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Gamma Spectrometric Efficiency Measurement Uncertainty of ^{137}Cs in the Vegetation Sample

Yusuf Ağuş*

TAEK, Department of Radiation and Accelerator Technologies, Kahramankazan /Ankara, Turkey
*yusufagus06@gmail.com

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Abstract

In this study, gamma spectrometric efficiency measurement uncertainty of ^{137}Cs in vegetation sample is investigated. Each uncertainty component is evaluated and quantified. For this purpose, n-type 70% efficiency HPGe detector was used. The combined standard uncertainty of ^{137}Cs efficiency was found to be 2.43%.

Keywords: Efficiency Uncertainty, Gamma Spectrometry, Cs-137, Vegetation.

1. Introduction

Artificial radionuclides occur as a result of the nuclear power plant accidents and nuclear tests. One of these artificial radionuclides is ^{137}Cs. Due to the relatively high half-life ($T_{1/2}$=30.05 years), ^{137}Cs remains in ecosystem many years after the nuclear tests and accidents [1]. After the Chernobyl (1986) and Fukushima Daiichi (2011) nuclear power plant accidents, artificial radionuclides released and contamination has occurred. Contamination takes place as deposition of the radionuclides in the air. Deposited radionuclides in the soil pass into the vegetation. Radionuclides in the vegetation pass into the human by the consumption of the vegetation within the food chain [2-6]. Therefore, determination and the evaluation of the contamination level and radioactivity concentrations for natural and artificial radionuclides are important for ensuring public health and food sector workers safety.

Gamma spectrometric analysis with the High Purity Germanium (HPGe) semiconductor detectors is a powerful radioanalytical analysis technique. Determination of the radioactivity concentration of the gamma-emitting radionuclide in the sample is the main goal of the gamma spectrometric analysis. Additionally, full energy peak (FEP) efficiency and photon emission intensities (f_γ) can be obtained with this technique [7]. The measurement of radioactivity concentration via gamma spectrometric analysis should be performed with high accuracy and precision. Therefore, FEP efficiency calibration must be done accurately and precisely before the radioactivity measurements. FEP efficiency calibration depends on the source-detector geometry, sample matrix and on the photon energy. Sample and standard must be counted exactly same geometry and in the same type of sample container for the well-established efficiency calibration.

The efficiency measurement uncertainty is an essential part of the determination of the radioactivity concentration measurement results. Nuclear analytical laboratories (NAL's) must take into account efficiency measurement uncertainty while preparing radioactivity concentration uncertainty budget. Additionally, measurement uncertainty evaluation is a part of the ISO/IEC 17025 technical requirements of the NAL's and quantifying uncertainty in nuclear analytical measurements. Therefore, more attention is given to the evaluation of the measurement uncertainty and the preparation of uncertainty budget [8-11].

In this study, components of the FEP efficiency measurement uncertainty of the ^{137}Cs in vegetation sample were evaluated and quantified using gamma spectrometric method. Contribution of the uncertainty components to the combined standard uncertainty evaluated and expanded uncertainty obtained multiplying by coverage factor k.

2. Materials and Methods

2.1. Sample Preparation and Experiment

Radioactive vegetation reference standard was used to obtain efficiency value at 661.6 keV ^{137}Cs photon energy. Vegetation standard had 9871.67 Bq.kg^{-1} ^{137}Cs radioactivity concentration at reference date. Standard were put into cylindrical plastic analysis container (6x5 cm diameter x height dimension). Then, analysis container sealed and weighed. Efficiency measurement was performed by using HPGe gamma spectrometer. Specifications of the gamma spectrometric system are given in Table 1. ^{241}Am, ^{137}Cs and ^{60}Co standard point sources were used for energy calibration. Before the measurements, an empty analysis container counted for the determination of the background effects.

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Table 1. Specifications of the gamma spectrometric system.

Detector		
Type		n-type
%Rel. Eff. (1.33MeV)		70%
Crystal diameter [mm]		73.2
Crystal length [mm]		65.0
Peak/Compton		60:1
Resolution	1332.5 keV ^{60}Co	2.3 keV
	122.0 keV ^{57}Co	1.1 keV

2.2. Efficiency Determination

Efficiency is calculated by the following formula [9]

$$\varepsilon = \frac{N}{A \cdot \gamma \cdot t_s \cdot m \cdot C_1 \cdot C_2 \cdot C_3 \cdot C_4 \cdot C_5} \quad (2.1)$$

where,

- N is corrected net peak area of the corresponding photo peak. It is calculated as $N = N_s - \frac{t_s}{t_b} \cdot N_b$. N_s and N_b are the net peak area in the sample spectrum and in the background spectrum respectively. t_s and t_b live time of the sample measurement and of the background measurement in seconds.
- A is radioactivity concentration of the ^{137}Cs in the sample.
- m is dried mass of the sample (kgs).
- γ is emission probability of the ^{137}Cs photo peak energy.
- C_1 is correction factor for nuclear decay during the sampling to measurement

$$C_1 = e^{-\frac{\ln(2)\cdot\Delta t}{T_{1/2}}} \quad (2.2)$$

where Δt is the elapsed time from the time of sampling to the beginning of measurement(s), $T_{1/2}$ is the half-life(s) [9]

- C_2 is correction factor for the nuclide decay during counting period

$$C_2 = \frac{T_{1/2}}{\ln 2 \cdot t}\left(1 - e^{-\frac{\ln(2)\cdot t}{T_{1/2}}}\right) \quad (2.3)$$

where t is the elapsed real time during the measurement[9].

- C_3 is self-attenuation correction factor

$$C_3 = \frac{1 - e^{-\mu x}}{\mu x} \quad (2.4)$$

where μ and x are the linear attenuation coefficient and thickness of the volume sample[8].

- C_4 is random summing correction factor.

$$C_4 = e^{-2 \cdot R \cdot \tau} \quad (2.5)$$

where τ is the resolution time of the measurement system(s). R is the mean count rate [9].

- C_5 is coincidence correction factor [9]. There is no true coincidence factor for ^{137}Cs.

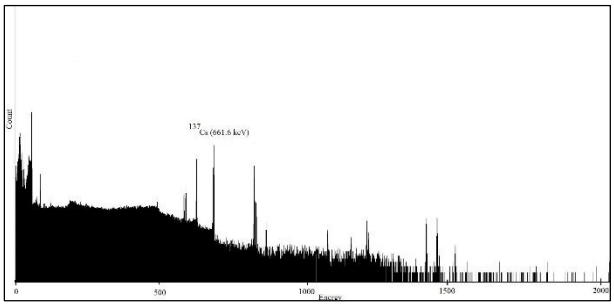

Figure 1. Gamma-ray spectrum of the radioactive vegetation reference standard.

Repeatability (precision) is the closeness of the result of the consecutive measurements. During the measurement process, experimental condition remains unchanged. In the repeatability measurements analyst, detection system, laboratory conditions are the same and successive measurements carrying out in short period of time. Repeatability (precision) is the random error and expressed as the standard deviation(s) or the percent relative standard deviation (RSD%). Reproducibility is closeness of results of measurements carried out under changed conditions. In the reproducibility measurements detection system, analyst, laboratory condition and measurement interval can change. Reproducibility of the efficiency measurement was determined by using different analyst's results.

3. Results and Discussions

Efficiency value was calculated by using equation 2.1. Activity concentration of the reference standard material was 9871.67 Bq.kg^{-1}. Measurement live time, background measurement live time, net photopeak area in sample spectrum, net photopeak area in spectrum of background and mass of sample were 1000 s, 263761 s, 10887, 109, 0.06983 kg respectively. The half-life and gamma emission probability of the radionuclide was 30.05 years and 0.8499 respectively [1]. Linear attenuation coefficient (μ) was taken from XCOM[12]. Linear attenuation coefficient was 0.009513 cm^{-1}. Mean count rate, resolution time of measurement system are R $\approx$ 1 cps and τ= 6 x 10^{-6} s respectively.

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Table 2. The uncertainty components of the efficiency measurement of ^{137}Cs in vegetation sample.

Variable Name	Value	Uncertainty value	Contribution to the Combined uncertainty (%)
Background corrected net peak area	10887.0	106.0	16.11
Sample mass	0.06983	0.00002	0.03
Gamma emission probability	0.8499	0.002	0.94
Radioactivity concentration	9871.67	178.68	55.68
Correction factor for decay between sampling and measurement	0.8114	4.5×10^{-4}	0.05
Correction factor for decay during counting period	1	9.7×10^{-10}	0
Correction factor for self-attenuation	0.9882	0.0002	0.01
Correction factor for random summing	1	1.7×10^{-05}	0
Coincidence correction factor	1	0.0	0
Repeatability	1	0.0091	14.3
Reproducibility	1	0.0086	12.9
Relative combined standard uncertainty (σ)	= 2.43%		
Relative expanded uncertainty (2σ)	= 4.86%		
Value of the efficiency ± uncertainty	= 0.0231 ± 0.001 (coverage factor k=2)		

The time between the start of the measurement and standard reference date was 3310 days. Correction factors values obtained as C_1=0.8114, C_2=1.0, C_3=0.9882 and C_4= 1.0 respectively. Since the nuclide (^{137}Cs) has no cascade of gamma rays, there is no need coincidence correction factor (C_5=1.0). Using equation 2.1, we obtained the efficiency of ^{137}Cs in the sample as 0.0231.

The combined standard uncertainty of the efficiency measurement (ε) is calculated in terms of component uncertainties by the following formula [8-10]:

$$u(\varepsilon(x_1,..,x_n)) = \sqrt{\sum_{i=1}^{n}\left(\frac{\partial\varepsilon}{\partial x_i}\right)^2 (u(x_i))^2} \qquad (3.1)$$

Each variable's contribution is the square of the associated uncertainty expressed multiplied by the square of the associated partial derivative. The expanded uncertainty U(ε) is obtained by multiplying the combined standard uncertainty by a suitable coverage factor k_α. The efficiency value was determined in the interval ε ± U(ε). Efficiency uncertainty calculation of the ^{137}Cs in the vegetation sample is given below [8,9]. Uncertainty of the net peak area calculated by the following formula:

$$u(N) = \sqrt{u^2(N_s) + \left(\frac{t_s}{t_b}\right)^2 \cdot u^2(N_b)} \qquad (3.2)$$

where, live time of the sample spectrum collection and live time of the background spectrum collection were t_s = 1000 s and t_b = 263761 s, respectively. Uncertainty of net photopeak area in sample spectrum ($u(N_s)$) was 106.0. Uncertainty of net photopeak area in spectrum of background ($u(N_b)$) was 2.6 and uncertainty of the net peak was u(N)=106.0. Relative uncertainty of standard activity concentration (u(A)) was taken from standard certificate as 178.68. Relative uncertainty of sample mass was estimated by calibration certificate of the balance as 0.0004. Uncertainty of sample mass was u(m) = (0.0004 x 0.04893) = 0.00002. The uncertainties of the half-life of the radionuclide and gamma emission probability were based on Nuclide-Lara data sheets [1]. Uncertainty of half-life: $u(T_{1/2})$= 0.08 years. Uncertainty of gamma emission probability was u(γ) = 0.002. Uncertainty of decay correction factor was $u(C_1) = C_1 \cdot \Delta t \cdot u(\lambda)$. Where $\lambda = \ln 2/T_{1/2}$ and its uncertainty was

$$u(\lambda) = \lambda \cdot u(T_{1/2})/T_{1/2}$$
$$u(C_1) = 4.5\times10^{-4} \qquad (3.3)$$

Uncertainty of the correction factor for the nuclide decay during counting period

$$u(C_2) = \frac{1-C_2(1+\lambda\cdot t)}{\lambda} u(\lambda) = 9.7\times10^{-10} \qquad (3.4)$$

Uncertainty of the correction factor for self-attenuation

$$u(C_3) = C_3 \cdot \left|\frac{e^{-\mu x}}{C_3} - 1\right| \cdot \sqrt{\left(\frac{u(x)}{x}\right)^2 + \left(\frac{u(\mu)}{\mu}\right)^2} = 0.0002 \qquad (3.5)$$

Uncertainty of the correction factor for pulses loss due to random summing

$$u(C_4) = \sqrt{(2\cdot R\cdot e^{-2\cdot R\cdot\tau})^2 \cdot u^2(\tau) + (2\cdot\tau\cdot e^{-2\cdot R\cdot\tau})^2 \cdot u^2(R)} = 1.7\times10^{-5} \qquad (3.6)$$

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Table 3. Repeatability measurement results

Measurement	Efficiency
1	0.0228
2	0.0232
3	0.0231
4	0.0234
5	0.0229
6	0.0233
7	0.0233
8	0.0234
9	0.0229
10	0.0233
Average	0.0232
Std. Dev.	0.0002
RSD%	0.9182

Table 4. Reproducibility measurement results

Measurement	Efficiency	
	Analyst#1	Analyst#2
1	0.0227	0.0235
2	0.0229	0.0235
3	0.0227	0.0231
4	0.0226	0.0231
5	0.0229	0.0233
6	0.0231	0.0234
7	0.0229	0.0235
8	0.0234	0.0231
9	0.0229	0.0233
10	0.0231	0.0235
Average	0.0229	0.0233
Std. Dev.	0.0002	0.0002
RSD%	0.9934	0.7253

The coincidence correction factor uncertainty ($u(C_5)$) is taken as 0, because there is no true coincidence factor for ^{137}Cs (C_5=1.0). Ten measurements carried out for determination of the repeatability contribution to the efficiency measurement uncertainty. RSD value was found to be 0.9180. Results are given in Table 3. Each of the two analysts was carried out ten measurements for determination of the reproducibility contribution to the efficiency measurement uncertainty. Pooled RSD value was found to be 0.8697. Results are given in Table 4.
Using equation 3.1 and all uncertainty components:

$$u_c(\varepsilon)=\frac{u(\varepsilon)}{\varepsilon}=\sqrt{\left(\frac{u(m)}{m}\right)^2+\left(\frac{u(N)}{N}\right)^2+\left(\frac{u(\gamma)}{\gamma}\right)^2+\left(\frac{u(A)}{A}\right)^2+\sum_{i=1}^{5}\left(\frac{u(C_i)}{C_i}\right)^2+\left(u_{Reproducibilty}\right)^2+\left(u_{Repeatability}\right)^2} \quad (3.7)$$

we obtained the relative combined standard uncertainty ($u_c(\varepsilon)$) as 2.43% and the value was 0.00055. Multiplying coverage factor (k=2), relative expanded uncertainty ($U(\varepsilon)$) found as 4.86 %. Value of the measured efficiency and expanded uncertainty were 0.0231 and 0.001 respectively. The results are summarized in Table 2.

4. Conclusion

In this paper, evaluation and quantification of gamma-spectrometric efficiency measurement uncertainty of ^{137}Cs in the vegetation sample was presented. For this purpose, radioactive vegetation reference standard was used. Uncertainty components and their contribution to the relative combined standard uncertainty were quantified. Main contributions to the uncertainty originated from corrected net area of the sample peak, radioactivity concentration, repeatability and reproducibility.

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Ethics

There are no ethical issues after the publication of this manuscript.

References

1. Nucléide – Lara Library for gamma and alpha emissions. http://www.nucleide.org/Laraweb (accessed at 21.03.2018).

2. Di Gregorio, DE, Huck, H, Aristegui, R, De Lazzari, G, Jech A. 2004. ^{137}Cs contamination in tea and yerba mate in South America. *Journal of the Environmental Radioactivity*; 76:273–281

3. Kuwahara, C, Fukumoto, A, Ohsone, A, Furuya, N, Shibata, H, Sugiyama, H, Kato, F. 2005. Accumulation of radiocesium in wild mushrooms collected from a Japanese forest and cesium uptake by microorganisms isolated from the mushroom-growing soils. *Science of the Total Environment*; 345:165– 173.

4. Lehto, J, Vaaramaa, K, Leskinen A. 2013. ^{137}Cs, $^{239,240}Pu$ and ^{241}Am in boreal forest soil and their transfer into wild mushrooms and berries. *Journal of Environmental Radioactivity;* 116:124-132.

5. Marzano, FN, Bracchi, PG, Pizzetti P. 2001. Radioactive and Conventional Pollutants Accumulated by Edible Mushrooms (Boletus sp.) Are Useful Indicators of Species Origin. *Environmental Research Section A.;* 85: 260-264

6. Topcuoglu, S, Güngür, N, Köse, A, Varinlioglu A. 1997. Translocation and depurtion of ^{137}Cs in tea plants. *Journal of Radioanaltical and Nuclear Chemistry;* 218: 2: 263-266.

7. Gordon, RG. Practical Gamma-ray Spectrometry; Wiley, 2nd ed.: Sussex, England, 2008; pp 150.

8. Lepy, MC, Pearce, A, Sima, O. 2015. Uncertainties in gamma-ray spectrometry. *Metrologia*; 52: S123-S145.

9. IAEA- International Atomic Energy Agency. Guide Quantifying Uncertainty in Nuclear Analytical Measurements. IAEA-TECDOC-1401, Vienna, 2004.

10. ISO. Guide to the Expression of Uncertainty in Measurement, 1995.

11. EURACHEM / CITAC GUIDE CG4. Quantifying Uncertainty in Analytical Measurement. 3nd Edition. 2012.P1.

12. XCOM, NIST, Pyhsical Meas. Laboratory. https://physics.nist.gov/PhysRefData/Xcom/html/xcom1.html (accessed at 21.03. 2018).