Radioactivity analysis of gamma-emitting radionuclides of environmental samples in a nuclear or radiological emergency

RCA Regional Office

Contents

1. Introduction

1.1. Background

The International Atomic Energy Agency (IAEA) provides general guidelines for assessing the dose of critical groups and populations based on the monitoring of radioactive materials and the environment during nuclear or radiological emergencies in nuclear and radiation-related facilities[1-3]. The purpose of environmental radiation monitoring is to provide information that can aid effective decision-making, such as prompt judgment in emergency situations and initiation of protective actions to safeguard emergency workers and the public. Therefore, in the event of an accident at a nuclear or other facility, environmental sampling and subsequent measurements are performed to provide data on the levels, time dependence, and spatial distribution of radionuclides in various environmental media (such as air, soil, plants, food, etc.) in order to assess the doses to critical groups and populations and to support decisions on protective and mitigation actions[1-3].

The emergency environmental monitoring program involves determining the types of environmental samples, collection points, radionuclides to be monitored, and the frequency of analysis[4]. The program varies based on the characteristics of the nuclear or radiological facility, site characteristics of the environment, type and magnitude of the accident, and the timing of monitoring. Depending on the type of accident, it may be necessary to prioritize the measurement of environmental media and radionuclides that have a greater environmental impact or are of greater exposure assessment significance. For instance, in reactor facilities, radioactive noble gases and oxides; in nuclear fuel facilities, uranium or plutonium released by fire, explosion, or leakage; and in criticality accidents, fission products, neutrons, and gamma rays. Therefore, in order to evaluate the radiation impact from the facility to the environment and detect unexpected situations, each country that operates the facility establishes and implements a radiation environmental monitoring plan during normal operation of the facility, and prepares for abnormal emissions and accidents by establishing radiation disaster prevention measures and emergency monitoring plans.

Table 1 summarizes the IAEA guidelines, which should be considered as a simplified framework for emergency monitoring. A unique program should be established that is appropriate for the operational characteristics of the facility in a given country, taking into account the site characteristics, emission levels, and relevant radionuclides. The intensity and duration of the monitoring activities should be determined by the severity of the emergency, which may range from a few days to several years. As the situation evolves, a phased monitoring program should be established.

Table 1. Environmental Monitoring of Radionuclides to be Performed Following Emergencies (IAEA RG-S-1.8)[4]

Release	Monitored constituents	Frequency	Remarks
Airborne	Measurements during the passage of a cloud		
	External radiation Gamma dose rate Neutron dose rate (if neutron radiation is foreseen)	Continuous Continuous	Near and far field, external dose rate map Only near field, if neutrons are expected
	Air Air Rain	Continuous collection, measurement every 2 h Continuous collection, measurement every 2 h	Near and far field Near and far field
	Measurements after the passage of a cloud		In contaminated areas
	External radiation Gamma dose rate	Continuous	external dose rate map
	Deposition Soil	Once	Contamination map for relevant radionuclides
	Foodstuffs/ingestion Leafy vegetables Milk Other vegetables and fruits Grain Meat Drinking water and/or ground water	Daily Daily At harvest At harvest Representative samples Representative samples	Good indicator for plant food Good indicator for animal food
Liquid	After release		Affected areas and water bodies are limited
	Aquatic dispersion Surface water Sediment	Continuous sampling, daily measurement Weekly	
	Aquatic foodstuffs Fish Shellfish	Selected samples Selected samples	
	Aquatic indicators Seaweeds	Selected samples	

Table 2. Monitoring Quantities and Measurement Guidance (IAEA RG-S-1.8)[4]

Quantity to be measured	Sampling/measurement	Application
Source monitoring		
Gamma dose rate at the source	Stationary on-line equipment, continuous measurement	Practice, emergency
Gases in released air	Stationary on-line equipment, continuous measurement	Practice, emergency
Aerosols in released air [a]	On-line equipment and/or sampling; nuclide specific analysis, total alpha and total beta	Practice, emergency
Activity in released water [a]	On-line equipment and/or sampling; nuclide specific analysis, total alpha and total beta	Practice, emergency
Environmental monitoring		
Gamma dose rate over ground	Field measurements; mobile or stationary devices	Practice, emergency, chronic (prolonged) exposure
Aerosol activity in air	Filter sampling; nuclide specific analysis	Practice, emergency, chronic (prolonged) exposure
Radioiodine in air	Sampling specific to physical and chemical form; nuclide specific analysis	Practice, emergency
Activity in rain	Sampling in rain collector; nuclide specific analysis	Practice, emergency
Deposited activity	In situ gamma spectrometry; planchet sampling and nuclide specific analysis	Practice, emergency
Activity in soil	In situ gamma spectrometry; field sampling and nuclide specific analysis	Practice, emergency, chronic (prolonged) exposure
Activity in foodstuffs and feedstuffs, waters, sediment	Field sampling; nuclide specific analysis	Practice, emergency, chronic (prolonged) exposure

[a] If the discharge limits for a practice are given in terms of total alpha activity and/or total beta activity, and not for specific radionuclides, radionuclide specific measurements on a routine basis may not be necessary

1.2. Objective

The aim of this handbook is to assist ASEANTOM Member States in effectively conducting radioactivity analyses of gamma-emitting radionuclides on environmental samples during nuclear and radiological emergencies. The handbook provides practical guidelines for the radioactivity analysis of gamma-emitting radionuclides in environmental samples, with a focus on laboratory analysis[5]. ASEANTOM Member States are recommended to establish procedures appropriate to the circumstances of their analytical laboratories in each country, based on this guide.

2. Considerations in radiological emergencies

2.1. Gamma-ray spectrometer

During normal times, radioactivity measurement devices for gamma-emitting radionuclides should be deployed and prepared for use in the event of an emergency. It is crucial that these devices are capable of achieving the targeted measurement uncertainty despite need for fast measurements, as a large number of samples may need to be analyzed during an emergency. To prepare for environmental samples that may be contaminated with high levels of radioactivity during emergencies, appropriate measures must be taken. Additionally, it is essential to prepare for situations where there are insufficient samples from the environment.

2.2. Environmental sample collection

The collection of environmental samples must be based on the type of sample to be collected, with the aim of, ensuring that the collected samples are representative of the target sample. As a result, it is crucial to determine which parts of the environmental sample should be collected and how they should be collected, depending on the type of sample used.

2.3. Sample preparation

Samples collected from the environment and transported to the laboratory must be prepared for rapid measurement. This can be achieved by placing them in prescribed volumes in reference containers after minimal pretreatment. It is crucial to prevent cross-contamination between samples and laboratory contamination, particularly contamination of measuring devices.

2.4. Measurement of Samples

In the case of an emergency, sample measurements are likely to be performed within a short period. However, it is essential to ensure that the target or requested measurement uncertainties are met. The uncertainty of measurements performed under normal conditions, such as calibration, is directly affected by the measurement time (spectral accumulation time).

2.5. Analysis of Measurement Results

In some cases, the results obtained by analyzing the spectra from the measurements of the samples may need to be corrected. To do so, correction factors for each contaminating radionuclide should be prepared for the selected measurement conditions.

2.6. Reporting results

To ensure that the measurement and analysis results are effectively utilized by relevant groups when making judgments, it is important to report them in accordance with metrological notations. Additionally, the recording and reporting should include details on sampling and measurement conditions.

3. Measurement equipment

3.1. High-purity germanium (HPGe) detector

High-purity germanium (Ge) detectors are commonly employed to measure the radioactivity of gamma-emitting radionuclides in samples. To detect gamma-rays with energies higher than 30 keV, p-type Ge detectors are typically used. This is because, while measuring lower-energy gamma rays, the X-rays generated during radioactive decay can cause coincidence summing with the gamma rays, which complicates post-measurement analysis. However, n-type detectors can also measure lower energies by placing an appropriate absorber between the detector and the sample to eliminate X-rays below a certain energy level.

To shield the detector from external radiation, it is necessary to install a shield around it. When placing the detector inside the shield, it is important to maximize the distance between the detector and the sample, as long as it does not compromise the external radiation shielding effectiveness. This will enable an appropriate relative dead time to be obtained, particularly in emergency situations involving highly contaminated samples.

Figure 1. (a) A shield installed with the detector, and (b) increased distance between the detector and the sample

3.2. Containers for environmental samples

To ensure representativeness of the sample being measured, the container holding the sample must have a sufficient volume (e.g., soil in a particular area) when preparing it for measurement in the laboratory. If required, environmental samples can be homogenized adequately before measuring. However, even in highly contaminated samples, selecting unreasonably small sample volumes can compromise the representativeness of the measurement. For a typical case with low contamination levels, it is recommended to use a large container with a volume of 1 L or more to hold sufficient samples and optimize measurement efficiency. In emergency situations where contamination levels are high, and the relative dead time of the measuring device is outside the appropriate range (e.g., approximately 10%), the relative dead time can be reduced by increasing the distance between the detector and the sample.

For highly contaminated samples and situations where smaller amounts of samples are collected in the environment, it is advisable to select additional cylindrical containers that can hold approximately one-tenth of the sample volume of the large containers.

Figure 2. Containers for environmental samples

3.3. Calibration sources

When calibrating the measuring device, it is essential to use a radioactive certified reference material that has the same quantity as the measurement sample in the containers selected previously. The certificate supplied with the certified reference material should contain information such as the radionuclides present in the calibration source, the radioactivity of each radionuclide, and the level of uncertainty associated with the calibration.

Figure 3. Certified reference materials

3.4. Sample holder

To ensure consistency in the measurement, a sample holder must be present inside the shield. This holder is used to position the calibration source or the prepared measurement sample under the same conditions (i.e., the same relative position of the sample with respect to the detector). It is recommended that the sample holder is located at the bottom of the shield to avoid exerting any direct force on the detector body. The sample holder must also be designed to return to the same position if temporarily removed for any reason. To prevent contamination of the inside of the shielding and the detector

by the calibration source and measurement sample, it is crucial to ensure that the sample does not come into direct contact with the detector. To facilitate decontamination in case of partial contamination, the surface of the detector and shielding should be protected with plastic wrap. For the sample holder, it is advisable to use a plastic material with low density.

Figure 4. A sample holder made of plastic

4. Detector calibration

4.1. Selection of calibration source

The calibration of a gamma-ray spectrometer is crucial in identifying the energy of gamma rays emitted by an unknown radionuclide in a sample and in calculating the radioactivity concentration of the radionuclide in the sample from its count rate. To achieve accurate calibration, the calibration source must contain a sufficient level of radionuclides that emit gamma rays over the appropriate range of energies necessary for energy and efficiency calibrations of the detector. Typical calibration sources used for gamma-ray spectroscopy should contain radionuclides that emit gamma rays in the energy range 50–2000 keV, covering a wide range of gamma ray energies.

When the calibration source is not in the same medium as the measured sample, the self-attenuation effect of the sample must be corrected. This

is because the gamma rays emitted by the radionuclide in the sample will be attenuated as they travel through the sample, reducing the count rate at the detector. Most calibration sources are produced and distributed only in water, which is not representative of all environmental media. Therefore, it is important to correct for the self-attenuation effect of the calibration source medium (usually water) and the measured sample medium using existing data or computer calculations.

When more than one sample container is used, and if the correction factor cannot be obtained with sufficient accuracy based on the target measurement uncertainty using computer calculations, a calibration source for each container should be prepared.

Each laboratory should have a quality control procedure for the detector, considering the characteristics of the detector and the measuring conditions. The quality control procedure should cover the operation of the detector, and include calibration and inspection.

4.2. Measurement of calibration source

If a calibration source with the same geometry and volume as the container required for sample preparation is available, the calibration should be at the selected detector-to-sample distance. The same calibration procedure should be repeated for other detector-to-sample distances using the same sample container. Additionally, calibration should be performed for other sample containers using the appropriate calibration sources to the containers, to the extent possible. In case a calibration source is not available, the measurement efficiency of the sample container and the detector-to-sample distance should be ensured through calculations such as computer simulations. When calibrating at different detector-to-sample distances using the same calibration source, the spectral accumulation times should differ (they

should be longer for longer distances) to achieve the desired uncertainty.

Figure 5. Calibration source and sample located at the same distance from the detector

Once the calibration source is selected and the detector-source distance (calibration point) is determined, the counts of each gamma ray of interest should be measured to ensure that they are sufficient to achieve the targeted calibration accuracy. The uncertainty of the measurement efficiency is mainly determined by the uncertainty of the certified reference material and the net count of each gamma ray of interest.
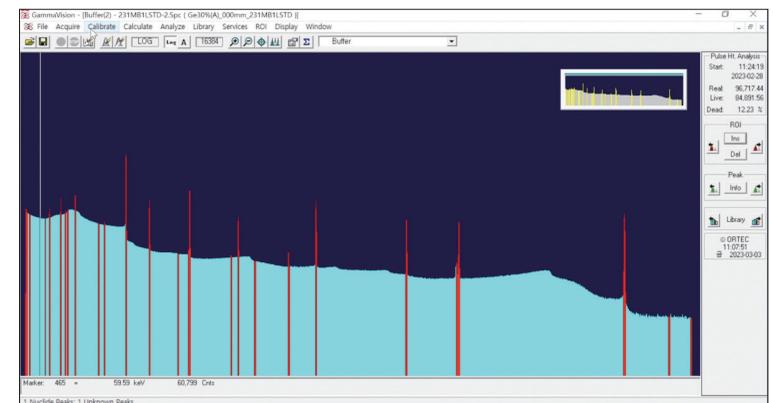
Figure 6. Spectrum of a calibration source

4.3. Energy calibration and energy resolution

The energy calibration procedure typically involves acquiring data using a calibration source that contains radionuclides emitting the gamma rays at known energies and correlating those energies with the multichannel analyzer (MCA) channels acquired by the data acquisition (DAQ) system. This requires using the reference data of the radionuclides[6] in the calibration source. The energy calibration procedure can be summarized as follows:

Energy calibration procedure

① Import a calibration source measurement spectrum
② Build and apply a gamma-emitting radionuclide library for calibration using reference data
③ Delete existing calibration results, if necessary
④ Perform an energy calibration (automatically or manually)
⑤ View and save energy calibration results

The energy calibration curve is typically a straight line, as shown in the figure 7. The difference between the linear fit and the measured data should be within ±0.15% for satisfactory results. After completing the energy calibration, it is important to check the energy resolution (FWHM), which generally increases proportionally with energy. A difference between the energy-resolution fitting result and the measured data within ±2% is considered satisfactory. If the result is unsatisfactory, then the DAQ measurement signal should be tuned separately.
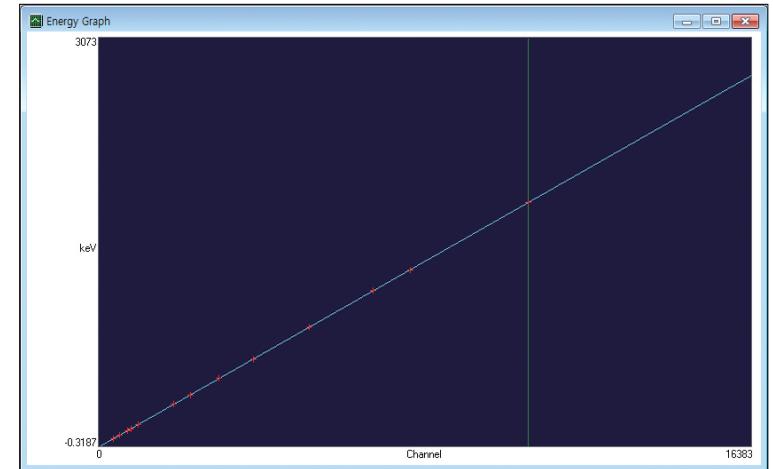
Figure 7. Energy calibration
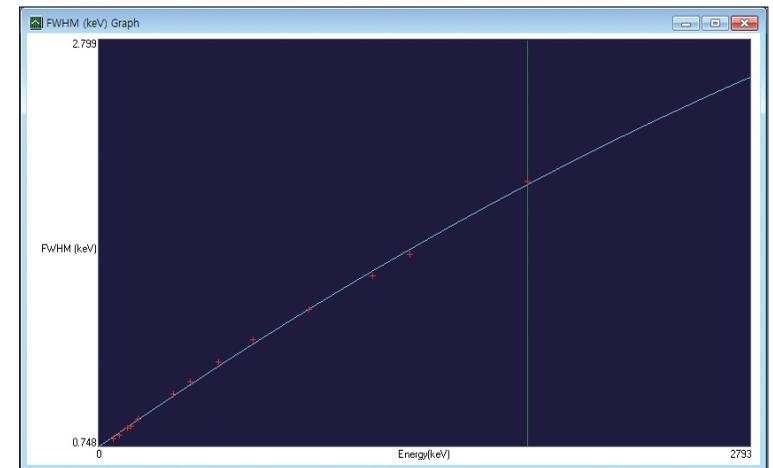
Figure 8. Energy resolution

4.4. Efficiency calibration

After completing energy calibration, the next step is to calibrate the gamma-ray detection efficiency of the HPGe detector. This requires data acquired from a calibration source, along with its calibration certificate, and a library file created during the energy calibration. The efficiency calibration procedure is as follows:

Efficiency calibration procedure

① Import the measured spectrum of the calibration source
② Apply the radionuclide library of the calibration source created during energy calibration
③ Verify that the radionuclides in a given peak are properly recognized
④ Enter the reference date of the certificate, radioactivity of the radionuclide, and relative standard uncertainty of the radioactivity by considering the units on the certificate.
⑤ Calculate peak efficiency by specifying an appropriate efficiency fitting curve function
⑥ View and save efficiency curve fitting results

After the efficiency calibration, it is important to check the appropriateness of the efficiency fitting curve function and examine the difference between the calculated and fitted efficiencies. In cases where the calibration source contains gamma-emitting radionuclides with a true coincidence summing effect, it is necessary to correct the efficiency of the peak. The figure below shows a typical example of the efficiency correction result.
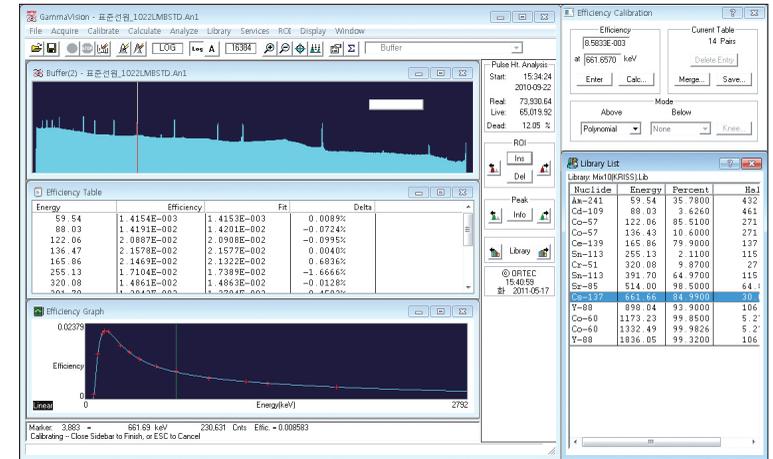
Figure 9. Efficiency calibration

The following shows the model equation of efficiency calibration and uncertainty evaluation. The model equations can also be used to perform the calculations and can be applied to the efficiency calibration results.

Efficiency Calibration Model Equation

• Calibration Efficiency

$$\varepsilon_k = \frac{C_k/t_L}{A_0 P_k} f_1 \cdots f_n$$

• Net count correction factor

$$f_1 = \frac{ln\,2\,t_R/T_{1/2}}{1 - e^{-ln2t_R/T_{1/2}}}$$

• Decay correction factor to the reference time, t_c

$$f_2 = e^{ln2(t_c - t_0)/T_{1/2}}$$

• True coincidence summing correction to k-gamma

$$f_3^k = \sum_j f_3^{kj}(P_k, \varepsilon_k, P_j, \varepsilon_j^T)$$

where

ε_k : Detection efficiency of k-gamma

A_0 : Certified radioactivity of k-gamma

P_k : Emission probability of k-gamma

C_k : Net count of k-gamma

t_L : Measurement live time

t_R : Measurement real time

$T_{1/2}$: Half-life of radionuclides

$f_1, \cdots, f_n^k$: Correction factors for k-gamma

t_0 : Reference time for certificates

t_c : Reference time for the calibration

Uncertainty Evaluation

$$r(\varepsilon_k) = [r^2(C_K) + r^2(t_L) + r^2(A_0) + r^2(P_k) + r^2(f_1) + r^2(f_2) + \cdots + r^2(f_n)]^{1/2}$$

$$r(f_1) = \left(1 - \frac{\ln 2\, t_R/T_{1/2}\, \varepsilon^{\ln 2\, t_R/T_{1/2}}}{1 - e^{-\ln 2 t_R/T_{1/2}}}\right)[r^2(t_R) + r^2(T_{1/2})]^{1/2}$$

$$r(f_2) = \frac{\ln 2\,(t_c - t_0)}{T_{1/2}}[r^2(t_c - t_0) + r^2(T_{1/2})]^{1/2}$$

4.5. Correction of true coincidence summing effect

Calibration sources used in gamma ray spectroscopy often contain ^{60}Co and ^{88}Y, which emit multiple gamma rays with a high emission probability. When two or more gamma rays with sufficient energy to be detected are emitted simultaneously from a single radionuclide and are detected, the detector cannot distinguish between them and records the combined energy, leading to a reduction in the count rate of each individual gamma ray. This reduction is greater at shorter distances between the detector and source. If the reduction is outside the targeted uncertainty range, the measurement efficiency of the affected gamma rays must be compensated. To compensate for the true coincidence summing effect of ^{60}Co and ^{88}Y in the calibration source, a correction factor can be obtained through computer simulations or using the shape of the calibration efficiency curve.
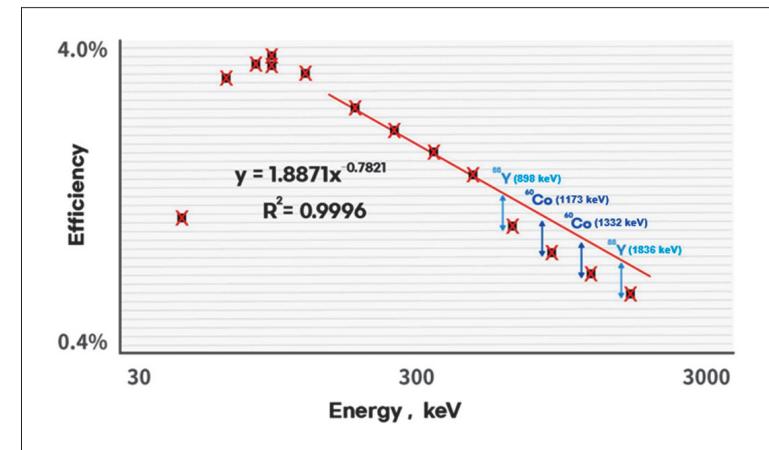
Figure 10. Correction method for true coincidence summing effects

In this manual, we present a method to compensate for the true coincidence summing effects in calibration sources for gamma-ray spectroscopy using the shape of the efficiency curve, which is plotted

as energy. For gamma-ray energies above approximately 200 keV, the logarithmic efficiency, plotted as logarithmic energy, shows a straight line. This enables us to extend the efficiency of gamma rays emitted by radionuclides without true coincidence summing effects in the range 200–700 keV up to 2000 keV, with some reduction in true coincidence summing effects at energies above 700 keV.

Commercial computational programs, such as GESPECOR[7], that use a combination of semi-empirical and Monte Carlo simulations, and full Monte Carlo simulation codes, such as Geant4[8] and PENELOPE[9], can be used to calculate the peak efficiency, total efficiency, self-attenuation, and true coincidence summing effect of HPGe detectors.
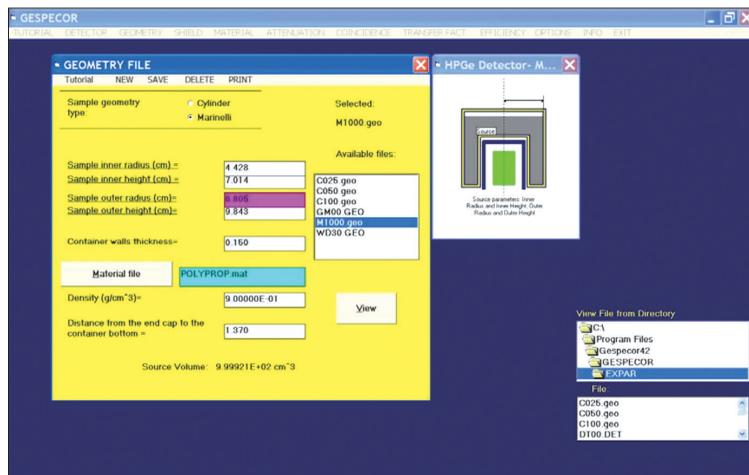
Figure 11. GESPECOR simulation

Figure 12. Geant4 simulation

5. Environmental sample preparation

5.1. Considerations for sample preparation

The sample preparation area should be separated based on the contamination level of the samples, into areas for low- and high-concentration samples. Disposable plastic should cover the work area, such as the floor or table, to prevent contamination and the area should be sanitized after any activity that could lead to contamination. All necessary tools should be prepared in advance, and workers must adhere to radiation protection rules for both internal and external exposure. In a logbook, the amount (mass or volume) of the environmental sample brought to the laboratory, the amount of the main sample after impurities removal, and the amount (mass or volume) of the net sample in the sample container should be recorded, along with information related to sample preparation, such as date, time, operator, and sample type.

The prepared sample should be refrigerated before measurement, if necessary, and then wrapped in a clean plastic bag or plastic wrap before being placed in the detector. The measurement room, where the detector is installed must be separated from the sample preparation workspace. Once the measurement is completed, the sample is stored or disposed of in accordance with the regulations.

5.2. Sample preparation procedure

The measurement samples should be prepared through an appropriate homogenization process for each sample. Then transfer the homogenized sample to a measuring container up to the specified volume.

• Soil and sediment

The soil(or sediment) first removes grass or other organic matter before manually picking up big stones and gravel. Use a sieve to do constitution if there are several tiny substances.

• Food

In an emergency, food samples are targeted at vegetables and fruits that may be contaminated. In the early stages of the accident, surface contamination of food is a major concern for radiation internal contamination of the target plant in the late stage. Since food may have been exposed to air deposition, sampling should be done on the upper section of the plant, or the edible part, and at locations such as marketplaces and distribution facilities. For foods, only the edible parts are sampled and then homogenized using a grinder.

Liquid samples are assumed to be homogenized and no further homogenization is required.

• Water

If the sample is to be stored for a certain period of time, add 11 M of hydrochloric acid at a ratio of 10 mL per liter of sample before or after sampling to prevent radioactive isotopes from being adsorbed onto the wall of the container.

To avoid cross-contamination, extreme radioactive contamination in water samples must be separated from other samples.

• Milk

In consideration of measurement time, milk samples are treated with preservatives to separate whey or prevent spoilage.

Measurement Sample Preparation Procedure

① Pretreatment of environmental sample

② Homogenization of environmental sample

③ Transfer of sample to a container
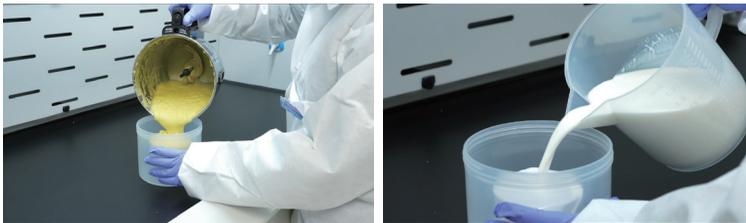
④ Addition of preservatives

⑤ Weighing (사진 교체 예정)

⑥ Sealing

⑦ Labeling
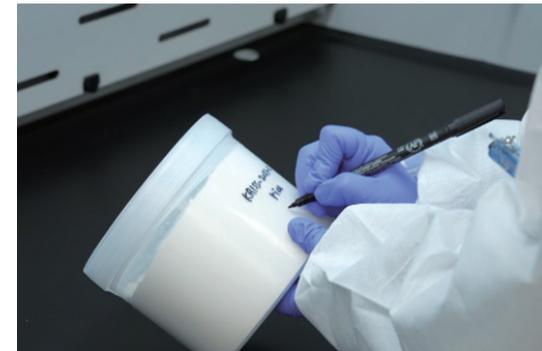
⑧ Storing of samples

⑨ Recording in Worksheet
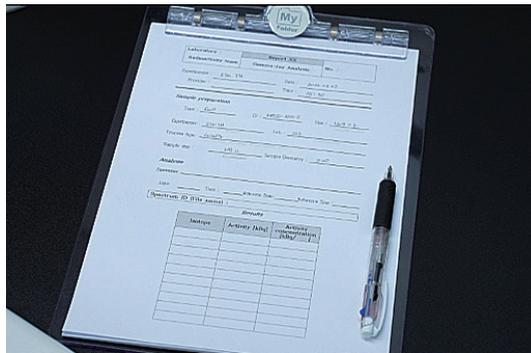
6. Sample measurement

6.1. System checking

In general, the position of the gamma-ray peak can shift, which can affect the analysis results of the spectrum. If a shift in the position of the target gamma-ray peak is identified (e.g., more than FWHM/3 or more than 2 keV of the gamma-ray peak), energy calibration should be performed again. The presence or absence of a peak shift can be determined by comparing the existing positions of specific gamma rays emitted by the background radiation or long-half-life radionuclides of the calibration source.

6.2. Background measurement

The measurement time for the sample spectrum was set in the range 100–1,000 s in case of an emergency. Prior to the sample measurement, a background radiation spectrum was taken without any sample in the shielding. The background spectrum should be saved with a unique number for future reference and calibration of the sample measurements, and all relevant information must be recorded in the logbook.

6.3. Sample loading

To determine the measurement point in the shield, which is dependent on the detector-source distance, the point that maximizes the measurement efficiency (as close as possible to the detector–source distance) is first selected based on the specific sample container. The detector is then calibrated using a calibration source in the same container. This measurement point is used when the radioactivity level of the sample to be measured is low and high measurement efficiency is required. If the radioactivity concentration of the prepared sample is high, an additional measurement point is selected at a distance

from the detector to the sample, where the measurement efficiency is reduced by approximately 1/10 compared to the measurement point at a near distance. For sample containers with different capacities, the measurement points are selected and calibrated in the same manner when preparing for emergency measurements.

6.4. Sample measurement procedure

Before measuring the prepared sample, the detector used for the measurement should be calibrated for energy and efficiency according to the measurement conditions, such as the sample container, sample volume, and detector-to-sample distance. Once the calibration is complete, the settings and adjustments of the device and the measurement geometry should not be changed. If more than one measuring device is used, appropriate measures must be taken to ensure that each measuring device is not contaminated by the sample, and to distinguish between different levels of sample contamination. Therefore, it is essential for each laboratory to have a working procedure that reflects the detectors and measurement geometries used and to record all information about the sample and measurement conditions in a logbook.

The target value of the minimum detectable activity (MDA), which depends on the detector's measurement efficiency and time, is 1 Bq or less for major radionuclides (e.g., 661 keV for ^{137}Cs and 364 keV for ^{131}I), and the target measurement uncertainty for major radionuclides is 10% or less.

Sample Measurement Procedure

① In the detector's logbook, record at least: 1) the sample type and unique number, 2) the sample container and detector geometry, 3) the sample mass and medium, 4) the date and time of sample collection, and 5) the background spectrum.

② To prevent contamination of the detector, wrap the detector part of the shielding in plastic wrap. Place the container with the sample at the selected position, set the measurement time, and initiate the measurement. Check the relative dead time, and if it exceeds 10%, change the position to a previously calibrated position with lower measurement efficiency and perform the measurement.
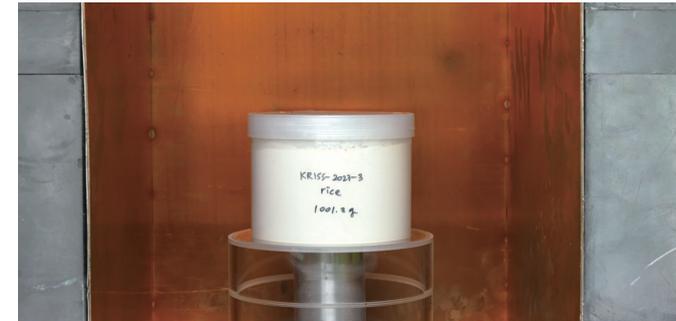
③ Save the measured spectrum with the ID of the sample and record the necessary information, such as the live time of the measurement, in the logbook. Check and record anomalies in the spectrum and peak shapes.
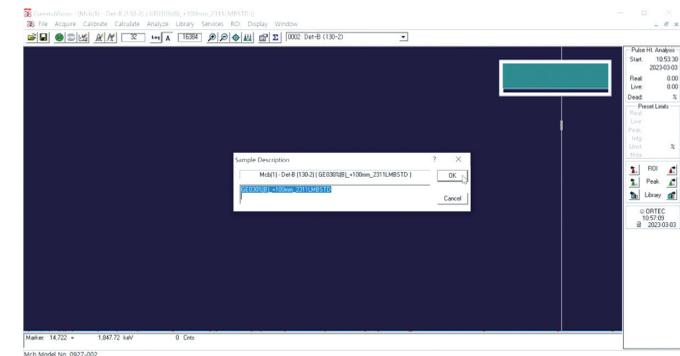
④ Repeat for all prepared samples.

7. Radioactivity analysis

7.1. Analysis of measurement results

Most laboratories use computer programs for gamma-ray spectroscopy to facilitate calibration and sample analysis. The process of analyzing a measured sample spectrum in these programs involves several steps, including (1) applying an energy calibration to the sample and background spectra to subtract the gamma-ray count measured in the background spectrum from the sample spectrum, (2) obtaining the area (net count) of the user-specified gamma-ray peak, and (3) applying an efficiency calibration to calculate the radioactivity of the specified radionuclide. In this process, the radioactivity concentration of each radionuclide is automatically calculated using the measurement time (real-time and real-time) stored in the spectrum, half-life of each radionuclide provided by the gamma-ray library, probability of emission of gamma rays emitted by each radionuclide specified by the user, date and time of sampling and measurement, and amount (mass or volume) of the measurement sample entered by the user. Additionally, these programs enable various fine calibration functions and inputs for uncertainty calculations.

To facilitate calibration and analysis, computer programs for gamma-ray spectrometry create and use a gamma-ray library in the database that contains information on radionuclides, including their name, half-life, and uncertainty, as well as information on gamma rays emitted by the radionuclides, including gamma-ray energy, emission probability, and probability uncertainty. This gamma-ray library is built for the analyzed contaminating radionuclides expected to be contained in the sample and the gamma rays emitted by those radionuclides. Once the gamma-ray library for sample analysis is established, the computer program

for gamma-ray spectroscopy can easily obtain the radioactivity of the contaminating radionuclides in the measured sample by applying the calibration results to the program.

Each laboratory should have a well-defined procedure that outlines the calibration and sample analysis processes specific to the gamma-ray spectrometry computer program used in the laboratory. Furthermore, gamma-ray libraries for calibration and sample analysis should be routinely created for each gamma-ray spectrometry computer program used.

The following is the model equation for determining radioactivity by gamma rays.

Gamma-Ray Specific Radiation Model Equations

• Radioactivity of k-gamma

$$A_k = \frac{C_k/t_L}{\varepsilon_k P_k} g_1 \cdots g_n$$

• Net count correction factor

$$g_1 = \frac{\ln 2\, t_R/T_{1/2}}{1 - e^{-\ln 2 t_R/T_{1/2}}}$$

• Decay correction factor to the sampling time, t_s

$$g_2 = e^{-\ln 2(t_s - t_m)T_{1/2}}$$

• True coincidence summing effect correction to k-gamma

$$g_3^k = \sum_j g_3^{kj}\left(P_k, \varepsilon_k, P_j, \varepsilon_j^T\right)$$

• Self-attenuation correction factor $\left(g_4^k\right)$

• Measurement geometry correction factor $\left(g_5^k\right)$

where,

A_k : Radioactivity of k-gamma

ε_k : Detection efficiency of k-gamma

P_k : Emission probability of k-gamma

C_k : Net count of k-gamma

t_L : Measurement live time

t_R : Measurement real time

$T_{1/2}$: Half-life of radionuclides

$g_1, \cdots, g_n$: Correction factors of k-gamma

t_M : Reference time for sample measurement

t_s : Reference time for sample collection

If the identified radionuclide emitted multiple gamma rays, its radioactivity is to be determined using the weighted average of the gamma rays.

Multiple gamma-emitting radionuclide radioactivity and uncertainty

• Radioactivity of multiple gamma-emitting radionuclide
$$A = \frac{\sum_{k=1}^{n} A_k / u_k^2 (A_k)}{\sum_{k=1}^{n} 1/u_k^2 (A_k)}$$

• Standard uncertainty of radioactivity of multiple gamma-emitting radionuclide
$$u(A) = \frac{1}{\sqrt{\sum_{k=1}^{n} 1/u_k^2 (A_K)}}$$

7.2. Corrections

7.2.1. Coincidence summing

The measurement sample may also contain radionuclides that simultaneously emit two or more gamma rays, such as [134]Cs, which have a true coincidence summing effect similar to the calibration sources. The true coincidence summing effect is higher at closer detector-to-sample distances, but the true coincidence summing effect usually decreases to 3% or less at detector-to-sample distances of 10 cm or more.
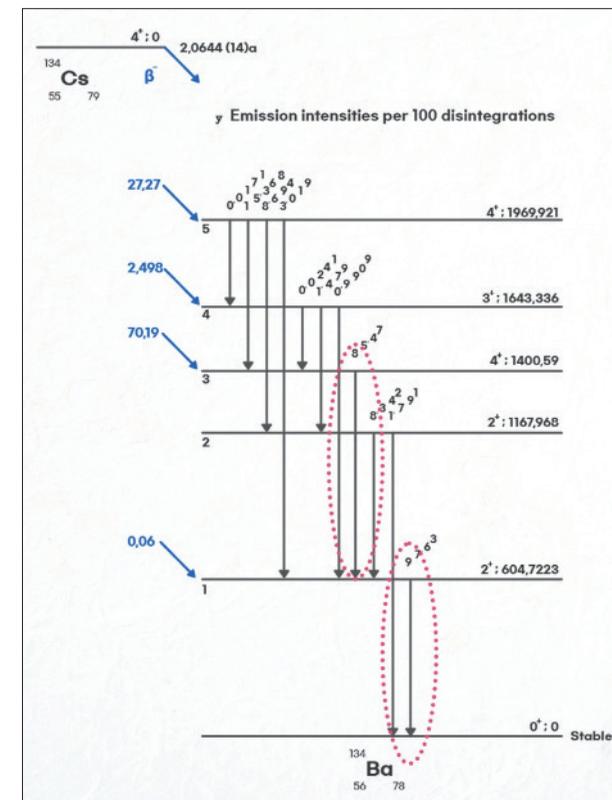
Figure 13. Decay scheme of [134]Cs[6]

7.2.2. Self-attenuation in a Media

Sometime, it is difficult to obtain calibration sources that use the same medium as the measurement sample. Hence, relative self-attenuation between the measurement sample and calibration source is often unavoidable. If the effect of media differences is sufficiently significant to prevent reaching the desired measurement uncertainty, then it may be necessary to compensate for the attenuation effect to achieve the target uncertainty.

The relative attenuation of the calibration source medium (usually water) by the medium of the measured sample depends on the distance between the detector and the sample. As the distance increases, the effect of attenuation decreases. This is because at a closer distance, the detector detects a wider range of anisotropic gamma rays emitted by the sample, whereas at a farther distance, the detector detects only a narrow range of isotropic gamma rays. This effect is most pronounced in the Marinelli beaker samples, where the thickness of the medium through which the gamma rays pass varies significantly.
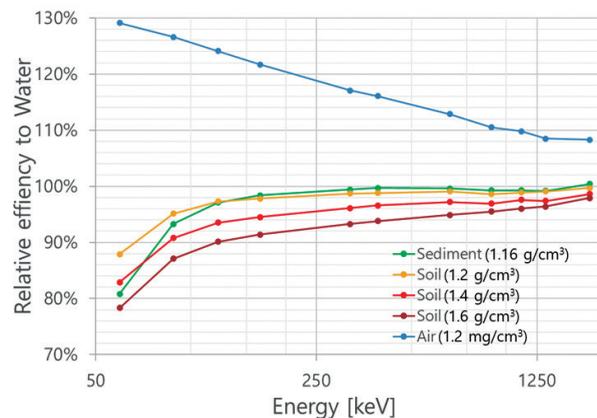
Figure 14. Self-attenuation effect for various media

8. Recording results

8.1. Recording information

8.1.1. Sampling information

- Worker's name
- Date of sample collection
- Sampling points
- Types of samples
- Sample collection methods
- Sample preparation type
- Measurement labs

8.1.2. Calibration information

- Worker's name
- Detector type & ID
- Calibration source
- Measurement geometry,
- Calibration date/time
- Calibration result

8.1.3. Measurement

- Worker's name
- Sample ID
- Measurement geometry
- Measurement date/time
- Measurement result

Figure 15. Worksheet sample[2]

8.2. Notation of data

All units used for recording and reporting are in the SI system. If necessary, they are indicated separately in parentheses.

The measurement results are recorded and reported as (measured value) $\pm$ (measurement uncertainty), and minimum detectable activity (MDA). The measurement uncertainty is approximately 95% confidence level, $k = 2$. If no radioactivity is measured, the measurement is reported as "< (MDA)" or "Not detected".

9. References

[1] IAEA-TECDOC-955 "Generic assessment procedures for determining protective actions during a reactor accident", IAEA (1997)

[2] IAEA-TECDOC-1092 "Generic procedures for monitoring in a nuclear or radiological emergency", IAEA (1999)

[3] IAEA-GSG-2 "Criteria for Use in Preparedness and Response for a Nuclear or Radiological Emergency", IAEA (2011)

[4] IAEA RS-G-1.8 "Environmental and Source Monitoring for Purposes of Radiation Protection", IAEA (2005)

[5] KINS/HR-1556 "Development of Procedure for Gamma-Emitting Nuclides in Emergency", KINS (2017)

[6] LNHB, 2023 Nuclear Data – Table – Laboratoire National Henri Becquerel, LNHB Webpage. http://www.lnhb.fr/nuclear-data/nuclear-data-table/, Accessed 30 Jan. 2023.

[7] Sima, O., Arnold, D. & Dovlete, C. "GESPECOR: A versatile tool in gamma-ray spectrometry", Journal of Radioanalytical Nuclear Chemistry 248, 359–364 (2001)

[8] S. Agostinelli et al. "Geant4 - A Simulation Toolkit", Nucl. Instrum. Meth. A 506 (2003) 250-303

[9] J. Sempau, E. Acosta, J. Baro, J.M. Fernandez-Varea and F.Salvat "An algorithm for Monte Carlo simulation of the coupled electron-photon transport" Nucl. Instrum. Meth. B 132 (1997) 377-390.

[10] Young In Scientific Co., Ltd. (2022) "GammaVision User Manual" Domestic education material.

[11] Neosiskorea Co., Ltd. (2023) "GENIE2000 Gamma Acquisition and Analysis" Domestic education material.

Appendix A. Utilizing GammaVision[10]

1. Preparation of Nuclide Library for Calibration

1) Run the S/W 'GammaVision' to prepare the nuclide library for calibration.

2) Click 'Library', 'Edit', and 'GammaVision Editor...' in order in the S/W.
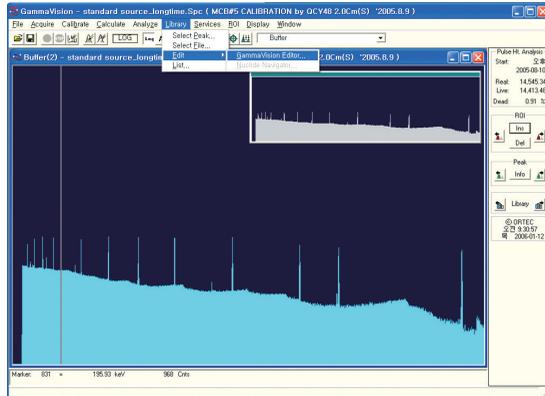
3) Copy the existing nuclide library most suitable for calibration and load it in the 'Editing:' window.
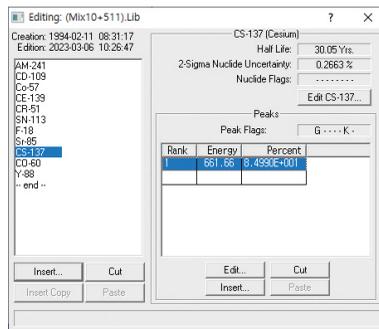
4) Modify the nuclide library by referring to the CRM certificate and the nuclear reference data designated by your institution.

5) When the modification of the nuclide library is completed, save it for calibration.

2. Preparation of Nuclide Library for Sample Analysis

1) Run the S/W 'GammaVision' to prepare the nuclide library for calibration.

2) Click 'Library', 'Edit', and 'GammaVision Editor...' in order in the S/W.
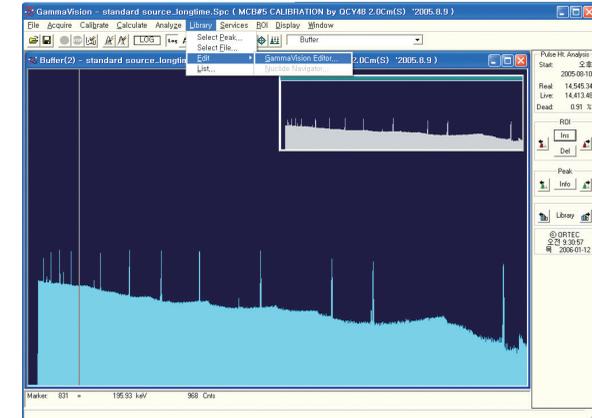
3) Copy the existing nuclide library most suitable for sample analysis and load it in the 'Editing:' window.
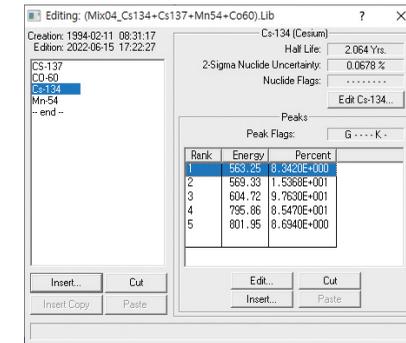
4) Modify the nuclide library by using the information on samples.

5) When the modification of the nuclide library is completed, save it for sample analysis.

3. Energy Calibration

1) Check whether the measurement system is operating normally.

2) When the system is ready, place the certified reference material (CRM) on the source holder.

3) Run the S/W 'GammaVision' to take a gamma-ray spectrum for CRM.

4) Determine the measurement time (live time) so that the net count of the gamma-ray peak of interest is greater than 100,000.

① Click 'Aquire' and then 'MCB Properties' in the S/W menu.
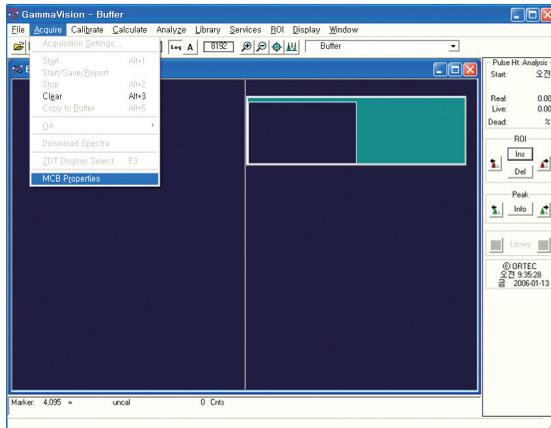
② Click 'Presets' tab in the 'Properties' window and enter the live time.
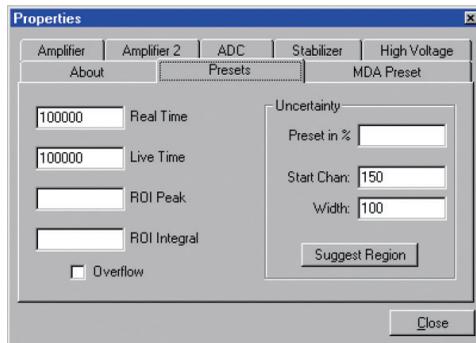
③ Make the distance between the detector and CRM the same as when measuring the sample.

5) Start data collection and obtain a gamma ray spectrum for calibration.

6) When data taking is finished, the spectrum for calibration is saved.

7) Load the nuclide library for calibration by clicking 'Library' and 'Select File…' in turn.

8) Press the 'Energy…' item in the 'Calibrate' menu to bring up the 'Energy Calibration' window.

9) Press the 'Auto Calibrate' button in the 'Energy Calibration' window to perform energy calibration.

10) Click the 'Energy' or 'FWHM' item in the 'Fit' area of the 'Energy Calibration' window to check the energy and energy resolution (FWHM) calibration results.

11) When energy calibration is completed by closing the 'Energy Calibration' window, the 'Energy…' item of the 'Calibrate' menu is checked.

12) Save the calibration result by clicking 'Save Calibration…' in the 'Calibrate' menu.

4. Efficiency Calibration

1) Click 'Calibrate' and 'Efficiency…' in turn. To calculate efficiencies of gamma-rays of interest, open the nuclide library for calibration and specify the corresponding gamma ray peak.
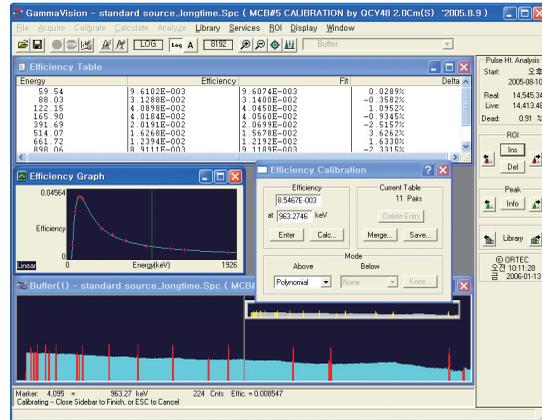
2) Click 'Calc…' in the 'Efficiency Calibration' window.

3) Fill in the blanks of the 'Efficiency Calculation Worksheet' using the CRM certificate.
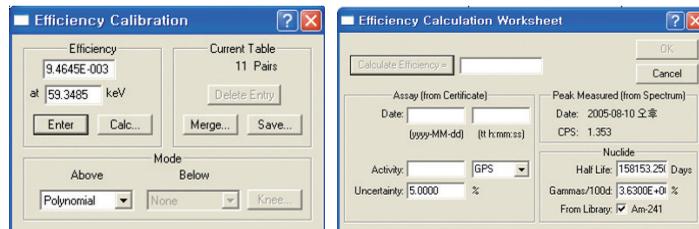
4) Calculate the efficiency of the gamma-ray by clicking the activated 'Calculate Efficiency =' button.

5) Then click the activated 'OK' button to accept the efficiency calculation result.

6) Calculate the efficiency for the remaining gamma-rays of interest in the same way.

7) After completing efficiency calculations for all gamma rays of interest, select a function formula (e.g. 'Polynomial') in the 'Mode' area of the 'Efficiency Calibration' window to create an efficiency fit curve.

8) After exiting the 'Efficiency Calibration' window, click 'Efficiency…' of 'Calibrate' again to check whether the efficiency fit curve was created well.

9) If energy and efficiency calibration is completed, 'Energy…' and 'Efficiency…' items should be checked when 'Calibrate' menu is clicked.

10) Save the calibration result by clicking 'Save Calibration…' in the 'Calibrate' menu.

5. Counting and Analysis for Sample Spectrum

1) Check whether the measurement system is operating normally.

2) When the system is ready, place a sample on the source holder.

3) Run the S/W 'GammaVision' to acquire the gamma-ray spectrum for the sample.

4) Determine the measurement time to satisfy the target MDA or measurement uncertainty.

5) When the measurement is complete, save the gamma ray spectrum of the sample.

6) Recall the saved sample spectrum using the 'Recall…' menu of 'File'.

7) Apply the saved calibration result using the 'Recall Calibration…' menu of 'Calibrate'.
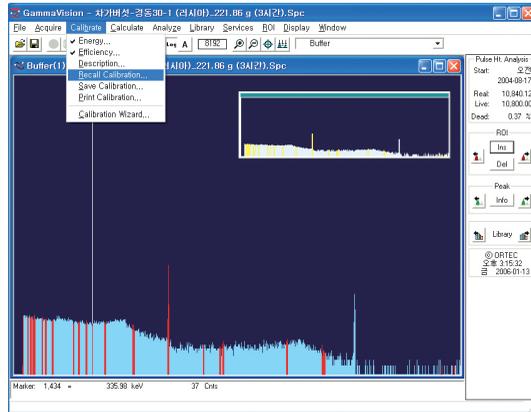
8) Set the analysis conditions by pressing the 'Analyze', 'Settings' and 'Sample Type...' menus in sequence.
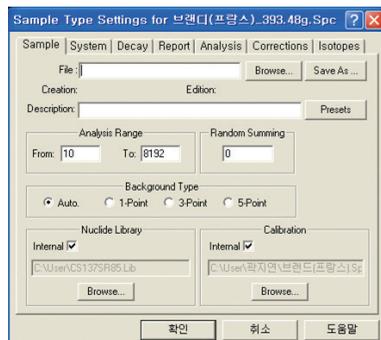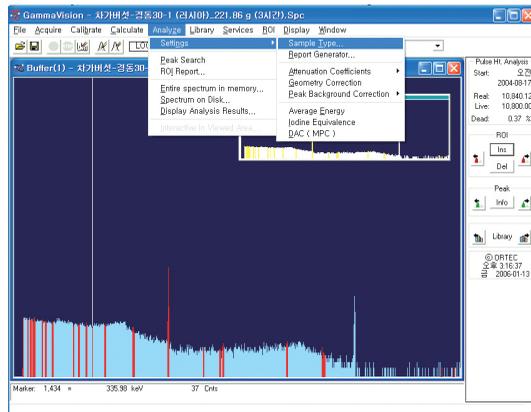
9) Load and apply the nuclide library for sample analysis by pressing the 'Library' and 'Select File...' menus in turn.

10) Analyze the sample spectrum by clicking 'Analyze' and 'Entire spectrum in memory…' in sequence.
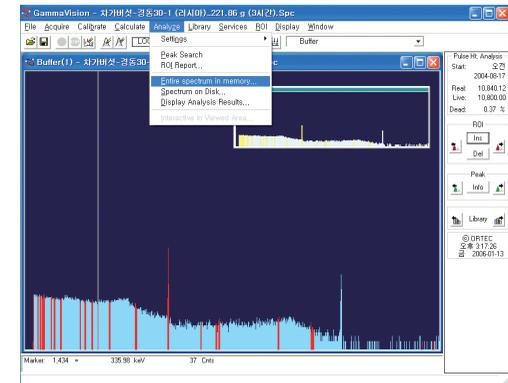
11) Depending on the S/W settings, the analysis results are displayed on the screen or saved.

파일(F) 편집(E) 서식(O) 보기(V) 도움말(H)
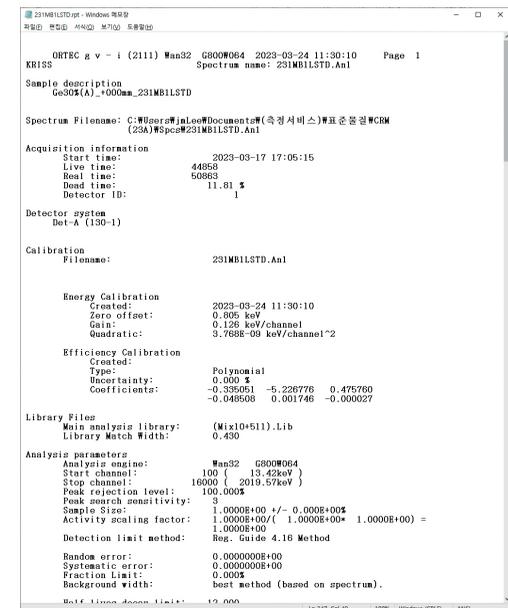
```
AM-241      3.5417E+03   3.7572E-01%   1.5476E+00%   7.967E+01
            3.5417E+03   3.5156E+03    3.5679E+03    3.980E+01    3.757E-01%

CD-109      1.8783E+04   3.5052E-01%   1.5246E+00%   2.912E+02
            1.8783E+04   1.8654E+04    1.8912E+04    1.455E+02    3.505E-01%

Co-57       1.1610E+03   2.4115E-01%   9.9537E-01%   1.033E+01
            1.1610E+03   1.1555E+03    1.1665E+03    5.164E+00    2.411E-01%

CE-139      1.2712E+03   2.5251E-01%   9.3307E-01%   1.058E+01
            1.2712E+03   1.2649E+03    1.2775E+03    5.285E+00    2.525E-01%

CR-51       3.0732E+05   2.4309E-02%   9.9319E-01%   1.011E+02
            3.0732E+05   3.0718E+05    3.0747E+05    5.051E+01    2.431E-02%
```

```
       ORTEC g v - i (2111) Wan32  G800W064  2023-03-24 11:30:10        Page  5
KRISS                              Spectrum name: 231MB1LSTD.Anl

SN-113      3.1704E+03   1.0531E-01%   8.4749E-01%   9.135E+00
            3.1704E+03   3.1639E+03    3.1770E+03    4.563E+00    1.053E-01%

F-18   #    0.0000E+00   1.0000E+03%   1.0000E+03%   0.0000E+00
            0.0000E+00   0.0000E+00    0.0000E+00    0.000E+00    0.000E+00%

Sr-85       5.1746E+03   7.1857E-02%   7.1269E-01%   7.768E+00
            5.1746E+03   5.1673E+03    5.1819E+03    3.880E+00    7.186E-02%

CS-137      1.5494E+03   2.1741E-01%   7.9725E-01%   8.237E+00
            1.5494E+03   1.5428E+03    1.5560E+03    4.113E+00    2.174E-01%

CO-60       2.1717E+03   1.8113E-01%   7.8459E-01%   1.743E+01
            2.1717E+03   2.1640E+03    2.1794E+03    8.707E+00    1.811E-01%

Y-88        5.2688E+03   9.7986E-02%   8.2855E-01%   1.091E+01
            5.2688E+03   5.2587E+03    5.2789E+03    5.449E+00    9.799E-02%


  # - All peaks for activity calculation had bad shape.
  * - Activity omitted from total
  & - Activity omitted from total and all peaks had bad shape.
  < - MDA value printed.
  A - Activity printed, but activity < MDA.
  B - Activity < MDA and failed test.
  C - Area < Critical level.
  F - Failed fraction or key line test.
  H - Halflife limit exceeded
  I - ISO NORM MDA ratio at maximum for one or more peaks

_____   S U M M A R Y   _____
Total Activity (   13.4 to  2019.6 keV)     3.494E+05  Bq/Kg

Analyzed by: _____
                    Lee, Jong-Man

Reviewed by: _____
                    Supervisor

Laboratory:  KRISS
```

Appendix B. Utilizing Genie 2000[11]

All GENIE-2000-related programs must always run the Virtual Data Manager (VDM) process before use. When Windows runs on the PC boot, VDM automatically starts and the corresponding icon is displayed in green. In order to prevent interruption (error) of VDM, screen saver, power saving mode, etc. are not set. Click Start Service if marked in red and contact the engineer if it does not start. And, MCA definition must be performed during the initial installation of equipment or during the replacement of MCA and PC.

1. Nuclide Library Editor

The Nuclide Library Editor program is a program that allows users to generate their own gamma-ray nuclide library files. Create library files for analysis and library files for energy calibration.

1) Run a program (Nuclide Library Editor): Run the Nuclide Library Editor by locating the installed path to generate the necessary nuclide library files.

2) A program for generating a nuclide library file is executed.

3) Click Extract in the Options menu to select the desired nuclide.

4) When the Extract Library Nuclides window opens, click the Open button
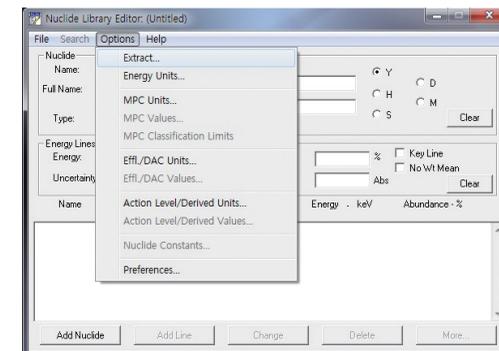
5) Pre-supplied *.Select the STDLIB.NLB file from the NLB file and click the Select button.
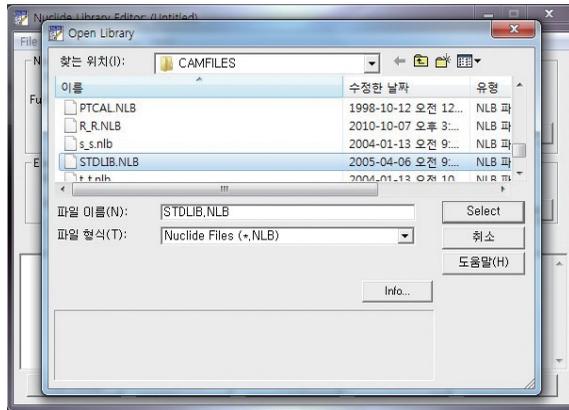
6) Select the desired nuclide in the Extract Library Nuclides window and click the OK button. (multiple selections available)

7) Make sure that all of the selected nuclides are attached to the list pane at the bottom of the program, and then change the half-life, energy, and emission probability shown in the certificate.
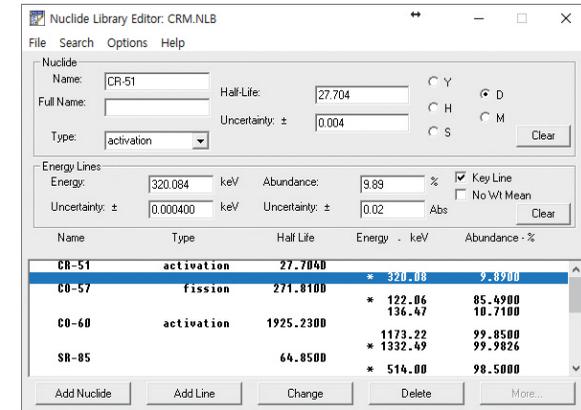
8) Click Save as in the File menu to save the list as a library file.
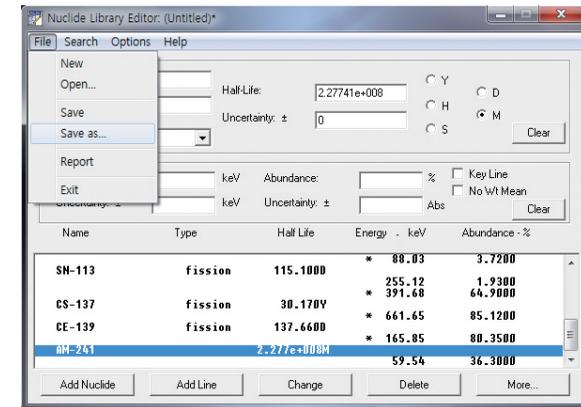
9) Enter the file name (required), Description, and Optional, and press the Save button to save it

10) Once the file is saved, you can check the file name stored at the top of the program.

11) 'Local Disk (C:) ▶ GENIE2K ▶ CAMFILES' You can check the file generated in the corresponding path.

2. Energy calibration

1) Energy full▶By Nuclide List in the File menu for energy calibration...
   Click .
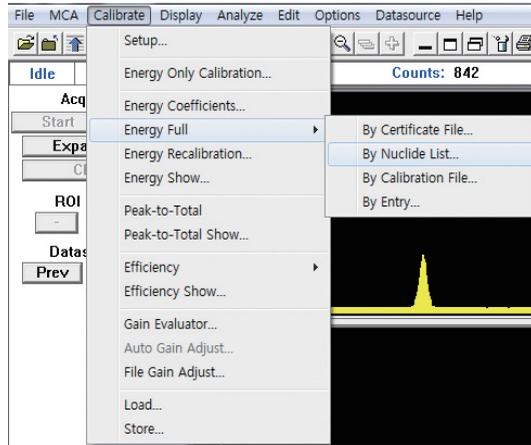
2) Select the pre-created calibration library (*.NLB) file by pressing the
   ▼ button on the Library in the Calibrate by Nuclide List window.

3) Select all Nuclides using the mouse and click the OK button.
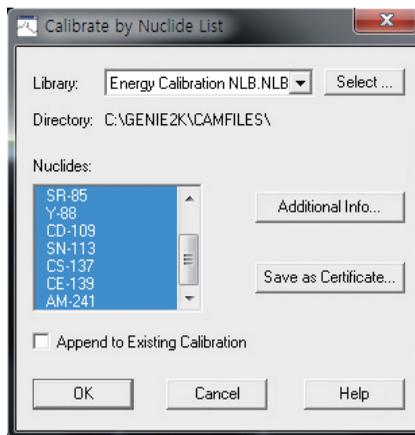
4) Mouse-click on the apex of the peak located at the front of the spectrum to locate the cursor.

5) In the Energy Calibration – Full window, click Energy keV 59.54 and press the Cursor button.
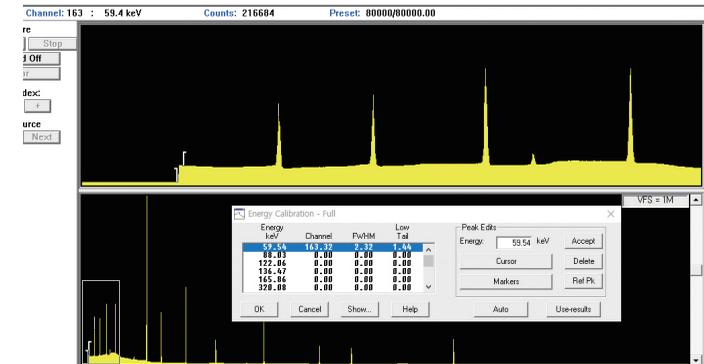
6) Verify that the channel value of the cursor corresponding to 59.54keV is substituted.

   ※ In the case of 59.54 keV (^{241}Am), it corresponds to about 163 channels when it is '8192 channels/3000 keV'.

7) Then, in the same way, mouse click on the apex of the ^{60}Co (1332.5 keV) peak.

8) In the Energy Calibration – Full window, click EnergykeV 1332.49 and press the Cursor button.

9) Verify that the channel value of the cursor corresponding to 1332.49 keV is substituted.

10) Click the Auto button to replace the channel values for the remaining energy. (It doesn't matter if you proceed manually)

11) In the Energy Calibration – Full window, verify that all energy corresponding channel values are substituted.

12) You can check the Energy Calibration Curves by pressing the Show button.

13) If the measured points are well substituted into the calculated line, it can be judged that the energy calibration is satisfactory.
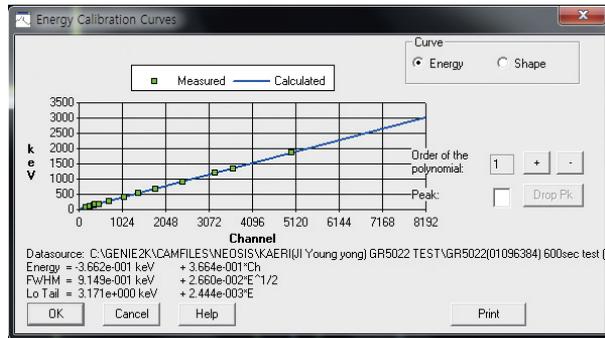
14) Click the OK button, and then click Save in the File menu to save energy calibration data.
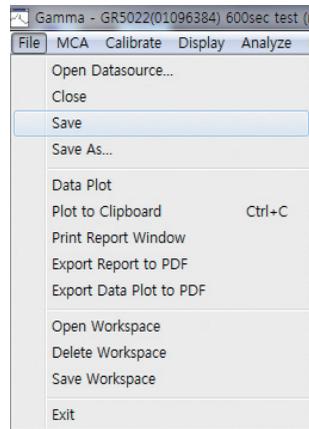
15) The last stored calibration data can be checked by clicking the Energy Show on the Calibrate menu.

3. Certificate File Editor

The Certificate file Editor program is used to generate a reference material certificate file for calibration. This must be done when using the nuclide library files and calibrating efficiency.

1) Run a program (Certificate File Editor): Locate the installed path to generate the required certificate file and run the Certificate File Editor.

2) A program for generating a certificate file is executed.

3) Click Library Extract on the Options menu to select the library file to apply to the file in the certificate.
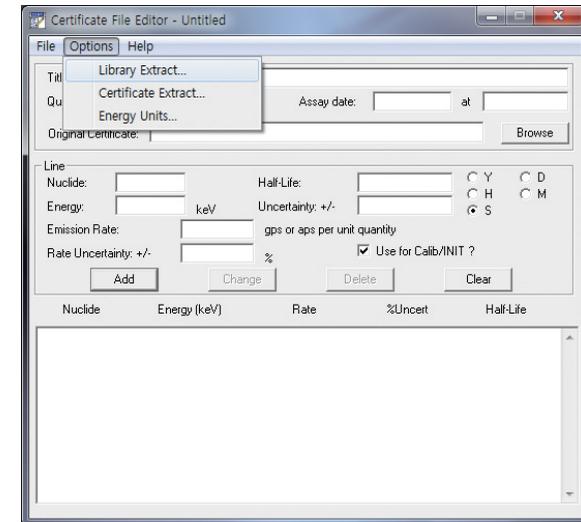
4) Select the library file to apply to the certificate and click the Open button.

5) Select the unit (Bq) listed on the certificate.

6) Apply the radioactivity value (Activity) and % Uncertainty listed on the certificate by pressing the Input Change button.
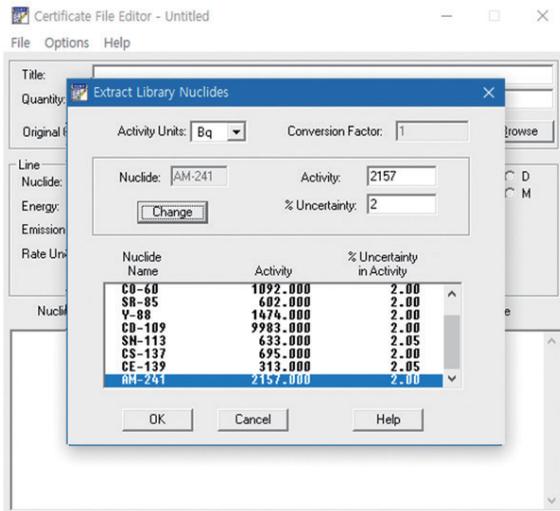
7) Ensure that all applied nuclide library files are attached to the list window.

8) Enter Title, Quantity, and Assay date at the top.

9) Click Save as in the File menu to save it as a certificate file when you have finished filling out everything.

10) Enter the file name and optional description, and press the Save button to save it.

11) When the file is saved, you can check the file name stored at the top of the program.

12) 'Local Disk (C:) ▶ GENIE2K ▶ CAMFILES' You can check the file generated in the corresponding path.

4. Efficiency calibration

1) Obtain a standard source measurement file of not less than 80,000 seconds for efficiency calibration.
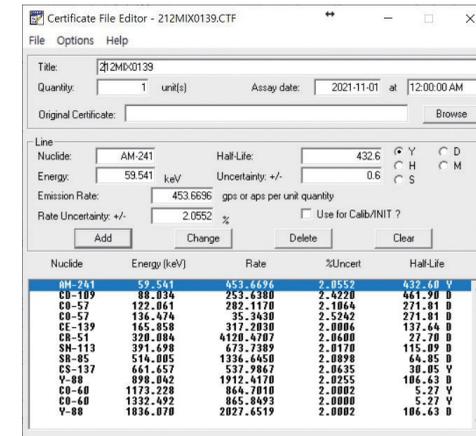
2) Click Efficiency ▶ By Certificate file on the Calibrate menu for efficiency calibration.

3) In the Open Certificate File window, select the pre-created certificate (*.CNF) file for efficiency calibration and open it.

4) Make sure that all calibration energy (keV) is in the list of the Efficiency Calibration window.
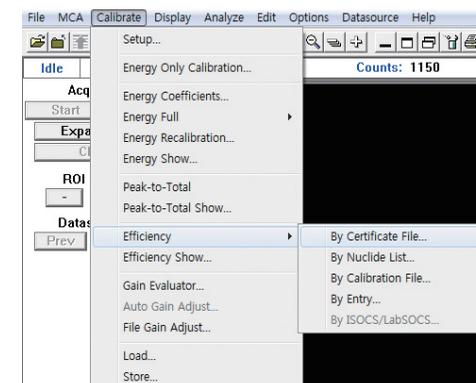
5) Select 136.48 keV to calibrate the low-energy and high-energy regions separately and click the Cross Over button.
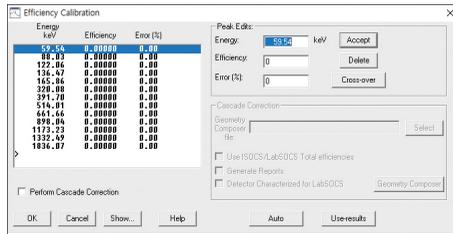
6) Click the Auto button to enter the Efficiency and Error (%) values for all energy.

7) Click the Show button on the Efficiency Calibration. Check the efficiency calibration curve.
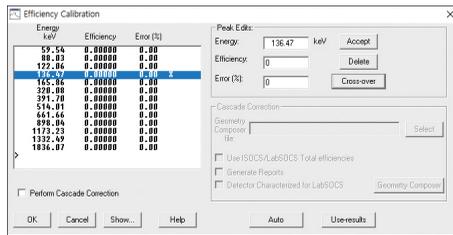
8) Click the List Pks button to check the Deviation value. In this case, if the Deviation (%) value falls within the range of ±5%, it is recognized as a satisfactory value.
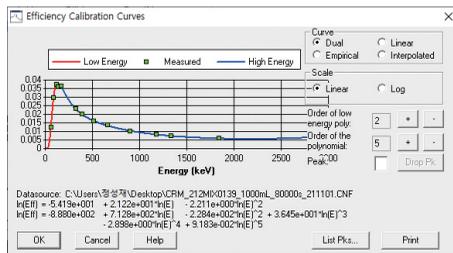
9) Click Store in the Calibrate menu to save efficiency calibration data as a file (*.CAL).

10) File name (required input), Description, Eff in the Store window. Enter Geom. ID and click the Store button.

11) Click Load on the Calibrate menu to apply the saved efficiency calibration file.

12) * to be applied. Select the CAL file and press the Load button to apply the calibration file.

5. Identification of radionuclides

1) Click the Activate Report window icon to check the analysis results.

2) Click Peak Location ▶ Unidentified 2nd Difference in the Analyze menu to find the peak in the measured spectrum
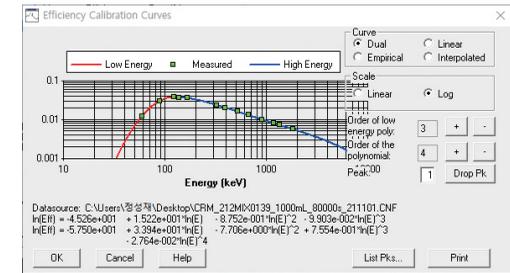
3) Click Peak Area ▶ Sum/Non-Linear LSQ Fit Setup in the Analyze menu to determine the area of the located peak.
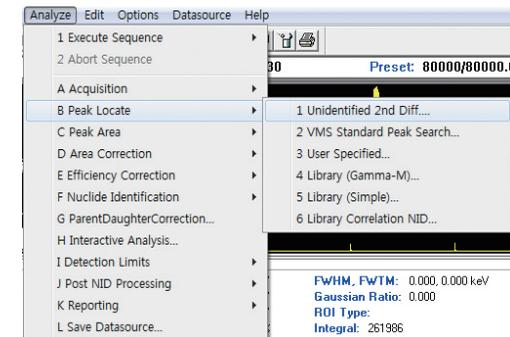
4) When analyzing radioactivity, select the same BKG file as the sample Geometry.
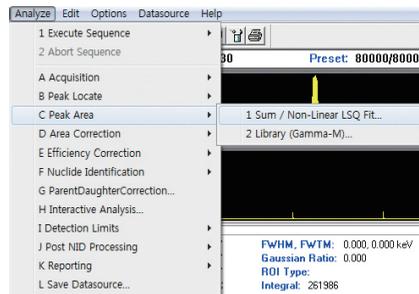
5) Click Efficiency Correction ▶ Standard on the Analyze menu for efficiency correction.

   ※ Dual: Dual efficiency calibration calculations are performed.

   ※ Linear: The linear efficiency correction calculation is performed.

   ※ Empirical: The efficiency correction calculation of the experiment is performed.
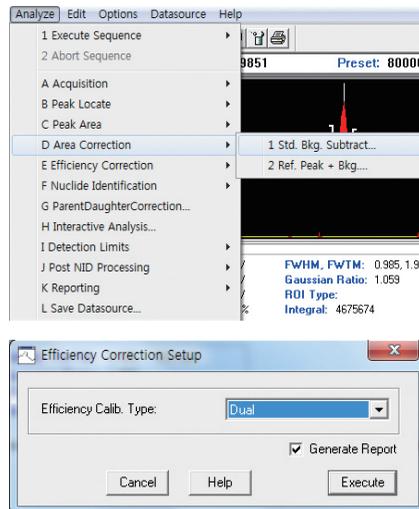
6) Nuclide Identification ▶ NID w/ Interf. Corr··· in the Analyze menu for nuclide discrimination/interference peak correction···click on.

   ※ NID library: Select the library file for analysis.