The first experimental test of the MEFFTRAN software on HPGe detector calibration for environmental samples

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ABSTRACT

The gamma spectrometry of environmental samples poses a series of specific problems for the practitioner. The determination of full energy peak efficiencies is, in the case of environmental samples, one of the greatest challenges. In this paper, the calibration of two HPGe detectors for environmental samples in Marinelli beakers was performed using the newly developed MEFFTRAN software. The results obtained with MEFFTRAN were compared to the experimental efficiencies measured using the calibration sample produced at the Laboratory for Radiation and Environment Protection, Institute for Nuclear Sciences Vinca, Belgrade, in order to confirm the validity of the calculation. It is shown that calculated and experimental efficiencies are in good agreement with the discrepancies from -3.9% to $+1.5\%$. The results were also validated by measuring secondary reference materials provided by the IAEA within several interlaboratory proficiency tests. The activity concentration of the radionuclides in the secondary reference materials was determined using the efficiency obtained by MEFFTRAN and compared with the target value given by the IAEA. All the results in materials provided by the IAEA were acceptable except one, proving that MEFFTRAN produces satisfactory results.

1. Introduction

The gamma spectrometry of environmental samples poses a series of specific problems for the practitioner. Environmental samples can vary significantly in terms of the chemical composition and structure, thus in principle requiring an efficiency calibration for measurement of each new sample. One way to overcome this problem is using some computing method (i.e. Monte Carlo simulation or efficiency transfer) to calculate the efficiency for the specific sample geometry. One such computing method is EFF-TRAN, software that calculates the efficiency transfer, using a method defined by Moens et al. (1981) and its newly developed version for Marinelli beakers - MEFFTRAN (http://www.efftran.com/MEFFTRAN.zip). Moens' theorem states that it is possible to calculate the desired full-energy-peak efficiency of the extended source as a product of the measured full-energy-peak efficiency of the calibrated source and the ratio of the computed total efficiencies for these two sources. In order to calculate the required total efficiencies, Monte Carlo integration is used in EFFTRAN and

MEFFTRAN as a suitable choice for many-dimensional systems. Because the model of the sample is constructed from cylinders only, the only complex operation required in the code is the calculation of the path length traversed through a cylinder of given dimensions by a gamma ray originating at an arbitrary location (Vidmar 2005).

The same basis for calculation of efficiencies was used in case of Marinelli beakers in MEFFTRAN. The self-absorption computation in MEFFTRAN (in the case of identical standard and sample dimensions) follows the method of Sima (1992). The definition of source and sample material, source filling height, as well as the definition of the detector geometry is done in the same way as in EFFTRAN, by defining the dimensions, chemical composition and densities in a dedicated module (Vidmar 2005). MEFFTRAN also contains the module which calculates coincidence correction factors based on the defined parameters of the detector and sample. Similar to its predecessor EFFTRAN, MEFFTRAN is also using Microsoft Excel as a user interface, making it very easy to use.

A great advantage of this method, as pointed out and verified by many users of EFFTRAN, is that in the calculated ratio, many inaccuracies in the detector model can be expected to cancel out, making it possible to work directly with non-optimized detector data supplied by the manufacturer. The cancellation of differences

also applies to the interaction cross-section data that this method may use (Vidmar et al., 2010, Vidmar and Likar 2004; Nikolic et al., 2014a). However, some uncertainties rising from poor knowledge of the composition of the sample as well as the lack of the precision in the detector geometry data cannot be avoided, therefore resulting in not negligible uncertainty.

This paper presents the results of the first experimental testing of the program MEFFTRAN done at the Laboratory for Radiation and Environment Protection, Institute for Nuclear Sciences Vinca, Belgrade. First, the measurement of reference material - Marinelli beaker, volume 500 ml filled with epoxy resin that contains radionuclides of certified total activity (product No. 9031-OL-208/18, named Marinelli Standard) issued by Czech Metrological Institute, was conducted in order to obtain a reference calibration curve. The software was then used to calculate efficiency for several secondary reference materials. The first secondary reference material (named Marinelli Sand) was produced in Laboratory fior Radiation and Environmental Protection, Institute for Nuclear Sciences Vinca, Serbia. The efficiencies calculated with MEFFTRAN were compared to the experimental efficiencies obtained by measuring the Marinelli Sand, in order to confirm the accordance between them. Also, secondary reference materials, issued by IAEA within the framework of several interlaboratory proficiency tests, were used for validation of MEFFTRAN. All these secondary reference materials were measured in the same 500 ml Marinelli as the Marinelli Standard and Marinelli Sand were. Their activity concentration was determined using efficiency calculated by MEFFTRAN and the results were compared to the target values. The comparison was performed using the u_{test} recommended by the IAEA in order to further validate the results obtained by MEFFTRAN.

2. Material and methods

The calculation of the efficiencies using MEFFTRAN begins by defining the reference calibration curve. Also, the geometrical characteristics of the detector have to be defined in the module Detector. The sample for which the efficiency is calculated, as well as the reference sample is defined in the module named „Sample". The measurement geometry is defined as Marinelli beaker with adjustable radii and sample filling height. Different matrices of the samples are defined via the chemical composition and density in the module „Material". The calculation is then performed in the module "Transfer" by inserting the reference calibration curve and pressing the button that starts the calculation.

To obtain the reference calibration curve, a reference material, Marinelli beaker filled with epoxy resin containing gamma emitting radionuclides (product 9031-OL-419/12 issued by Czech Metrological Institute), was measured for 60,000 s. The list of radionuclides contained in the reference material is given in Table 1. The full energy peak efficiency was calculated according to the well known expression:

$$\varepsilon = \frac{N \cdot C(E)}{t \cdot P_\gamma \cdot A} \tag{1}$$

where N represents the net count, $C(E)$ is the coincidence summing correction factor (also calculated using MEFFTRAN), t is counting time, P_γ is the photon emission probability and A is the source total activity for the radionuclides emitting gamma photons with different energies E, with the decay correction. The duration of the measurement was set on live time, so there was no need for dead time loss correction.

Relative combined measurement uncertainty for the experimental values was calculated according to the following equation:

Table 1
The radionuclides contained in the reference material Marinelli beaker and secondary reference material (http://www.nucleide.org/DDEP_WG/DDEPdata.htm) and their total activity at reference date 31.08.2012.

Radionuclide	E [keV]	Pγ [%]	A [Bq]	
			Marinelli sand	Marinelli standard[a]
241Am	59.5409	35.92	497.4	3691 + 1.1
109Cd	88.0336	3.66	2284.7	16950 + 1.4
139Ce	165.8575	79.90	148.3	1101 + 1.1
57Co	122.0606	85.51	124.2	921 + 1.1
	136.473	10.71		
60Co	1173.24	99.85	378.2	2806 + 1.1
	1332.5	99.98		
137Cs	661.657	84.99	277.1	2041 + 1.2
203Hg	279.195	81.48	278.5	3117 + 2.4
113Sn	391.698	64.97	432.3	4295 + 1.2
85Sr	514.005	98.50	595.6	4308 + 1.2
88Y	898.036	93.70	597.4	1973 + 2.3
	1836.05	99.35		

[a] Taken from the certificate No.9031-OL-419/12.

$$u(\varepsilon) = \sqrt{(\delta A)^2 + (\delta N)^2 + (\delta C(E))^2} \tag{2}$$

where δA represents the relative uncertainty of the total activity in the standard given by the manufacturer, δN is the relative counting uncertainty and $\delta C(E)$ is the relative uncertainty introduced via coincidence correction factors (a relative statistical uncertainty due to Monte Carlo integration in MEFFTRAN). The uncertainties of the measurement time t, mass of the Marinelli Standard m and P_γ were negligible. Relative combined measurement uncertainty $u(\varepsilon)$ for all energies did not exceed 5% at 2σ level of confidence (Nikolic et al., 2014a).

The calibration curve was obtained by fitting the measured values using the following fitting function (Osae et al., 1999):

$$\ln\varepsilon = \sum_{i=1}^{5} a_i (\ln E)^{i-1} \tag{3}$$

where a_i are fitting coefficients and E is the energy.

The first measured secondary reference material (named Marinelli Sand) was produced at the Laboratory for Radiation and Environmental Protection, Institute for Nuclear Sciences Vinca, Serbia, by spiking the active charcoal carrier with the standard radioactive solution issued by the Czech Metrological Institute (type ERX-170712-1370434, certificate number 9031-OL -427/12) and mixing the carrier with the sand matrix (Vukanac et al., 2008). After thorough homogenization, the matrix was placed into a 500 ml Marinelli beaker and measured. The list of radionuclides present in the Marinelli Sand is listed in Table 1.

Other measured samples were issued by IAEA within several interlaboratory proficiency tests. These samples are: Sediment IAEA-Tel-2014, Grass IAEA-TEL-2012, Seaweed IAEA-Tel-2014 and Soil IAEA-Tel-2013. They were placed into 500 ml Marinelli beakers and measured immediately.

The characteristics of all measured samples were defined in the software via their chemical composition and density of the matrix, as presented in Table 2. Chemical composition and density were defined according to Ref. (Nikolic et al., 2014a, b; Savenko, 2007). The sample geometry is predefined as a Marinelli beaker and the only variable is the sample filling height, which was measured for each sample. The composition of the Marinelli beaker was defined via the container material (polystyrene), diameter (110 mm), thickness (1 mm), hole diameter (80 mm) and hole length (40 mm).

The measurements were conducted on two HPGe detectors,