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Validation of an advanced analytical procedure applied to the measurement of environmental radioactivity

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ABSTRACT

In this work, an advanced analytical procedure was applied to calculate radioactivity in spiked water samples in a close geometry gamma spectroscopy. It included MCNP-CP code in order to calculate the coincidence summing correction factor (CSF). The CSF results were validated by a deterministic method using ETNA code for both p-type HPGe detectors. It showed that a good agreement for both codes. Finally, the validity of the developed procedure was confirmed by a proficiency test to calculate the activities of various radionuclides. The results of the radioactivity measurement with both detectors using the advanced analytical procedure were received the "Accepted" statuses following the proficiency test.

1. Introduction

In gamma-ray spectrometry, the quantitative information can be derived from the detection efficiency calibration, radionuclide activity, and gamma emission intensity (Lépy et al., 2015). Moreover, a low-level measurement generally requires determining the accurate efficiency calibration and getting reliable quantitative analysis results from sample measurements, especially in close sample-detector geometries. In these cases, a well-known phenomenon is called "true coincidence" or "cascade summing", and "coincidence summing", the latter term will be used in this work. This coincidence summing effect in the gamma-ray spectra depends on the source-to-detector distance, the complexity of the decay scheme, and the detection solid angle (Andreev et al., 1972; Semkow et al., 1990). The determination of coincidence summing correction factors (CSF) can be calculated by different and available code such as EFFTRAN (Vidmar et al., 2011), EGS4 (Celik et al., 2015), ETNA (Lépy et al., 2012a), GEANT4 (Giubrone et al., 2016), GESPECOR (Arnold, and Sima., 2006), and PENELOPE (García-Toraño et al., 2017). An intercomparison exercise (Lépy et al., 2012b) and a testing equivalence of computer codes (Vidmar et al., 2016) on the calculation of CSF in gamma-ray spectrometry considerable volume geometry have been performed.

The goal of this work was to validate the coincidence summing correction factor on volume sources by the MCNP-CP and ETNA (an acronym standing for Efficiency Transfer for Nuclide Activity measurements) codes. In addition, the coincidence summing correction will be considered for the radionuclides on the spiked water samples using the MCNP-CP code. The mass activities from measurements of radionuclides with an advanced procedure including the coincidence summing correction using Monte Carlo method will be evaluated in the IAEA proficiency test.

2. Materials and methods

2.1. Experimental set-up

In this study, we utilized two p-type Ge detectors with different efficiencies: 50% (Model GEM50 supplied by Ortec, Inc., laboratory code GEM50), and 35% (Model GC3520 supplied by Canberra, Inc., code GC3520), which used the Lynx based on advanced digital signal processing techniques. Gamma spectra of the former were acquired by Maestro software (Maestro®-32., 2003) with 16384 channels while that of the latter were recorded by Genie software (Genie 2000., 2009) with 32748 channels. The detector diagrams and specifications were provided by the manufacturer (see Table 1). Both detectors are surrounded by a cylindrical low-background passive shielding to record photon energy up to 3 MeV.

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Table 1
Parameters of the HPGe detectors.

Geometrical parameters of the detector	GEM50 (mm)	GC3520 (mm)
Side cap diameter	83.0	76.2
Window thickness	1.0	1.5
Crystal-window distance	4.0	5.0
Crystal-dead layer thickness (outer)	0.7	0.46
Crystal-dead layer thickness (inner)	0.0003	0.0003
Crystal diameter	65.9	50.1
Crystal length	77.0	62.2
Crystal hole depth	64.9	23
Crystal hole diameter	11.5	7.5

Table 2
Radioactivities of the standard solution.

Radionuclides	Energy (keV)	Activity (Bq)	Uncertainty (%)
^{210}Pb	46.54	563.85	4.1
^{241}Am	59.54	55.81	3.0
^{109}Cd	88.03	414.71	3.1
^{57}Co	122.06	11.60	3.1
^{123m}Te	158.97	8.50	3.0
^{113}Sn	391.70	30.37	3.0
^{85}Sr	514.00	14.51	3.0
^{137}Cs	661.66	81.34	3.1
^{88}Y	898.01	52.17	3.0
^{60}Co	1173.23	94.71	3.0
^{60}Co	1332.49	94.71	3.0
^{88}Y	1836.07	52.17	3.0

2.2. Standard and sample solutions

The standard solutions were packed in a 500 mL cylindrical container and were filled to height 143.5 mm. The efficiency curve of the full energy peak (FEP) was obtained by a mixed standard that included radionuclides such as ^{210}Pb, ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co, and ^{88}Y with energy range 47–1836 keV. The mixed standard solution was purchased from Eckert & Ziegler Isotope Products Company with around 3% combined uncertainties. (see Table 2). One must be noted that it is necessary to include correction factors for coincidence summing of the radionuclides ^{60}Co and ^{88}Y. In the case of ^{60}Co (1173.23 keV, and 1332.49 keV), and ^{88}Y(898.01 keV, and 1836.07 keV) two gamma-rays are emitted during the same decay event of the nucleus so that they appear to be emitted instantaneously. This seemingly instantaneous emission of separate gamma-rays is known as coincidence summing. In this situation, the detector will see both of the gamma-ray energies as one larger (2505.72 keV, and 2734.08 keV) energy deposited in the detector. This effect will be causing the count of the first gamma-ray to be "lost" from its FEP. They were corrected using MCNP-CP code (Berlizov, 2012) of about 10% (see Table 3). The ACORES code (Lépy et al., 2008) was used to fit the experimental efficiency curves with the log-log polynomial. Fig. 1 presents the efficiencies of both detectors including the fitting function. At the energy lower than 300 keV, the efficiency of the detector GC3520 is higher than the detector GEM50 because of the dead-layer. However, the result is reversed for the energy higher than 300 keV.

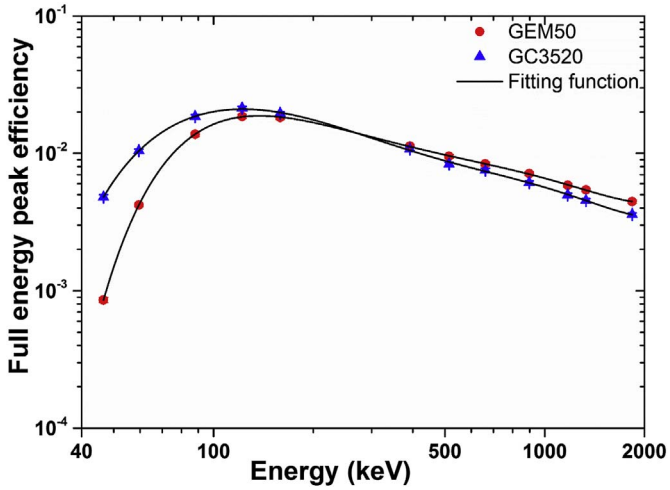

Fig. 1. Experimental full energy peak efficiency calibration curve of the standard solution.

The three series spiked water proficiency tests are IAEA-TEL-2014 (spiked 01, and spiked 02), IAEA-TEL-2015 (spiked 03, and spiked 04), and IAEA-TEL-2016 (spiked 05, and spiked 06), respectively. They were

Table 3
The coincidence summing corrective factors (CSF) were taken into account MCNP-CP and ETNA codes, respectively.

Radionuclide	E (keV)	GC3520			GEM50		
		MCNP-CP	ETNA	RD(%)	MCNP-CP	ETNA	RD (%)
^{134}Cs	563.25	1.22	1.28	4.5	1.24	1.27	3.0
^{134}Cs	569.33	1.24	1.28	3.0	1.25	1.27	1.6
^{134}Cs	604.72	1.13	1.16	1.9	1.14	1.15	0.9
^{134}Cs	795.86	1.13	1.15	2.0	1.14	1.15	0.6
^{134}Cs	801.95	1.20	1.24	4.0	1.21	1.24	2.4
^{152}Eu	121.78	1.15	1.17	1.9	1.15	1.18	2.7
^{152}Eu	244.70	1.20	1.22	1.9	1.18	1.23	3.8
^{152}Eu	344.28	1.09	1.10	0.7	1.09	1.11	2.1
^{152}Eu	411.12	1.20	1.24	3.3	1.21	1.27	4.8
^{152}Eu	443.97	1.16	1.18	2.3	1.15	1.19	3.2
^{152}Eu	778.90	1.11	1.14	2.6	1.12	1.15	3.3
^{152}Eu	964.10	1.10	1.10	0.2	1.07	1.09	1.7
^{152}Eu	1085.84	0.98	0.98	0.5	0.97	0.98	1.1
^{152}Eu	1112.08	1.06	1.07	0.3	1.05	1.06	1.2
^{152}Eu	1408.01	1.07	1.07	0.0	1.06	1.06	0.3
^{22}Na	1274.54	1.22	1.26	3.9	1.23	1.31	6.0
^{88}Y	898.01	1.08	1.09	0.9	1.09	1.10	0.9
^{88}Y	1836.06	1.09	1.10	0.9	1.09	1.10	0.9
^{60}Co	1173.32	1.09	1.11	1.8	1.10	1.10	0.0
^{60}Co	1332.49	1.10	1.11	0.9	1.10	1.10	0.0

RD is relative deviation, $RD = \frac{|CSF_{MCNP-CP} - CSF_{ETNA}|}{CSF_{MCNP-CP}} \times 100\%$.