HPGe detector efficiency optimization for the atypical measurement geometry of simulated aerosol filters

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***Abstract* — Gamma spectrometry is widely used method of choice for measurement of environmental samples conducted during monitoring of the environment and contamination control, as well as measurement of radionuclide content in various materials. However, one of the main challenges in this method of spectrometry is the determination of detection efficiency for different energies, different source-detector geometries and different composition of samples. This task is defined as an efficiency calibration of the detector. When using a commercial calibration sources is not possible, or the available sources are not adequate, the optimization of the efficiency calibration has to be performed.**

In this paper, the results of the optimization of efficiency calibration for the atypical geometry and composition of the simulated aerosol samples, measured within the Proficiency tests organized by International Atomic Energy Agency (IAEA), performed using EFFTRAN efficiency transfer software, will be presented and discussed.

***Index Terms*— gamma spectrometry; efficiency calibration, EFFTRAN; optimization**

I. INTRODUCTION

Gamma spectrometry is one of the mostly often used measurement methods for determining the radionuclide content in various samples. It is a non-destructive method which can be applied for a wide range of environmental samples measured within the framework of a monitoring, as well as for contamination control.

This method is based on the interaction of gamma rays emitted from the sample with the active volume of the detector. Semiconductor detectors and among them, high purity germanium (HPGe) detectors, are mostly used due to their high sensitivity and good energy and time resolution. The result of a gamma spectrometric measurement is represented by the spectrum of photons originating from the source, that are collected by the multichannel analyzer (MCA) and the number of photons detected is proportional to the activity of the given radionuclide. The main challenge in this method of measurement is the determination of the detection efficiency which is dependent not only on the type of the detector and the energy of the emitted gamma photons, but also on the type of the measured sample: its geometry and chemical composition, the sample – detector geometry and the presence of the absorber. This task is defined as an efficiency calibration of the detector [1].

The most often used approach to the efficiency calibration is a direct measurement of different calibration sources containing γ-ray emitters and subsequent fitting of obtained results to a parametric function, thus obtaining the efficiency curve – a functional dependence of the efficiency with respect to the energy. Different sample types require different calibration curves. Due to that, this approach requires a large number of calibration sources, produced to mimic the real measured samples to the largest possible degree, which may not be available. This problem is especially pronounced when environmental samples are of interest due to their diversity in composition and structure [2].

When the sample of the atypical geometry or composition is presented, an optimization of the calibration curve using the means available in the laboratory has to be performed. One of the methods of optimization is the efficiency transfer using some type of software. The software calculates the efficiency transfer factors with which the original efficiency for a given energy needs to be multiplied in order to obtain the efficiency in the special case of the measured sample [3]. One of these software is EFFTRAN [4], a user friendly software that calculates efficiency transfer factors and coincidence summing correction factors for cylindrical samples.

In this paper, the results of the efficiency calibration for the atypical geometry and composition of the simulated aerosol samples, measured within the Proficiency tests organized by International Atomic Energy Agency (IAEA), performed using EFFTRAN efficiency transfer software,

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will be presented and discussed.

II. MAIN RESULTS AND DISCUSSION

A. Efficiency transfer

The calculations of the efficiency transfer factors are based on the assumption that the detector efficiency for the special case of measured sample can be obtained by multiplying the reference efficiency (obtained by measuring the commercial or laboratory calibration source) by the efficiency transfer factors. In order to calculate these factors, a set of partial differential equations needs to be solved. For the purpose of the efficiency transfer, in our laboratory, EFFTRAN software is often used. It is organized as an user friendly Excel file with three modules. The software performs the needed calculations using a Monte Carlo integration, given the specific data are provided.

The data that the software requires are the detector characteristics (crystal material, diameter and length, thickness of the dead layer, housing geometry and composition, material of the window and window to crystal gap,) and the characteristics of both calibration sample used for the reference calibration curve and the measured sample (the diameter, filling height and thickness of the container, chemical composition and density of the sample matrix). . Because the model of the sample, as well as the detector crystal, can be 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 photon originating from an arbitrary location [4].

The choice of the reference efficiency plays a significant role in the final result, therefore it has to be chosen with care. This is especially important when the geometry of the measured sample differs significantly from the calibration source used for the efficiency calibration of the detector. Also the definition of the calibration source as well as the measured sample has to be performed as precise as possible, especially the chemical composition which has the largest influence on transfer factors. The final result of the calculation is the efficiency for the measured sample which is dependent on the reference efficiency used. The measurement uncertainty of the calculated efficiency is determined according to the following equation:
[5]:

$$u(\varepsilon) = \sqrt{(u(\varepsilon_{ref}))^2 + (u(C))^2 + (u_D)^2 + (u_S)^2} \tag{1}$$

where $u(\varepsilon)$ represents the combined measurement uncertainty of the efficiency for the measured sample, $u(\varepsilon_{ref})$ is the relative uncertainty of the reference efficiency value which has to be calculated, $u(C)$ is the uncertainty of the transfer factors calculated by the program as a statistical uncertainty of the Monte Carlo integration (≈ 1.2%), u_D is the uncertainty associated with the geometry of the detector and u_S is the uncertainty associated with the characteristics of the sample. The, for the measurement uncertainty of the measured activity, this component is combined with other contributions to obtain the total combined measurement uncertainty.

B. Results and Discussion

As it was said in the previous section, the final efficiency for the measured sample is dependent on the reference efficiency used. It is therefore crucial to perform some sort of validation of the results, when a choice of different reference efficiency is available.

In this investigation, the efficiency for an atypical geometry and composition has been calculated using three different reference efficiencies. The measured samples were simulated aerosol filters containing different artificial radionuclides, printed on a cellulose filter paper, diameter 43 mm, thickness of 1mm. These samples were measured within the World-Wide Open Proficiency test IAEA-TEL-2019-03, World-Wide Open Proficiency test IAEA-TEL-2020-03 and World-Wide Open Proficiency test IAEA-TEL-2020-05, organized by International Atomic Energy Agency (IAEA) during the year 2019 and 2020 [https://nucleus.iaea.org/sites/ReferenceMaterials/Pages/Interlaboratory-Studies.aspx]. The simulated aerosol filters contained Cs-134, Cs-134 (IAEA-TEL-2019-03 and IAEA-TEL-2020-05) and Ag-110m and Se-75 (IAEA-TEL-2020-03).

Three existing efficiency calibration curves were used for the reference efficiency: spiked charcoal in cylindrical geometry of 100 ml filled to a full, spiked mineralized grass in cylindrical geometry of 100 ml filled with 6.03g of matrix, and 50 ml vial, filled with 4.22g of aerosol [6]. The charcoal efficiency curve was used as it has the similar composition and density, the grass had the closest measurement geometry and the aerosol was used because it is readily used for the measurement of the prepared aerosol filters in the laboratory.

The simulated aerosol filters were measured on 2 p-type HPGe detectors. The duration of the measurement was 5100 s, 60000 s and 240000 s for the filter from IAEA-TEL-2019-03, IAEA-TEL-2020-03 and IAEA-TEL-2020-05 respectively. After the measurement, the activity of the present radionuclides was calculated using the grass matrix reference efficiencies (as it was the closest with the respect to the measurement geometry) in order to obtain the uncorrected results. Then the efficiency transfer was performed using EFFTRAN and the calculated transfer factors were applied in order to obtain the corrected result. Both uncorrected and corrected results were compared to the target value provided by the IAEA in the final report of the said Proficiency tests.

The uncorrected results, the corrected results and the target value for one simulated aerosol filter from each Proficiency test are presented in the Table I

TABLE I
THE RESULTS OF THE SIMULATED AEROSOL FILTER MEASUREMENTS USING DIFFERENT EFFICIENCIES AND THE TARGET VALUE, THE RESULTS WERE GIVEN WITH THE APPROPRIATE MEASUREMENT UNCERTAINTY, COVERAGE FACTOR 1

Element	**Uncorrected result [Bq/sample]**	**Charcoal to filter efficiency transfer [Bq/sample]**	**Grass to filter efficiency transfer [Bq/sample]**	**Vial to filter efficiency transfer [Bq/sample]**	**Target value [Bq/sample]**
IAEA-TEL-2019-03					
Cs-137	17.8 ± 0.7	9.6 ± 0.5	12.7 ± 0.6	13.1 ± 0.6	**13.02 ± 0.40**
Cs-134	21 ± 2	15 ± 1	20 ± 2	21 ± 2	**20.28 ± 0.61**
IAEA-TEL-2020-03					
Se-75	51 ± 2	23.5 ± 1.1	31 ± 2	29 ± 1	**31.3 ± 1.5**
Ag-110m	57 ± 2	30 ± 2	35 ± 2	35 ± 2	**35.1 ± 3.0**
IAEA-TEL-2020-05					
Cs-137	47.7 ± 0.8	25 ± 1	31 ± 1	29 ± 1	**28.6 ± 1.5**
Cs-134	27 ± 1	16.1 ± 0.7	20.4 ± 0.9	19.1 ± 0.9	**20.5 ± 1.1**

As it can be seen from the Table I, the uncorrected results differ significantly from the ones obtained using the efficiency transfer, although the composition of the mineralized grass (mainly cellulose and carbon) and the geometry were similar. Also, the transfer from the reference efficiency with the coal matrix produced the results that are significantly lower than the target value, meaning that the obtained efficiency is significantly overestimated. This can be explained by the large difference between the geometry of the reference efficiency which has greater diameter and sample height and therefore is the most diverse from the measured sample. Contrary to that, the transfer from the other two reference efficiency curves produced the results that are in agreement with the target values. For the elements that have multiple gamma lines, the coincidence correction factors were obtained using also EFFTRAN software. As it can be seen, the values for Cs-134, Se-75 and Ag-110m which are corrected for the coincidence summing effect and efficiency transfer from the grass reference efficiency proved to be the closest to the target value. For Cs-137, which has only one gamma emission and do not require coincidence summing correction, better results are obtained by transferring the aerosol reference efficiency. There is a local minimum at the energy of 661 keV in all efficiency curves regardless of the matrix of the calibration source. This leads to underestimation of the efficiency for this energy, which in turn produces an underestimated transferred efficiency. The recommendation for this energy is to use the efficiency obtained directly from the calibration source measurement, rather than from the calibration curve. Also, it is evident that the aerosol calibration source, although it closely represents the real aerosol samples, is not the best choice for the simulated aerosol filters which have an atypical geometry and composition. The mineralized grass calibration source proves to be the best reference calibration for the efficiency transfer since its diameter is very close to the diameter of the measured sample and more important, its thickness and chemical composition are virtually the same.

All the results obtained by using the efficiency transfer from the grass and aerosol matrix are acceptable, while none of the uncorrected results are acceptable. This obviously proves that the efficiency transfer has to be performed with the adequate reference calibration curve.

III. CONCLUSION

In this paper we presented the optimization of the efficiency calibration of HPGe detectors for the measurement of the simulated aerosol filters, measured within three Proficiency tests organized by IAEA. In case of the atypical geometry and composition of the measured sample, the efficiency transfer is inevitable, since the uncorrected activities are not in agreement with the target values, although the calibration source used for the efficiency calibration is of the similar geometry and composition. The choice of the reference efficiency curve for the efficiency transfer should be based on the similarities between the thickness and composition of the calibration source and the measured sample, since this choice produces the best results.

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