# Development of virtual gamma-ray energy spectrum database under various conditions using MCNP simulation

Yoomi Choi, Young-Yong Ji, Sungyeop Joung
*Korea Atomic Energy Research Institute, 989-111 Daedeok-daero, Yuseong-gu, Daejeon 34057, Republic of Korea*
*Corresponding author: cym1031@kaeri.re.kr*

## 1. Introduction

Gamma-ray spectrometry is one of the most effective methods for detecting radionuclides and measuring the radioactivity. It can apply not only to laboratory measurement, but also to *in situ* measurements for various field, such as decommissioning, environmental monitoring and radioactive waste(hereinafter referred to as "radwaste") management etc. As the demand in the field of application increases, a training system for technical profession is also required. In order to increase the proficiency of spectrometry, it is necessary to experience directly analyzing the measured spectrum. However, it is very difficult to obtain a measured spectrum example for which measurement conditions are accurately known. As an alternative to this problem, a database for generating a virtual gamma-ray energy spectrum of artificial radionuclide was developed by computational simulation. This paper describes the methodology for development of virtual gamma-ray energy spectrum database.

## 2. Methods and Results

### *2.1. Measurement situations and conditions*

The conditions for measurement situations that are mainly used in practice were set in advance to establish the database for virtual spectrum generator. The measurement situations considered are as *in situ* measurement of ground surface (for environmental monitoring), concrete wall (for facilities decommission) and radioactive waste drum (for radwaste management). In each situation, it is assumed that the detector is located at a distance of 1 m from the measured target. Figure 1 shows the geometry structure of assumed measurement situation. The types of detector were selected as HPGe detector, 3"φ×3" NaI(Tl) scintillation detector and 2 2 2"φ×2" $LaBr_3$(Ce) scintillation detector, which were widely used for gamma-ray energy spectrometry.

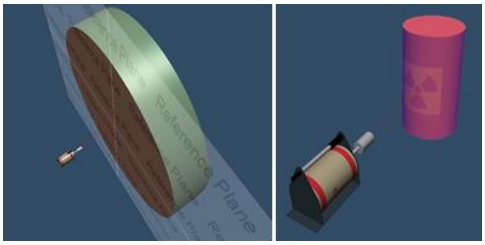

**Figure 1. The geometry of measurement situations**

The types and distributions of radioactive nuclides were determined for each measurement situation. For the ground surface measurement, 5 representative gamma-emitting nuclides that can be released from the nuclear facilities into the atmosphere during accident ($^{60}$Co, $^{134}$Cs, $^{137}$Cs, $^{131}$I, $^{103}$Ru) were selected. In order to reflect that the radionuclides deposited on the surface diffuse into the soil over time and have a vertical distribution according to the depth, the exponential distribution function with the maximum concentration at the surface suggested by ICRU report 53[1] was applied. The Equation (1) for the concentration of radioactivity in the ground is as follows:

$$A = A_0 \times e^{-\rho_s z/\beta} \quad (1)$$

Where, A and $A_0$ are the radioactivity per unit mass at any point and at the surface [Bq/kg], $\rho_s$ is the density of soil [g/cm$^3$], z is the depth at the point [cm] and β is the relaxation mass per unit area [g/cm$^2$], which is the parameter determining the depth distribution function. In this study, the distribution functions with 7 value of β (0, 0.3, 0.5, 1, 3, 5, 10) were considered. β values of 0 and 0.3 represent the surface distribution, and 0.3 responds to the consideration of the roughness of the soil surface. [2] β values of 0.5 and 1 are representative of less than 1 year after the fallout, 3 and 5 are representative less than 10 years after fallout, and 10 represents more than 10 years after fallout. [1,3] The radius of the nuclide distribution region was also considered as various values. (1, 5, 10, 20, 30, 50, 100 m)

For the concrete wall, 10 representative gamma-emitting nuclides that may exist in normal decommissioning wastes were selected. ($^{60}$Co, $^{94}$Nb, $^{134}$Cs, $^{137}$Cs, $^{108m}$Ag, $^{110m}$Ag, $^{152}$Eu, $^{154}$Eu, $^{124}$Sb, $^{125}$Sb) The radionuclides in the concrete wall was assumed to be distributed uniformly up to an area of 5 cm in the depth direction.

For radwaste drum, 5 representative gamma-emitting nuclides in the regulation on the delivery of low and intermediate level radwastes were selected. ($^{58}$Co, $^{60}$Co, $^{94}$Nb, $^{137}$Cs, $^{144}$Ce) The distribution of radionuclides in the drum was assumed to be uniform. Solid waste and a cement were considered as filling materials inside the drum, respectively.

### *2.2. Background measurement*

Since a large amount of natural radionuclides exist in actual measurement, the energy spectrum of background radiation of these natural radionuclides is always measured in actual *in situ* gamma spectrometry. To reproduce the virtual spectrum similarly to the actual spectrum, it is necessary to consider not only artificial radionuclides but also natural radionuclides that exist in the background. In addition, the various gamma-ray peaks caused by these natural radionuclides can be helpful in training analytical capabilities by identifying peak interference. Therefore, the background spectrum for synthesizing into the virtual spectrum was obtained through real measurement.

The background measurements were performed at a height of 1 m from the ground and the floor of laboratory inside the building, where the three measurement situations under consideration mainly occur. The background spectra for the ground were measured surface in several region, because the background spectrum may be different according to various concentration conditions. For NaI(Tl) and $LaBr_3$(Ce) detector, the background spectra was measured in Daejeon and Busan(Gijang), and HPGe detector (Canberra GC4019 model) was used for measurement in Daejeon and Jeju where background gamma dose rate is low. For laboratory, all types of detectors were used for the background measurement. Figure 2 shows the measured background gamma-ray energy spectrum. In these spectra, gamma peaks derived from several natural radionuclides, such as $^{40}$K, $^{214}$Bi, $^{208}$Tl, were detected. Energy calibration was performed using the gamma energies of known natural radionuclides.

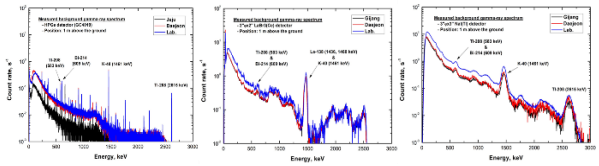

**Figure 2. The measured background gamma-ray energy spectra**

*2.3. Monte Carlo simulation*

To obtain the virtual spectra of artificial radionuclides, Monte Carlo simulation was performed for each measurement condition for each detector using MCNP 6.

The geometry of detectors in the simulation were shown in Figure 3. The source information was input as the gamma-ray energy and gamma-ray emission rate of each nuclide. In this study, only gamma-ray emission was considered, and the generation of annihilation gamma-ray due to other decay (e.g. $\beta^+$ decay) was not considered. To obtain the energy spectrum measured by the detector, calculations were performed by simulating the radionuclides and distribution for each measurement condition previously set using F8 tally. In the case of NaI and $LaBr_3$ that have low resolution, gamma energy broadening effect (GEB) function was applied to reproduce the peak shape in the measured spectrum of an actual detector. In the case of a large radius like 100m, the DXT function and larger number of nps were applied to reduce uncertainty and secure sufficient peaks.

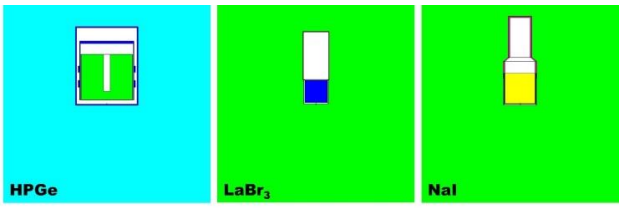


**Figure 3. The geometry of detectors in the simulation**

2.3.1. Ground surface measurement situation

A total of 245 simulations were performed for 5 gamma nuclides, 7 depth distribution, and 7 radius length for each detector type. The simulated source distributions in the ground according to the β value were shown in Figure 4. The detector was located at a height of 1m from the ground surface. The density of soil was set as 1.6 g/cm$^3$.

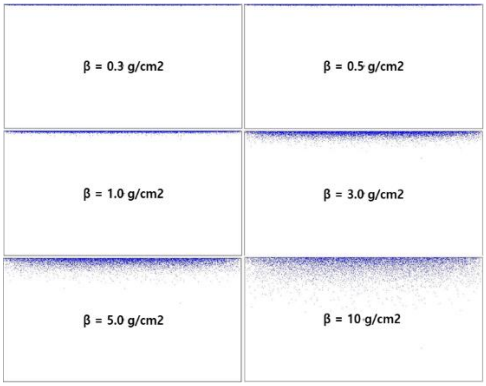


**Figure 4. The distribution of source in the simulation according to the value of β**

The calculation results were finally converted to values in cps/Bq/cm$^2$ by correcting the gamma emission rate for each nuclide and the radial distribution in the input file.

2.3.2. Concrete wall measurement situation

The concrete wall was simulated with concrete with a size of 10m×10m×0.5m and a density of 2.3 g/cm$^3$. The source was uniformly distributed in the radial direction and uniformly distributed in the thickness direction from the surface to a depth of 5 cm. The detector was positioned 1 m from the surface of wall. Simulations for 10 nuclides were performed for each detector, respectively. The results were converted to count rates per unit Bq/kg.

2.3.3. Radiowaste drum measurement situation

In the simulation, it was assumed that the drum was made of carbon steel with a density of 7.82 g/cm$^3$ and the interior was 100% filled with solids(0.252 g/cm$^3$) or cement(1.665 g/cm$^3$). The structure of the drum was set as a 200 L cylindrical container. The detector was located 1m from the center of the drum. Simulations for 5 nuclides were performed for each detector. As in the case of the concrete wall, the results were converted to count rates per unit Bq/kg by correcting the density of the filling material.

### *2.4. Establishment and verification of database*

#### 2.4.1. Database for virtual spectrum generation

The database consisted of the measured background spectra and the simulation results for all combinations of the measurement conditions. The virtual gamma-ray energy spectrum can be generated by synthesizing the background spectrum and the simulation results for nuclides of interest as in Equation (2).

$$c_V(\mathrm{ch}) = [cps_B(ch) + \sum_i cps_i(ch) \times A_i] \times t_s \qquad (2)$$

Where, $c_V$ is the number of counts for each channel of the virtual spectrum, $cps_B$ is the count rate [cps] for each channel of the background spectrum, $cps_i$ is the count rate [cps] for each channel of the simulated spectrum of nuclide i. $A_i$ is the radioactivity concentration of nuclide i [Bq/cm$^2$ or Bq/kg]. $t_s$ is the counting time [sec].

In the process of summing the spectra, gamma peak locations may be different due to different channel-energy correction information. To align this, when the count rate for each energy in the simulated spectrum is combined with the background spectrum, the theoretical counts value obtained by linear interpolation was allocated to the matching channel of background spectrum. The number of channels of the simulated spectrum was adjusted to 1k for NaI and $LaBr_3$, which have lower energy resolution, and 16k for HPGe, which has higher energy resolution, for high-accuracy linear interpolation.

#### 2.4.2. Verification of virtual spectrum using ERS

To verify the adequacy of the generated virtual spectra, radioactivity analysis was performed with the previously developed program such as ERS (Environmental Radiation Survey) program[4] and ISOCS (In-situ Object Counting System, Canberra). The analysis results were compared with the true values of radioactivity input during virtual spectrum generation.

In the case of the ERS, since EPS program is only applicable for the semi-infinite ground contamination, the virtual spectra for ground surface contamination with a radius of 100m were used for verification. The considered nuclides were $^{131}$I, $^{134}$Cs and $^{137}$Cs, which can be applicable for ERS program. The representative gamma-ray energies of these nuclides were respectively selected as 364, 605 and 662 keV for HPGe, and as 364, 796 and 662 keV for NaI and $LaBr_3$. The reason for choosing different energies depending on the detector type is that the peaks at 605 keV of $^{134}$Cs and 609 keV of $^{214}$Bi (natural radionuclide) in the spectrum for NaI and $LaBr_3$ overlap due to the low resolution of scintillator. However, since the peak at 796 keV overlaps with the peak at 802 keV of $^{134}$Cs, subtraction evaluation was performed through the following Equation (3) to induce the net counts of only the peak of 796 keV.

$$N_{796} = N_{796+802}/(1 + \frac{\gamma_{802}}{\gamma_{796}}) \qquad (3)$$

Where, N means the net counts, γ means the gamma-ray emission rate and the numbers in the subscripts represent the gamma-ray energy. This equation assumes that the counting efficiency values are the same as the energies of the two peaks are almost similar.

The net counts of peaks were obtained by peak analysis using Genie 2000 and entered into the EPR program. The analysis results for each nuclide were compared to the true radioactivity input values. Most of the relative errors were within about 20%, which in an acceptable value considering the *in situ* measurement on a large scale. Therefore, it can be considered that the analysis results were in good agreement with the true values.

#### 2.4.2. Verification of virtual spectrum using ISCOS

ISOCS is a counting efficiency calculation program provided by Canberra, a manufacturer of HPGe detectors used in the study, and is applicable only to the well-characterized product, so it was only used to verify the virtual spectrum for the HPGe detector. The considered nuclides were $^{131}$I, $^{134}$Cs, $^{137}$Cs and $^{60}$Co for ground surface measurement, $^{137}$Cs and $^{60}$Co for concrete wall, and $^{58}$Co and $^{60}$Co for radiowaste drum.

The applied geometry was an exponential circular plane for ground surface, simple box for concrete wall, and simple cylinder for the radiowaste drum. Like the verification using the EPS program, the analysis results were compared with the input true values. In the case of the ground surface, as the β value of depth distribution function increases, the relative error tends to increase. When the value of β is 10, it shows a relative error of more than 20% for all nuclides. This is considered to be an error resulting from the implementation of the depth distribution function in the MCNP simulation. Nevertheless, all other results showed relative errors within 20%, confirming the validity of the virtual spectra. In particular, the relative error values for the radiowaste drum measurement were smaller than those for the ground surface or those for concrete wall measurement. This is thought to be because the simulation scale is relatively small in the case of drum measurement. In other words, if the scale of the measurement geometry is

small, the validity of the virtual spectrum created based on the simulation is expected to be high.

## 3. Conclusion

In this study, for training to increase proficiency in gamma spectrometry used in various fields, an attempt was made to build a database that can generate virtual gamma energy spectra for artificial nuclides that are difficult to obtain with actual *in situ* measurement. To this end, a method for deriving virtual gamma-ray energy spectra through MCNP simulation for various preset measurement conditions was developed.

As the detector, HPGe, NaI and $LaBr_3$ detector, which are widely used detectors for *in situ* gamma spectrometry, were selected, and for the measurement situations, ground surface measurement, measurement of concrete walls of decommissioned facilities, and measurement of radiowaste drums were selected.

By combining the simulation results for various gamma emitting nuclides that can be detected in each measurement situation and the actually measured background gamma-ray energy spectrum, the database was structured so that virtual spectrum can be derived according to the artificial nuclides concentration. The generated virtual spectra were validated by comparing the analysis results using ERS program and ISOCS to the true values. As a results, most of the results showed a difference within about 20% of the concentration value for each nuclide input in the virtual spectrum generation, confirming that it is possible to generate a virtual spectrum suitable for simulating a given measurement situation.

As a result of this study, the database that can be used for educational gamma-ray energy spectrum simulators was developed. It is expected that it will be improved to generate a spectrum more similar to the actual measured spectrum and expanded by adding various measurement conditions in the future.

## ACKNOWLEDGEMENTS

This work was supported by KOREA HYDRO & NUCLEAR POWER CO., LTD (No. No. G20IO02)

## REFERENCES

[1] ICRU, Gamma-ray spectrometry in the environment, ICRU Report 53, 1994.
[2] ISO, Measurement of Radioactivity in the Environment – Soil – Part 7: In Situ Measurement of Gamma-Emitting Radionuclides, ISO 185889-7, 2013.
[3] Y.Y. Ji et al., Development of Correction Factors and Their Evaluations in the Aerial Radiological Survey, KAERI/CR-573/2014, 2014.
[4] Y.Y. Ji et al., Development of the Environmental Radiation Survey Program and Its Application to the In-Situ Measurement, KAERI/TR-6876/2017, 2017.