of the radioactivity monitor for ^{40}K was measured at 0.184±0.005 cps/(Bq/L), the Monte Carlo simulations showed the similar result of 0.182±0.002 cps/(Bq/L), and the detection efficiency of ^{137}Cs was estimated to be 0.224±0.009 cps/(Bq/L) from the simulations. For 3h measurement in the water tank based condition, the MDA of ^{137}Cs was estimated to be 0.077±0.003 Bq/L. Future research will include detailed studies for detector sizes and seawater salinities.

1. Introduction

Geographically, Korean peninsula is surrounded by Yellow Sea to the west adjacent to China, the East Sea to the east adjacent to Japan, and Korea Strait and the East China Sea to the south. Because South Korea and its neighboring countries use nuclear power plants as one of their main energy sources, the environmental radioactivity monitoring including marine radioactivity has been performed continuously. Recently, the public concern to the marine radioactivity has become more significant, especially after Fukushima accident in 2011. It is important to measure ^{137}Cs, which is a gamma-emitting radionuclide, in marine water within the framework of radiation monitoring.

For these reasons, Korea Institute of Nuclear Safety (KINS) has chaosen NaI(Tl) scintillation detector for real-time marine radioactivity monitoring system in Korea and KINS has been running 18 detectors around Korean peninsula.

Marine radioactivity monitoring, especially for ^{137}Cs, can be conducted on in-situ or in laboratory measurement. In the laboratory, High Purity Germanium (HPGe) detector is commonly used to analyze samples (Eleftheriou et al., 2013; Povinec et al., 1996). Because the HPGe detector profited to thoroughly analyze the radioisotopes of samples due to high energy resolution than other detectors. However, the detector is difficult to use in-situ measurement because Liquid nitrogen or temperature controller is necessary to continues measure samples. Therefore, some other methods are needed for the real-time marine monitoring, which is necessary to provide early radioactive contamination warning, especially in emergencies.

For the establishment of the important underwater gamma-ray monitoring system there have been a few research papers on the use of NaI(Tl) scintillation detectors (Tsabaris and Thanos, 2004; Osvath et al., 2005; Tsabaris et al., 2005; Tsabaris and Ballas, 2005; Tsabaris, 2008; Tsabaris et al., 2008; Cinelli et al., 2016) including actual measurements and simulations, which worked as a useful guide for the authors. Recently, some Greek researchers reported the findings on the use of low-resolution spectrometers with stainless steel enclosure for the deep-sea applications (Tsabaris et al., 2018) and the scientific features on the probable use of 2 in. × 2 in. $CeBr_3$ (Tsabaris et al., 2019) which can distinguish photo-peaks with neighboring energies, especially the 606 and 796 keV of ^{134}Cs and 662 keV of ^{137}Cs because of its better resolution (3.5% at 662 keV).

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Notwithstanding the wide utilization of NaI(Tl) detectors, some disadvantages must be considered and resolved for more accurate measurement. One of the disadvantages is a relatively poor resolution, which may leads to overlapping of spectral lines so that makes the system difficult to identify precise peaks of the various gamma ray contribution (Vlastou et al., 2006).

Efficiency calibration of radiation detectors is an important part of the procedure for obtaining quantitative results to determine the activity for each radionuclide. The efficiency calibration is usually performed by measuring radioactivity quantities of Certified Reference Materials (CRM) called the standard source. In the marine environment monitoring field, to execute the energy calibration process of the detector for accurate marine radioactivity measurement is difficult. However, the calibration is possible by substitute measurement in a laboratory instead of calibrating at seawater (Tsabaris et al., 2005, 2018). If it is difficult to reproduce the marine environment in a laboratory, the efficiency calibration by the Monte Carlo method can be an alternative. The Monte Carlo simulation has been commonly used as a convenient tool for obtaining statistical properties as a particle tracking method that enables analyze some virtual situations (Zhang et al., 2015; Tsabaris et al., 2018). There are limitations to reproducing the real marine environment in the laboratory, the Monte Carlo simulation makes possible for testing and analyzing a detector in the isotropic aquatic environment of various situations.

In this research, to develop the practical way of evaluating the performance of a marine radioactivity monitoring system in the aquatic environment, Monte Carlo N-Particle (MCNP) Transport code and measurement in a laboratory were utilized in the beginning. MCNP code was performed to determine the effective detection radius for experimental setup. In the main part of the research, MCNP was used to simulate the experiments conducted in the air and with the water tank. As calibration work, the CRM of the disk type for calibration was measured by the NaI(Tl) detector and simulated in the air. Validation works for the reliability of MCNP geometries were made by comparing measurement and simulation results. In addition, the CRM and different ^{40}K background levels were measured in the water tank and the same experimental setup was simulated. After the validation of the simulation model, the Minimum Detectable Activity (MDA) of ^{137}Cs in the virtual marine environment was estimated.

2. Materials and methods

For real-time measurement in marine environment in Korea, in-situ marine radioactivity monitoring system is used as shown in Fig. 1(a). The system consists of a 3 in. × 3 in. NaI (Tl) scintillation crystal connected to a photomultiplier tube (PMT) (SCIONIX, less than 7% energy resolution at 662 keV) and a multi-channel pulse-height analyzer (SI detection). The whole system is enclosed in a MC nylon cylinder. The measurement results of the system are stored on computer using a serial port. An operating temperature range of the system is between −10 °C and 60 °C. The system automatically can correct the channel shift of ^{40}K peak from the change of external environmental conditions. The system electricity consumption is less than 2 W as such in Table 1. In this research, the monitoring system was simulated and used in the laboratory experiment.

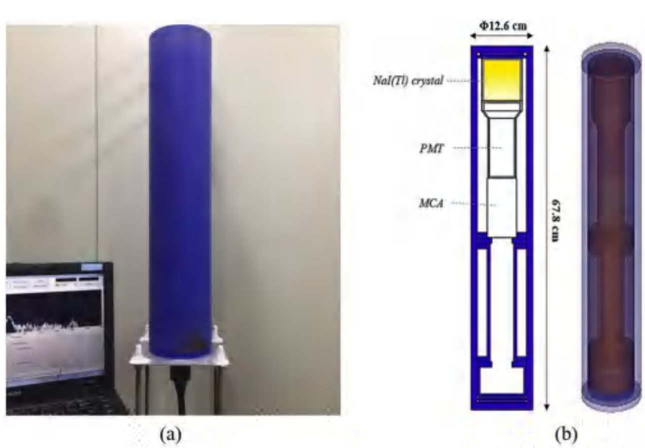

Fig. 1. (a) Marine radioactivity monitoring system (b) Monte Carlo simulation geometry of marine radioactivity monitoring system.

2.1. Monte Carlo simulation

2.1.1. Simulation of marine radioactivity monitoring system

Monte Carlo simulation with the Monte Carlo N-Particle (MCNP) radiation transport code (Los Alamos National Laboratory) was performed for the marine radioactivity monitoring system by considering the detailed dimensions and the materials of the experiment, as shown in Fig. 1(b) and Table 2. The density of the NaI(Tl) crystal is 3.667 g·cm^{-3}, the MgO reflector is 2 g·cm^{-3} (Saito and Moriuchi, 1981) and the aluminum housing is 2.702 g·cm^{-3}. The photomultiplier tube (PMT) is separated from the NaI(Tl) crystal by a 3 mm thick glass window of which effective density is 2.2 g·cm^{-3} (Mouhti et al., 2018). In addition, other parts of the photomultiplier tube, the glass housing of PMT and copper wires of PMT electrical circuits, are considered as an MCNP cell. The material of whole system enclosure is the MC nylon, a type of engineering plastic, the shape of the enclosure is a cylinder. It has an external diameter of 12.6 cm and a height of 67.8 cm, as shown in Fig. 1 (b).

The MCNP calculations are performed in mode p, taking into account the photon interactions with materials. In MCNP code, F8 tally is used to determine the energy distribution of pulses created in a detector (Booth, 2002). In order to simulate the Gaussian-shaped spectrum obtained from realistic response of the detector, a Gaussian Energy Broadening (GEB) option is used as a special treatment for F8 tally. The GEB is defined by Full Width at Half Maximum (FWHM):

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

where E is an gamma-ray energy (MeV) and the constants, a, b and c, are derived from experimental results. To determine the parameters of above equation from the measured gamma-ray spectrum, the CRM disc source was used which has various gamma energies, listed in Table 3. As a result, the extracted a, b and c are equal to −0.0120879 MeV, 0.0700906 MeV$^{1/2}$ and −0.1073321 MeV^{-1}, respectively.

2.1.2. Effective detection radius

In organizing an aquatic environment experiment in the laboratory, there was a size limit on the water tank. In order to determine the practical size of the water tank, MCNP simulations were performed for the marine radioactivity monitoring system in different radius of spherical seawater geometry, as shown in Fig. 2(a). In the seawater volume, gamma-ray energies of radionuclides (^{137}Cs, ^{40}K) were diluted to calculate the detection efficiency at different detection radius. As

Table 1
The specifications of the marine environment monitoring system.

Component	Specification
Detector parts	Scintillation: NaI(Tl), 3 in. × 3 in. Energy range: 50 – 3000 keV Resolution: ~7% at 662 keV
Analog digital converter	1024 channels
Weight	Detector parts: <10 kg Local control parts: 3 kg
Operating temperature	−10 to +60 °C
Commutation protocol	Bluetooth/Lan/Wi-Fi/GSM/CMDA/3G
Electricity consumption	<2 W

Table 2
Material information for Monte Carlo simulations.

Material	Constituents	Density (g/cm^3)
NaI detector	Na, I	3.667
Reflector	Mg, O	2.0
PMT	Cu, Air	0.838
Detector housing	Al	2.702
MC-nylon enclosure (engineering plastic)	H, C, O	1.15
Seawater	H, Br, O, Na, Mg, S, Cl, K, Ca, Br, Sr	1.027

shown in Fig. 2(b), the trend of the detection efficiency curve reached equilibrium when the detection radius increased. The trend of detection effective radius was similar to another research result (Bagatelas et al., 2010). From the exponential fitting, the effective detection radii at which detection efficiencies are 95% of their saturated value, which were determined for each gamma-ray energy. From the simulated results, the effective detection radius of 662 keV (^{137}Cs) is 45 cm and of 1460 keV (^{40}K) is 62 cm.

2.2. Experimental setup

2.2.1. Aquatic environment for the laboratory experiment

To determine an adequate size of the water tank for emulating a marine environment, gamma-ray transmission in the water was also considered. The transmission probability of gamma-ray energies was calculated with the XCOM attenuation coefficient data. In case of 662 keV, the transmission probability is 0.57% when the 60 cm thick water is surrounding the detector. After due consideration, the 3-ton water tank, which can cover the effective detection distance (i.e. radius) of mainly target gamma-energy (662 keV, ^{137}Cs), was used for laboratory experiments.

The marine radioactivity monitoring system was located in the water tank and the center of the NaI(Tl) crystal was equidistant from the wall of the water tank, as shown in Fig. 3. In the water, potassium chloride (KCl) was diluted for three different cases to control ^{40}K background levels. To avoid sedimentation, nitric acid (70% HNO_3 1 L) was also diluted (Naumenko et al., 2018). Additionally, for the homogeneous condition in the water tank, an electric circulation pump was equipped. The measurement was conducted in manual mode to cancel an energy auto-calibration, and the stability of water temperature condition was controlled by air conditioner during the experiments.

2.2.2. Radiation source

The certified reference materials in disc source (Table 3, Fig. 4) certified by Korea Research Institute of Standards and Science (KRISS) were used to conduct the energy and efficiency calibration of NaI(Tl) detector. The radioisotopes of the CRM were shielded by a polyethylene (thickness: 15 μm). The measurement was performed in the water tank, positioning the CRM source as close to the head of the detector as possible.

After diluting the KCl in the water tank, three different activities of ^{40}K were measured and by using HPGe detector for 86,000 s measurement. For the case 1, 2 and 3, each activity of ^{40}K is equal to 5.28±0.17 Bq/L, 10.13±0.18 Bq/L and 14.41±0.26 Bq/L, respectively.

3. Results

3.1. Validation of Monte Carlo simulation model

For validation of the Monte Carlo simulation model, air-based CRM

Table 3
The specification of Certified Reference Materials (CRM) in disc-type source, certified by Korea Research Institute of Standards and Science (KRISS).

Nuclides	Half life [day]		Activity [Bq]		γ-ray energy [keV]	γ-ray emission probability [%]	
	Half life	**Uncertainty**	**Activity**	**Uncertainty**		**Prob.**	**Uncertainty**
^{57}Co	271.81	0.04	539	22	122.06	85.40	0.14
					136.47	10.71	0.15
^{139}Ce	137.641	0.020	673	27	165.86	79.9	0.04
^{51}Cr	27.704	0.004	53875	2200	320.08	9.89	0.02
^{113}Sn	115.09	0.03	1245	50	391.7	64.97	0.17
^{137}Cs	10976	29	1255	51	661.66	84.99	0.20
^{60}Co	1925.23	0.29	1477	59	1173.23	99.85	0.03
					1332.49	99.98	0.0006
^{88}Y	106.63	0.05	3212	130	898.04	93.7	0.3
					1836.05	99.34	0.025

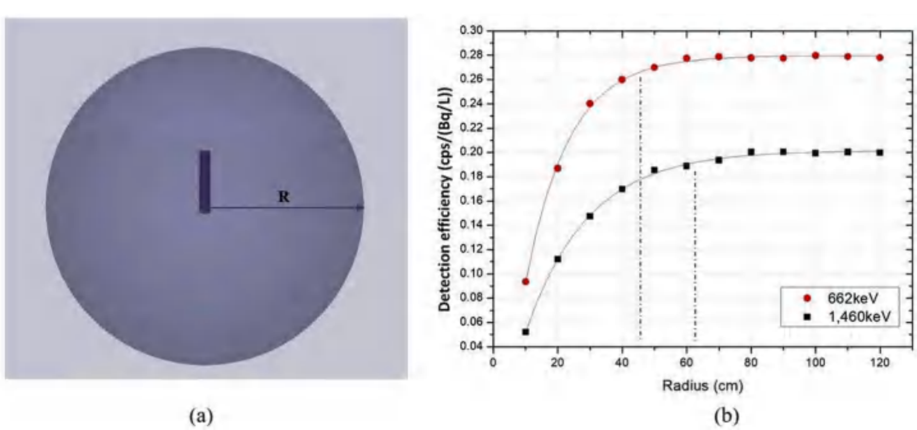

Fig. 2. (a) Monte Carlo simulation for calculating effective detection radius in the aquatic environment, (b) Detection efficiency of different energies in the aquatic environment.

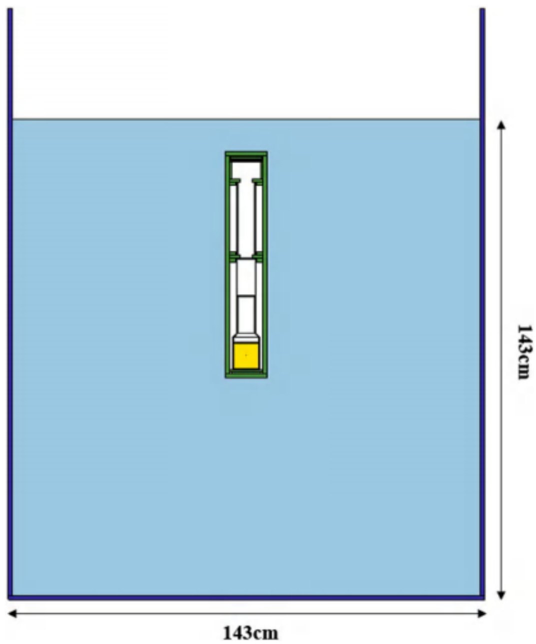

Fig. 3. Monte Carlo simulation geometry of monitoring system in the water tank.

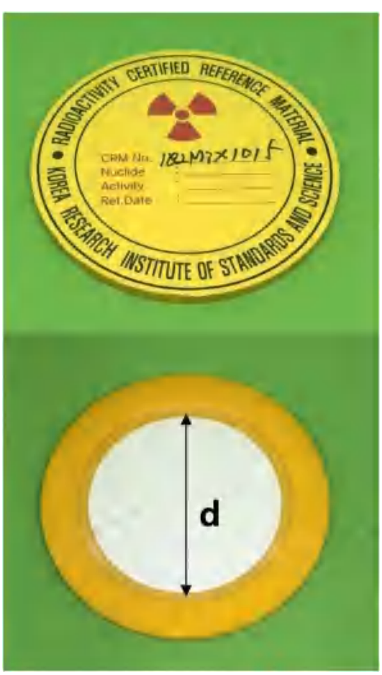

Fig. 4. Radioactivity Certified Reference Materials (CRM) in disc type source (d = 47 mm).

source measurement has been done and get the reliability of geometries. The comparison between the simulation and measurement results showed the conformity of detection efficiency that shown in Fig. 5. There is a good agreement between the two results with an R^2 of 0.982.

As part of the calibration process of the marine radioactivity monitoring system, the CRM source in the water tank was measured and simulated by the MCNP code. In order to validate the simulation, spectra acquired from the 3-h measurement and MCNP code were compared as presented in Fig. 6. The figure shows a good agreement neglecting peaks in over 2 MeV, and two high energy peaks of the figure are presumed to be sum peaks of ^{88}Y and ^{60}Co (Al-Dargazelli and Mahmood, 1990).

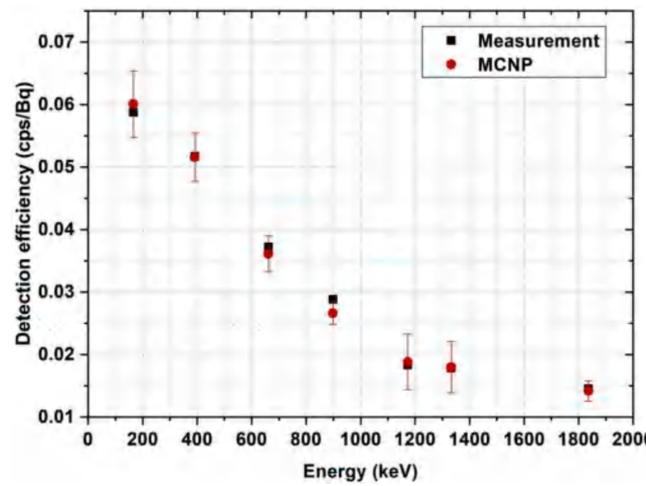

Fig. 5. Detection efficiencies from experiment and simulation with the CRM disc source in front of NaI(Tl) detector (measurement in the air).

Detection efficiencies for each gamma-ray energy were obtained using the following equation:

$$\varepsilon = N / (A \cdot \boldsymbol{I}_{\gamma} \cdot T) \tag{2}$$

where N is net count, T is live time of measurement in second, A is nuclide activity in Bq, which were corrected with decay time, and I_{γ} is gamma-ray emission probability. The values with their statistical uncertainties of these parameters used for calculation are listed in Table 3. The detection efficiencies derived from simulated results were compared to those of the experimental results, as shown in Fig. 7. There is a good fit between the two results with an R^2 of 0.997.

In the case of the diluted radioactivity source in water, the detection efficiency curve of distributed gamma-ray energies was obtained by the MCNP calculation as shown in Fig. 8. Measurement results of the three different activities of ^{40}K in the water tank is provided in Table 4. In three cases, an average value of the detection efficiency, resulting from KCl, is 0.184±0.005 cps/(Bq/L). Similarly, the fitted value for efficiency at 1460 keV was 0.182±0.002 cps/(Bq/L), generated by simulated results.

Additionally, we calculated the detection efficiency in aquatic environment with different salinities (17.09 – 40.82‰ as salinity range of normal seawater surrounding Korea) (Lee et al., 2003; Park et al.,

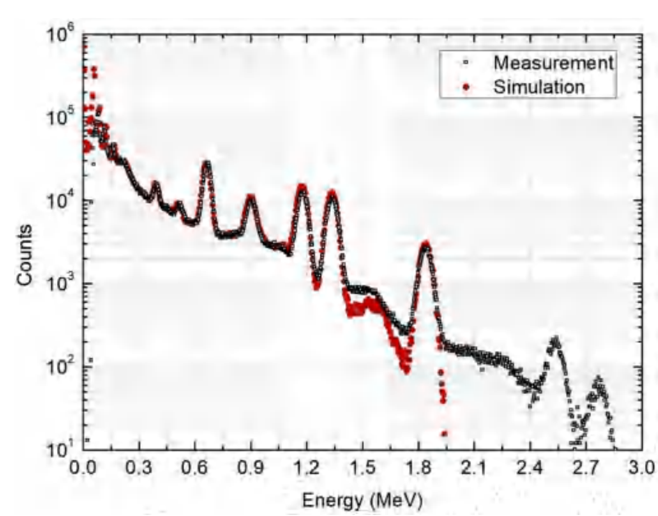

Fig. 6. Experimental and simulated spectra of the marine radioactivity monitoring system in the case of CRM disc source measurement in the water tank.

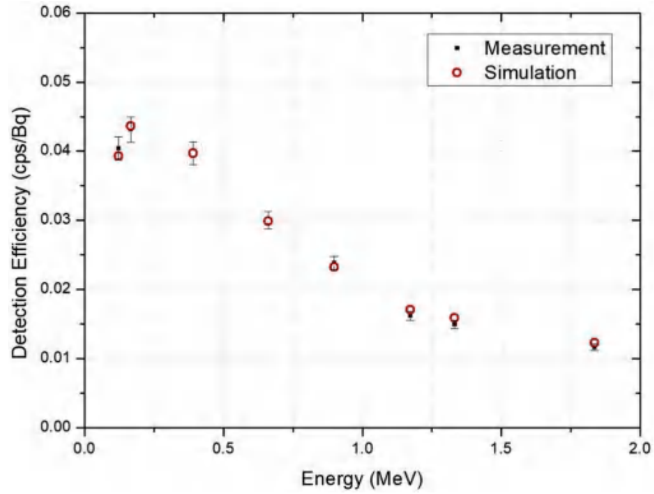

Fig. 7. Detection efficiencies from experiment and simulation with the CRM disc source in front of NaI(Tl) detector (measurement in the water tank).

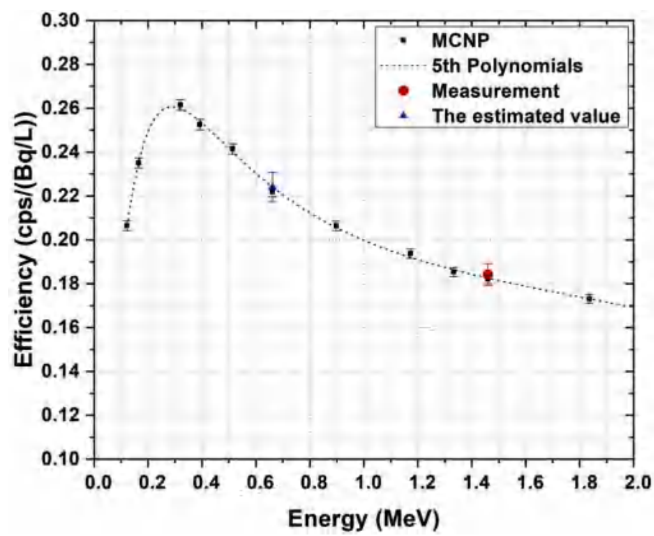

Fig. 8. Detection efficiency curve of marine radioactivity monitoring system obtained by simulation in the case of distributed radionuclide measurement in the water tank.

Table 4
The results of 3 h measurements at different ^{40}K levels and estimation of ^{137}Cs MDA in the aquatic environment.

		Case 1	Case 2	Case 3
^{40}K	**Activity [Bq/L]**	5.28±0.17	10.13±0.18	14.42±0.26
	Count rate [cps]	0.107±0.004	0.202±0.008	0.297±0.012
	Detection efficiency [cps/(Bq/L)]	0.184±0.010	0.181±0.008	0.187±0.008
^{137}Cs	**MDA (3 h meas.) [Bq/L]**	0.049±0.003	0.077±0.003	0.085±0.004

2017). The detection efficiency at 662 keV ranges 0.220 to 0.216 cps/(Bq/L). This results present that the detection efficiency is weakly affected by density of environment, but the change in ^{40}K level will change the background level around 662 keV and will change the MDA estimation (Lee et al., 2003; Park and Lee, 2017).

3.2. ^{137}Cs detection efficiency and minimum detectable activity

Through the aforementioned validations, the detection efficiency for ^{137}Cs was obtained based on the reliability of the simulation model. As illustrated in Fig. 8, the detection efficiency for 662 keV is 0.224±0.009 cps/(Bq/L) or 0.224 × 10^{-3}±0.9 × 10^{-5} cps/(Bq/m^3), which allows quantitative evaluation of marine radioactivity monitoring system in the aquatic environment. And the results are similar to the estimated detection efficiency 0.21±0.01 cps/(Bq/L) of an acetal enclosure material at 662 keV (Tsabaris et al., 2018).

In an attempt to evaluate the performance of the marine radioactivity monitoring system, the minimum detectable activity (MDA) for a certain time is estimated. The MDA of ^{137}Cs in the aquatic environment is calculated according to the following Currie's method (Currie, 1968):

$$MDA = L_D / (\varepsilon \cdot I_\gamma \cdot T) \tag{3}$$

where ^{137}Cs detection efficiency ε is 0.224±0.009 cps/(Bq/L) and L_D is the detection limit in counts with a 95% confidence level, given by:

$$L_D = 2.71 + 4.65\sqrt{B} \tag{4}$$

where B is the number of counts of background continuum in a specific Region of Interest (ROI). For the estimation of ^{137}Cs in the aquatic environment, an interval of 1.275 times the FWHM on each side of the peak was used as the width of the expected peak i.e., providing 99.73% peak area coverage (International Atomic Energy Agency (IAEA), 2017). The background spectra in the water tank were recorded for 3-h varying ^{40}K activity as listed in Table 4. The estimated ^{137}Cs MDA of three cases are 0.049±0.003, 0.077±0.003, and 0.085±0.004 Bq/L. As the ^{40}K activity increased, the background region of 662 keV increased, consequently the estimated result of ^{137}Cs MDA also increased. The preliminary uncertainty of MDA was calculated by the error propagation method of each parameter like Poisson distribution of the detection counts, the detection efficiency calculated by MCNP, and the emission probability of the gamma ray. More detailed research on MDA uncertainties will be conducted in the next stage. At this state of research the ^{137}Cs MDA is mainly determined by background provided by ^{40}K Compton effect. For the possible ^{137}Cs contamination of NaI(Tl) detector the authors expect the increase of ^{137}Cs MDA. One possible approach for the cases will be accurate background subtraction method.

4. Conclusions

In-situ marine radioactivity monitoring system using 3 in. × 3 in. NaI (Tl) scintillation detector is playing a fundamental role in Korea marine radiation protection. As a real-time system, the keys of the detector performance which are the detection efficiency as a function of gamma energy and the MDA of ^{137}Cs are investigated with measurements in air and water tank and MCNP simulations.

Prior to the water tank experiment, we measured the CRM source by the detection system in the air to get the reliability of MCNP geometries. After the validation, taking into consideration the experimental setup (i.e. water tank) for the aquatic environment, the effective detection distance of the monitoring system was investigated with Monte Carlo simulation using MCNP code and the size of the water tank was determined. Then the efficiency calibration was performed by measurement of the CRM disc source in the water tank and simulations. As the result of the efficiency calibration, the experimental and simulated detection efficiencies matched with each other well. Furthermore, in the case of added ^{40}K concentration which is similar to real marine conditions, the simulated value of detection efficiency was 0.182±0.002 cps/(Bq/L) in good agreement with experimental averaged-value, 0.184±0.005 cps/(Bq/L).

From these comparisons, the validity of simulations was obtained that the simulation model provides fairly reliable results for continued quantitative analysis, such as estimation of MDA in the aquatic environment. Therefore, ^{137}Cs MDA in various ^{40}K background levels was estimated by Currie's method with simulated detection efficiency. For the ^{40}K activity of case 2 (10.13 ± 0.18 Bq/L), similar to that of Korea marine environment, the ^{137}Cs MDA was about 0.077 ± 0.003 Bq/L for 3 h measurement time.

In the further study, the MDAs of various nuclides including ^{131}I and ^{134}Cs in the real marine environment will be estimated through continuous validations and improvements of Monte Carlo simulation model. And different kinds of detectors will be studied to analyze the environment radiation, such as 4 in. × 4 in. NaI(Tl) detector or $LaBr_3$ scintillation detector, which has internal contamination as a drawback but advantages radiation analysis for energy calibration and offers relatively better resolution than NaI(Tl) detector.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2020.106222.

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