Activity standardization of ^{134}Cs and ^{137}Cs by 4πβ(LS)-γ coincidence counting method

Agung Agusbudiman[1,2,3*], Kyoung Beom Lee[1,2], Jong Man Lee[1,2], Sang Hoon Hwang[2]

[1]*Korea University of Science and Technology (UST), Daejeon, 34113 Republic of Korea*
[2]*Korea Research Institute of Standards and Science (KRISS), Daejeon, 34113 Republic of Korea*
[3]*National Nuclear Energy Agency (BATAN), Jakarta, 12440 Republic of Indonesia*

*Corresponding author's e-mail: agusbudiman@batan.go.id

Abstract. A primary activity standardization of ^{134}Cs and ^{137}Cs has been performed at the Korea Research Institute of Standards and Science (KRISS) by 4πβ(LS)-γ coincidence counting method to provide a reference standard for activity measurement of the radionuclides. For the case of ^{134}Cs, the method can be applied directly to determine its activity value. The complex decay scheme of ^{134}Cs allows more than one gamma window setting to be used for the standardization. In this experiment, five different gamma windows are used to standardize the activity of ^{134}Cs. The mean activity values from these measurements are used as the final activity value, which gives a value of (1145.6 ± 5.2) kBq/g at the reference time. For the case of ^{137}Cs, the long decay time of its gamma emissions makes the 4πβ(LS)-γ coincidence counting method can not be applied directly to determine its activity value. As a solution, we applied the efficiency tracing technique into this method by using the standardized ^{134}Cs as the tracer. The final activity value of ^{137}Cs obtained from this standardization is (963.5 ± 9.4) kBq/ g at the reference time. The quoted uncertainty in all results is evaluated at $k = 1$.

KEYWORDS: Activity standardization, ^{134}Cs, ^{137}Cs, 4πβ(LS)-γ coincidence, efficiency tracing method

1 INTRODUCTION

Caesium-134 (^{134}Cs) and Caesium-137 (^{137}Cs) are two important radioisotopes widely used in both the research and industrial fields. For example, in most nuclear accidents, these isotopes are often used as parameters for radiation contamination in the environment, marine organisms, and foodstuffs. The ratio of these two isotopes in the environment was also utilized to identify the reactor units that caused atmospheric releases during the Fukushima nuclear accident [1]. In terms of activity measurement, the two isotopes are very important for the calibration of γ-ray spectrometer and the study of coincidence summing on gamma spectrum analysis. The importance and the wide application of these two isotopes make the activity standardization of these isotopes has become essential to provide an accurate and traceable activity measurement of these isotopes.

The 4πβ-γ coincidence counting method is one of a well-known method that is widely used for primary activity standardization of radionuclides, particularly for a β - γ emitter. This method can be directly applied to standardize the ^{134}Cs since the nuclide emits both β and γ promptly. The ^{134}Cs disintegrates mainly by beta minus to the excited states of ^{134}Ba with a half-life of 2.0644(14) years. Three main beta emissions are decays to several excited states of ^{134}Ba with end-point energy of 89.06 keV (27.27%), 415.64 keV (2.49%), and 658.39 keV (70.19%). The most dominant beta emission decays to the excited states of ^{134}Ba with end-point energy of 658.39 keV (70.19%), followed by a dominant gamma emission decay with the energy of 795.86 keV (85.47%) to another excited state of ^{134}Ba. Another dominant gamma emission is the 604.72 keV (97.63%), which decays to the ground state of ^{134}Ba [2]. Due to the complex decay scheme of ^{134}Cs, A.Rytz suggested that the standardization is performed based on the γ-window around the 605 keV photopeak, around the peaks at 796 and 802 keV, or around the peak at 1365 keV as the other γ-windows may give more difficult results to interpret [3].

In the case of ^{137}Cs, the nuclide disintegrates with a half-life of 30.05 (8) years by β-ray emissions. The emissions occur 94.36% to the excited level of ^{137m}Ba with end-point energy of 513.97 (17) keV and 5.46% to the ground state of ^{137}Ba with end-point energy of 1175.63 (17) keV. The meta-stable ^{137m}Ba further decays with a 661.6 keV γ-ray emission to the ground state with a decay time of 2.552 minutes [4]. This long decay time makes the time correlation between the 513.97 keV β-ray and the 661.6 keV γ-ray emission events nearly completely disappeared. Therefore, for the case of activity measurement, ^{137}Cs

needs to be treated as a pure β-ray emitter that makes the conventional 4πβ-γ coincidence method cannot be applied directly to determine its activity. However, with some additional techniques, i.e., efficiency tracing technique, the coincidence counting method can still be used to standardize the activity of ^{137}Cs.

The efficiency tracing technique can be applied to the activity standardization of ^{137}Cs by adding another nuclide with a known activity amount as a tracer into the solution of ^{137}Cs. To be used as the tracer, the other nuclide should be able to be measured using the 4πβ-γ coincidence method with sufficient accuracy. It also needs to have a low self-absorption and have a similar characteristic with the ^{137}Cs, i.e., similar β-spectrum shape and β end-point energy and the mix of the nuclides must be homogenous [5]. Considering the above requirements, in this experiment, we use ^{134}Cs as the tracer to determine the activity of ^{137}Cs by the 4πβ-γ coincidence method.

The following sections will describe the application of the 4πβ(LS)-γ coincidence counting method for activity standardization of ^{134}Cs and ^{137}Cs with efficiency tracing technique.

2 EXPERIMENTAL METHOD

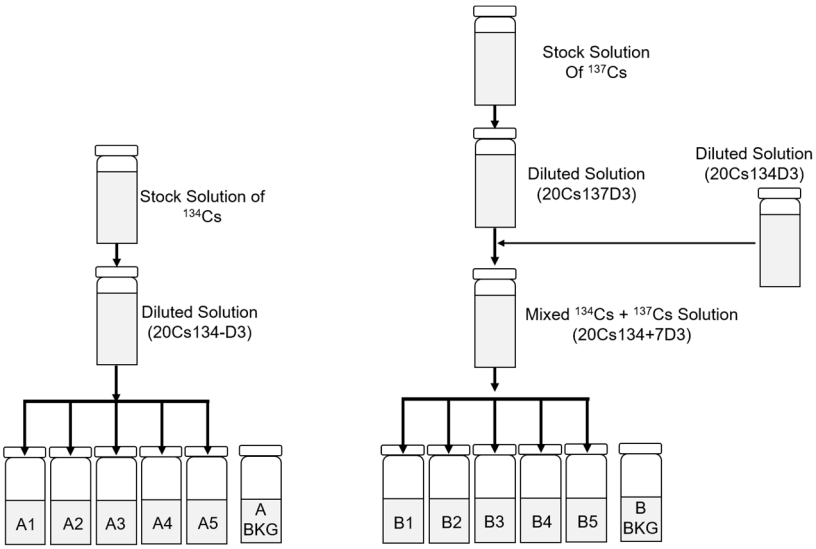
2.1 Measurement system

The 4πβ(LS)-γ coincidence counting system is used to standardizes the activity of ^{134}Cs and ^{137}Cs. The system comprises a light-tight optical chamber that can accommodate a standard 20-mL LSC vial, and a detector system consists of two Burle 8850 photomultiplier tubes (PMT) and two 2-inch NaI(Tl) detectors for beta and gamma detection. For data acquisition, the digital sampling technique for 4πβ(LS)-γ coincidence counting (DCC) is applied to reduce the data acquisition time and optimize the data analysis using digital data processing. Detail of the hardware configuration and DCC technique is explained in [6] and [7].
In this experiment, the high voltages on both gamma detectors were set at +750 V, and for the PMT, the high voltages were set at +2.61 kV and +2.62 kV with focus voltage set at +700V. The shaping time for both gamma and beta channels were set at 3μs with the threshold level set at 30 and 100 mV for beta and gamma, respectively. Data acquisition was conducted for 5 minutes for each LS sample and 10 hours for the background measurement. Raw data obtained from measurements were analyzed using the analysis routine explained in [7].

2.2 Sample preparation

A stock solution of ^{134}Cs and ^{137}Cs were prepared for activity measurement with the 4πβ(LS)-γ coincidence counting system. The stock solution of ^{134}Cs was gravimetrically added into a 10 mL of UG solution prepared in a 20 mL high-performance glass vial for activity measurement of ^{134}Cs. For activity determination of ^{137}Cs by efficiency tracing method, a mix solution of ^{134}Cs and ^{137}Cs was prepared using the same method. The ratio of ^{134}Cs and ^{137}Cs is 1:1. Five LS samples were prepared in this experiment for each ^{134}Cs and ^{137}Cs measurement. Each sample series comprised one sample without an active solution to measure the background counting rate, which was then subtracted. Due to the stock solution's high activity, dilution processes were performed before preparing the LS samples. A schematic diagram of the sampling process is shown in Fig. 1.

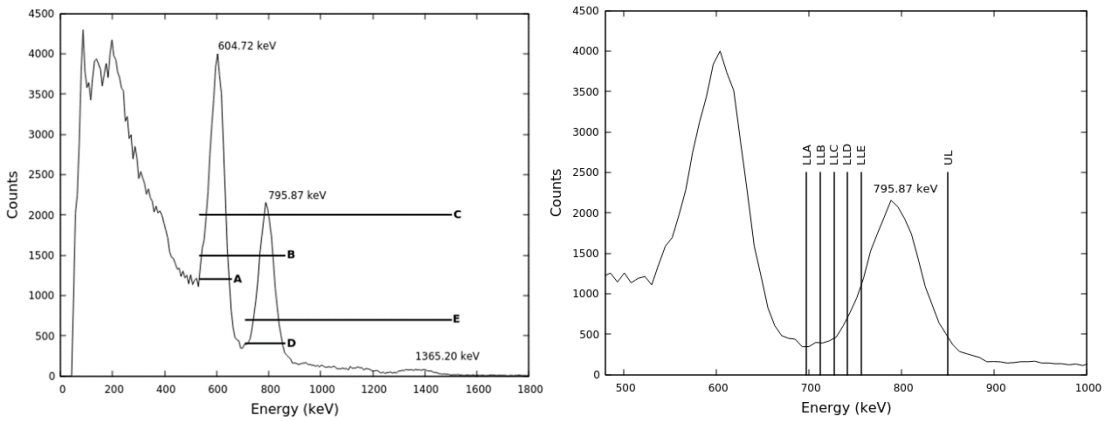
Figure 1: Schematic diagram of sample preparation for ^{134}Cs and mix of ^{134}Cs and ^{137}Cs.



2.3 Standardization of ^{134}Cs

The standardization of ^{134}Cs was conducted based on five different gamma window settings, as shown in Fig. 2., and the final value is determined from the mean of activity value obtained from all gamma window settings. For the purpose of the tracing method used in activity standardization of ^{137}Cs, the effect of the lower level (LL) of gamma window settings was also studied by conducting measurements with different LL setting on gamma 795.87 keV.

Figure 2: Gamma window settings for activity measurement of ^{134}Cs (left) and LL settings for gamma 795.87 keV (right).



For a complex decay scheme like ^{134}Cs, the β-efficiency extrapolation method is required for determining the activity value. The basic equations used for the β, γ, and coincidence channels are as follow:

$$\rho_\beta = A \sum a_r \left[\varepsilon_{\beta r} + \frac{(1 - \varepsilon_{\beta r})(\alpha \varepsilon_{\text{œ}} + \varepsilon_{\beta\gamma})}{1 + \alpha} \right] \quad (1)$$

$$\rho_\gamma = A \sum a_r \, \varepsilon_{\gamma r} \quad (2)$$

$$\rho_c = A \sum a_r \{ [\varepsilon_{\beta r} \varepsilon_{\gamma r}] + (1 - \varepsilon_{\beta r}) \varepsilon_{\alpha r} \} \quad (3)$$

Where :

A = activity of the source

ρ_β, ρ_γ, ρ_c = counting rates for the beta, gamma, and coincidence channel, respectively (corrected for background, dead-time, and resolving-time).

ε_β, ε_{ce}, $\varepsilon_{\beta\gamma}$ = beta detector efficiency for the beta, conversion electron, and gamma emission, respectively.

α = total conversion coefficient

$\varepsilon_{\gamma r}$ = efficiency of gamma detectors at r^{th} branch

ε_{cr} = probability of recording the coincidence when the associated beta particle is not detected.

2.4 Standardization of ^{137}Cs.

The activity standardization of ^{137}Cs was performed at the gamma window D setting to avoid the contribution of the 661.7 keV gamma decay of ^{137}Cs. In principle, the activity of ^{137}Cs, A_7, is determined from the β-count rate of ^{137}Cs, $\rho_{\beta 7}$, multiply by its efficiency, ε_7, with correction for conversion electrons' contribution and unconverted photons, C. The quantitative addition of ^{134}Cs into ^{137}Cs allows us to approximate the ε_7 value as a function of the efficiency (or inefficiency) of ^{134}Cs.

From the measurement of the mixed solution, the β-count rates of ^{137}Cs, $\rho_{\beta 7}$, is then determined by subtracting total β-count rate, $\rho_{\beta m}$, from the measurement with the β-count rate of ^{134}Cs, $\rho_{\beta 4}$.

$$\rho_{\beta 7} = \rho_{\beta m} - \rho_{\beta 4} \quad (4)$$

$$\rho_{\beta m} - \rho_{\beta 4} = A_7[f_2(1-\varepsilon_4) + C] \quad (5)$$

$$\rho_{\beta m} - A_4\, f_1(1-\varepsilon_4) = A_7[\, f_2(1-\varepsilon_4) + C\,] \quad (6)$$

The use of the digital sampling technique in this experiment allows us to consider the decay ratio between the two nuclides to be constant during the measurement time. Therefore, the efficiency function can be approached as a single polynomial. Since the value of ε_4 can be obtained from the measurement of mix source as $\rho_{cm}/\rho_{\gamma m}$, makes the above equation becomes:

$$\rho_{\beta m} - A_4\, f_1\left(1 - \frac{\rho_{cm}}{\rho_{\gamma m}}\right) = A_7[\, f_2\left(1 - \frac{\rho_{cm}}{\rho_{\gamma m}}\right) + C\,] \quad (7)$$

If we normalized with the β-efficiency of the mix source, $\varepsilon_m = \rho_{cm}/\rho_{\gamma m}$, the efficiency extrapolation becomes:

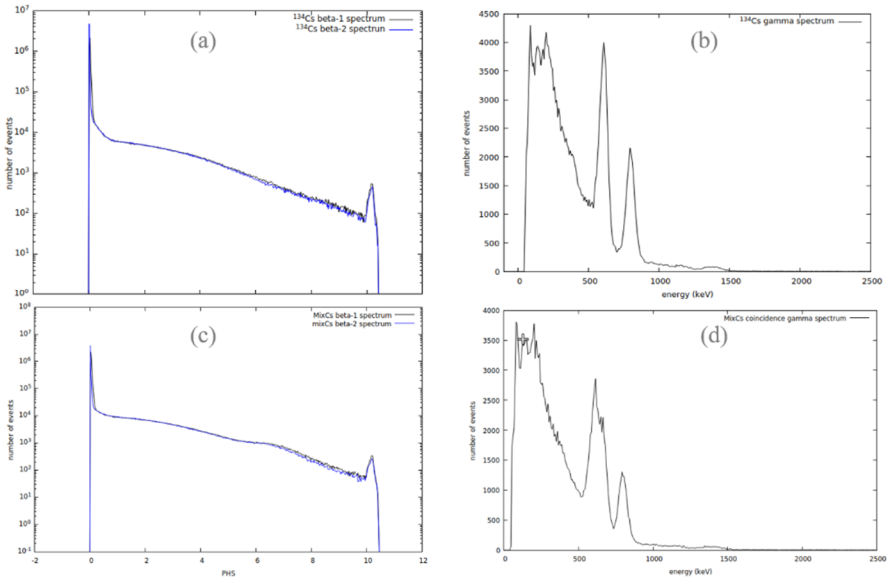
$$\frac{\rho_{\beta m}\rho_{\gamma m}}{\rho_{cm}} - A_4\, f_1\left(\frac{1-\frac{\rho_{cm}}{\rho_{\gamma m}}}{\rho_{cm}/\rho_{\gamma m}}\right) = A_7[\, f_2\left(\frac{1-\frac{\rho_{cm}}{\rho_{\gamma m}}}{\rho_{cm}/\rho_{\gamma m}}\right) + \frac{C\,\rho_{\gamma m}}{\rho_{cm}}\,] \quad (8)$$

Extrapolating the left side of the equation above in terms of $\left(\frac{1-\frac{\rho_{cm}}{\rho_{\gamma m}}}{\rho_{cm}/\rho_{\gamma m}}\right)$ to the limit $\rho_{cm}/\rho_{\gamma m} \to 1$ will then give the $A_7[1+C]$. The value of A7 is then obtained by dividing the extrapolated value to the factor [1+C].

3 RESULTS AND DISCUSSION

Figure 3. shows the typical beta and gamma spectrum obtained from the measurement of the ^{134}Cs sample (fig.3a and fig.3b) and the mix ^{134}Cs and ^{137}Cs sample (fig.3c and fig3.d). The contribution of the ^{137}Cs in the mixed sample can clearly be seen from both the beta and gamma spectrum.

Figure 3: Typical beta and gamma spectrum of ^{134}Cs (a,b) and mix of ^{134}Cs and ^{137}Cs samples (c,d).



Applying the data processing and analysis routine in [7] to the raw data above, the β-efficiency curve is obtained. The least-square method is used to fit the curve to obtain the activity value. The fitting range covers around 20% of β-efficiency, with the maximum β-efficiency obtained in this measurement is 86%. The efficiency curve is shown in Figure 4.

Figure 4: The β-efficiency curve of ^{134}Cs.

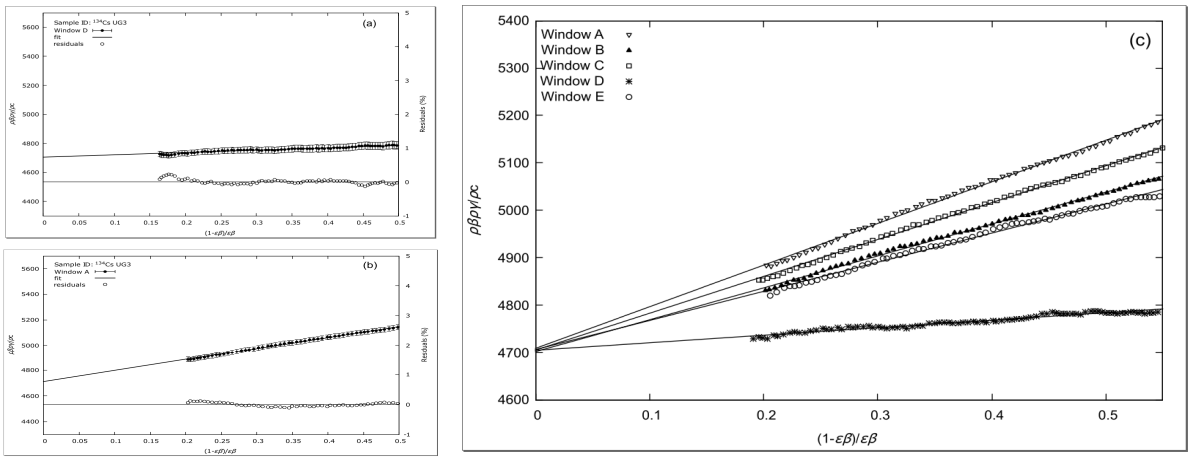


Figure 4 shows that only window D gives a nearly horizontal slope while other windows show a steeper slope. The window D covers the most dominant 796 keV gamma emission in the decay process with the influence of 802 keV gamma. Since we use the NaI(Tl) as the gamma detector, the two gamma emissions are recorded as a single peak. Therefore, the gamma window D case can be considered equivalent to a simple β-γ coincidence by disregarding the weak contribution of the 1365 keV gamma emission. For other gamma windows, the Compton contribute to the coincidence channel as the $\varepsilon_{cr,}$ causing a steeper slope of the efficiency curve.

Despite the different slope given by the five gamma window settings, the activity value obtained from the settings showing a difference within 0.33%. The comparison is shown in Figure 5, with the error bars representing the measurement's combined uncertainty. The mean activity value from the five gamma window settings is used as the final activity value of the ^{134}Cs. The value is (1145.6 ± 5.2) kBq/g at the reference time with quoted uncertainty evaluated at k = 1. The detail of the uncertainty budget is shown in Table 1.

Figure 5: Comparison of the activity value of ^{134}Cs obtained from five gamma window settings.

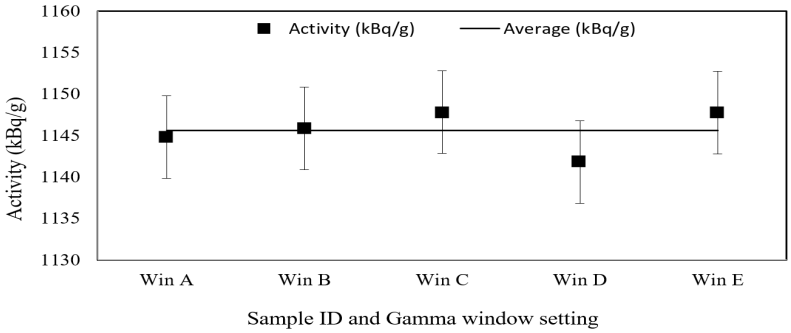
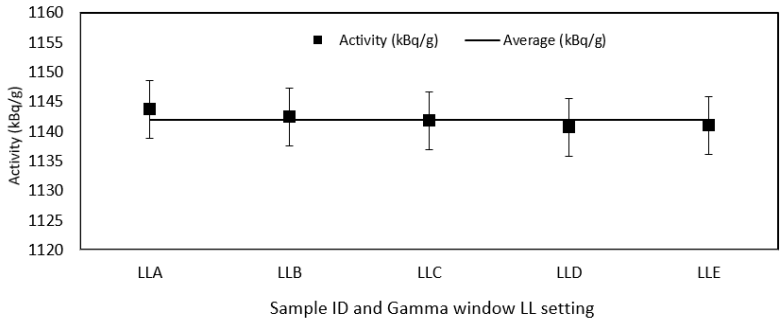


Table 1: Uncertainty budget for the activity measurement of ^{134}Cs.

component	comments	u (%)
measurement variability	S.D. of the mean for the sample measurements	0.19
Efficiency extrapolation	S.D. of the distribution obtained with a different efficiency range	0.10
β-β resolving time	the difference of activity values obtained using different resolving time values	0.38
^{134}Cs half-life	^{134}Cs half-life uncertainty propagating to the activity values referred to on reference date	0.001
Mass determination (including dilution)	Gravimetricmass determination uncertainty	0.10
^{134}Cs impurity	Measured impurity concentration fraction	0.02
	Combined uncertainty (u_c)	0.45
	Expanded uncertainty (k = 2)	0.90

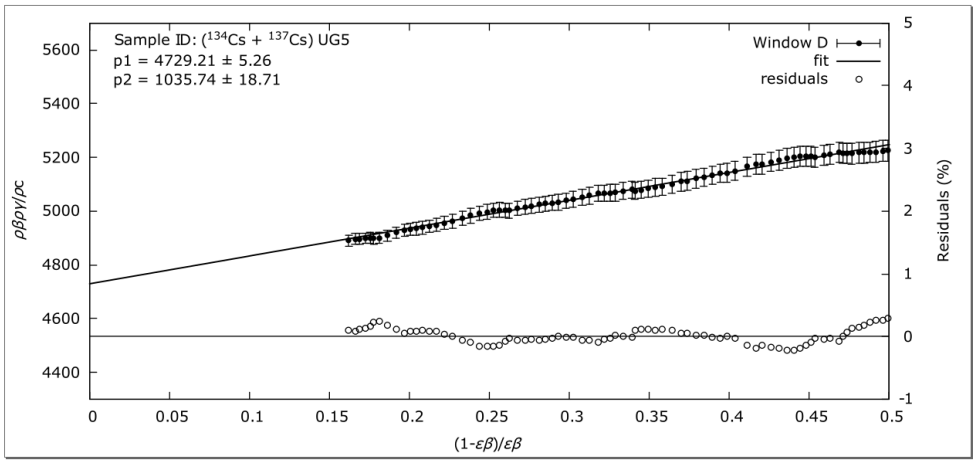
To completely avoid the 661.7 keV gamma emission contribution in the standardization of ^{137}Cs, the LL setting at window D needs to be appropriately set. The study activity measurement of ^{134}Cs at window D with different LL settings was conducted to evaluate the LL setting's effect on the activity of ^{134}Cs. The difference between the setting is within 0.16%. From this result, we applied the LLE for the setting of window D in the measurement of the ^{137}Cs to avoid the contribution of the 661.7 keV gamma completely. The result is shown in Figure 6.

Figure 6: Comparison of the activity value of ^{134}Cs obtained from five LL settings of window D.



Using the gamma window D and LLE setting, the mixed samples were measured to obtain the $\rho_{\beta 7}$. A program written in Fortran is used to bin-by-bin subtracting the $\rho_{\beta m}$ with the $\rho_{\beta 4}$. Once the $\rho_{\beta 7}$ is obtained, the analysis routine used in the activity standardization of ^{134}Cs is applied to obtain the efficiency curve. The least-square method is also used to fit the curve, and the extrapolated value is then divided by C to obtain the activity value. The C value is approximated to be 0.0915 [8]. Figure 7 is showing the typical efficiency curve for the ^{137}Cs.

Figure 7: Typical β-efficiency curve of ^{137}Cs.



The final activity value of 137Cs obtained from the measurement is (963.5 ± 9.4) kBq/ g at the reference time. The quoted uncertainty is evaluated at k = 1, with a detailed uncertainty budget is shown in Table 2.

Table 2: Uncertainty budget for the activity measurement of ^{137}Cs.

component	comments	u (%)
measurement variability	S.D. of the mean for the sample measurements	0.66
Efficiency extrapolation	S.D. of the distribution obtained with a different efficiency range	0.10
β-β resolving time	the difference of activity values obtained using different resolving time values	0.38
^{137}Cs half-life	^{137}Cs half-life uncertainty propagating to the activity values referred to on reference date	0.001
Mass determination (including dilution)	Gravimetric mass determination uncertainty	0.10
^{137}Cs impurity	Measured impurity concentration fraction	0.02
	Combined uncertainty (u_c)	0.74
	Expanded uncertainty (k = 2)	1.48

4 CONCLUSION

The activity standardization of ^{134}Cs and ^{137}Cs was conducted using the 4πβ(LS)-γ coincidence method with the application of efficiency tracing technique for the case of ^{137}Cs. For the case of activity standardization of ^{134}Cs, the selection of different gamma window settings gave different efficiency functions, which can be observed from the different slopes in their efficiency curve. However, the activity value is only differed by 0.33%. The final activity value for the ^{134}Cs and ^{137}Cs is obtained as (1145.6 ± 5.2) kBq/g and (963.5 ± 9.4) kBq/ g at the reference time, respectively. All quoted uncertainty is evaluated at k = 1.

5 ACKNOWLEDGEMENTS

The authors would like to thank the Korea Research Institute of Standards and Science (KRISS) for supporting this research activity.

6 REFERENCES

[1] Chino M, Terada H, Nagai H, et al., 2016. Utilization of 134Cs/137 Cs in the environment to identify the reactor units that caused atmospheric releases during the Fukushima Daiichi accident. Sci Rep 6:. https://doi.org/10.1038/srep31376

[2] Bé M-M, Chisté V, Dulieu C, et al., 2013. Table of Radionuclides (Vol. 7 – A = 14 to 245). BIPM, Sèvres

[3] Rytz A (1982) Activity measurements of a solution of 134Cs: Report on an international comparison. Rapp BIPM-80/2 192:427–431. https://doi.org/10.1016/0029-554X(82)90855-2

[4] Bé M-M, Chisté V, Dulieu C, et al., 2006. Table of Radionuclides Vol. 3. In: Monographie BIPM-5. pp 23–28

[5] Sochorová J, Auerbach P., 2014. Activity standardization of 134Cs and 137Cs. Appl Radiat Isot 87:207–210. https://doi.org/10.1016/J.APRADISO.2013.11.127

[6] Lee KB, Lee JM, Park TS, Lee SH, 2010. Development of a High-Ef fi ciency 4 (LS)– Coincidence System for Direct Measurements of Activity in Radioactive Decay. 57:2613–2616

[7] Lee KB, Lee JM, Park TS, et al., 2011. Application of digital sampling techniques for 4πβ (LS) γ coincidence counting. Nucl Instruments Methods Phys Res Sect A Accel Spectrometers, Detect Assoc Equip 626–627:72–76. https://doi.org/10.1016/j.nima.2010.10.091

[8] Sochorová J, Auerbach P, 2014. Activity standardization of 134Cs and 137Cs. Appl Radiat Isot 87:207–210. https://doi.org/10.1016/j.apradiso.2013.11.127