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PARTICIPATION OF THE LDPM, IFIN-HH, IN THE PROFICIENCY TEST ON A RADIOACTIVE SOLUTION, USING THE RAPID SAMPLE EVAPORATION METHOD*

ANA STOCHIOIU*, AURELIAN LUCA, MARIA SAHAGIA

"Horia Hulubei" National Institute for R&D in Physics and Nuclear Engineering, IFIN-HH, Bucharest, 30 Reactorului Str. Magurele, Ilfov County, code 077125, Romania
*Corresponding author: stoc@nipne.ro

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Abstract. Participation of the Dosimetry Laboratory for Personnel and Environment Laboratory (LDPM) from IFIN-HH in the AQUACHECK-2011 proficiency test (PT), organized by the LGC Standards, UK, is presented, in terms of the following aspects:

– Type of sample: radioactive solution, in the domain of low radioactivity concentration, consisting from a mixture of unknown radionuclides.

– Type of required measurements and report was the gross alpha and beta equivalent radioactive concentration: Alpha – ^{239}Pu and ^{241}Am; Beta – ^{40}K, ^{137}Cs and ^{90}Sr.

– The method of processing of the solution, in order to perform activity measurements. The new scientific information consists in the fact that the measurements implied two parallel independent methods. They are intended to check their validity, by the comparison of the results: i) A preliminary gamma-ray spectrometry measurement, to identify the gamma-ray emitters in the mixture and to determine their content; ii) Measurement of the gross alpha and respectively gross beta activity of the prepared samples, applied to thick samples, with the use of selfabsorption corrections, using the equipment calibrated in response for all radionuclides from the list provided by LGC Standards; iii) The third step in PT was the processing of the measurement data, by applying the necessary corrections and reporting the results. The sample was slowly evaporated to dryness in order to obtain a constant mass, at a temperature under 90 ^{o}C, to avoid loss of radionuclides.

After the transmission of the measurement results, from all the 11 participating laboratories, the organizer of the PT, LGC Standards, processed the received values and published the following data: the reported values of the laboratories and the accomplishment of the z'-score performance. Our laboratory, code AQ2757, obtained satisfactory results, passing the z'-score test for all the reported values. On the other side, the comparison of the two methods: gamma-ray spectrometry and gross alpha, beta measurement confirms the agreement between the results obtained for ^{241}Am and ^{40}K.

Key words: gross alpha and gross beta, gamma-ray spectrometry, activity measurement, proficiency test.

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1. INTRODUCTION

The Dosimetry Laboratory for Personnel and Environment (LDPM) from IFIN-HH deploys activities for dosimetric personnel survey and monitoring of the environment radioactivity [1]. The monitoring is accomplished by the measurement of the radioactive concentration of soil, water, vegetation, sediment and aerosols [2]. The laboratory is accredited by the national accreditation body, RENAR, and designed by the nuclear authority CNCAN, according to EN ISO/IEC 17025:2005 standard, for low level activity measurement and reporting. One of the mandatory requirements for the accredited laboratories is to prove their technical expertise in the field, by participation at proficiency tests (PT) or Inter Laboratory Comparisons (ILC).

The work and the results obtained by the LDPM at the AQUACHECK 2011-PT, organized by the LGC Standards, UK, are presented. The exercise consisted from the conditioning for analysis of an acidulated water sample and the measurement of the gross alpha and beta activity concentration in equivalents of ^{239}Pu, ^{241}Am (for alpha) and respectively ^{40}K, ^{90}Sr and ^{137}Cs (for beta). As it is well known, the standard method consists in the radiochemical separation of alpha and beta emitters and preparation of the measurement samples by electroplating, or by precipitation as a very thin deposit [3, 4]. The paper presents a more rapid and simple method for preparation and measurement of samples; of course, it is less precise than the standard one, but it can be useful when not advanced radiochemistry is available in the laboratory [5]. On the other side, the complementary gamma-ray spectrometry measurement results confirmed the validity of the method and allowed us to state the real radionuclidic composition of the sample. The aim of this work is to demonstrate that a simple, rapid method of preparation and measurement may be used, with satisfactory results.

2. PREPARATION AND CONDITIONING OF SAMPLE

The sample received for analysis consisted of 2 L of 0.5% nitric acid solution. It was evaporated slowly at a temperature under 90 ^{o}C, till the constant mass, in order to avoid the loss of radioactive particles. The evaporation was made in a porcelain vessel until a dry residuum was obtained. The residuum was then crushed and carefully transferred on the measurement support; the control of the recovery yield was done by parallel weighing of the vessel before and after the transfer of the residuum and of the support before and after the transfer with an analytical balance model WAX 220, and comparison of the two obtained values. The measurement support consisted from a stainless steel plate with the useful diameter of 50 ± 2 mm and height of 6.0 ± 0.2 mm, as a spare part of the measurement equipment. The amount of recovered residuum, dry mass, was 1.3946 g. The residuum was then uniformly stretched on the entire surface of the

plate and then covered with 1 mL of acetic acid, to avoid its spreading in the windowless counter volume during the measurement.

3. MEASUREMENT EQUIPMENT AND ITS CALIBRATION DESCRIPTION

– The installation of gamma-ray spectrometry of the Radionuclide Metrology Laboratory (RML) from IFIN-HH, with HPGe detector has the following components: The Hyper-Pure Germanium semiconductor detector (HPGe), Model GEM25P4, is introduced in a graded shielding made of lead (10 cm thick), tin (1 mm) and copper (1 mm), in order to reduce the background radiations. The main technical parameters of the system are (measured values): energy resolution (FWHM) of 1.67 keV at 1.33 MeV (^{60}Co) and 0.64 keV at 122 keV (^{57}Co); relative efficiency 28.9 %; the peak-to-Compton ratio, ^{60}Co, is 62:1. The analysis system consists from: A Digital gamma-ray spectrometer (including a digital signal processor, «DSP») ORTEC, model DSPEC ^{PLUS} and a Personal Computer (PC) UltraPro. Three software's are implemented in the system: ORTEC GammaVision-32; ORTEC MAESTRO-32 for operation and processing of data. GESPECOR, for the computation of coincidence summing corrections and for the transfer of efficiency calibration from a measurement geometry to another and from a source geometry and matrix to another one. The energy and efficiency calibrations of system were done with gamma-ray spectrometry standard sources produced by the RML. The measurement method by gamma-ray spectrometry was validated in many international comparisons [6, 7].

– Two gamma-ray spectrometry installations for the measurement of low activities belonging to the LDPM.

– Installation for measurement of gross alpha-beta activity in ultra low background, Model 9300 PC-GFL, provided with a windowless proportional counter and a Soft VISTA 2000. Background counting rates are: alpha: 0.050 ± 0.041cpm and minimum detectable activity (MDA) is 0.010 Bq; beta: 0.600 ± 0.141cpm and MDA is 0.029 Bq, for a measurement time of 30 minutes.

– Installation for measurement of gross alpha, beta, gamma activity in low background, with automatic sample changer Model S 5 XLB-G, Soft ECLIPSE. Minimum detectable activities are: alpha 0.010 Bq, beta 0.030 Bq and gamma 1.12 Bq, for a measurement time of 30 minutes. They were calibrated by the Radiation Metrology Laboratory from IFIN-HH (CMRID), using ^{239}Pu, ^{241}Am, ^{40}K, ^{90}Sr and ^{137}Cs standard sources certified by the RML. The counting efficiencies for the two installations are presented in Table 1.

Table 1

The counting efficiencies for the two installations

Installation	Alpha efficiency [s^{-1}/Bq]		Beta efficiency [s^{-1}/Bq]		
	^{239}Pu	^{241}Am	^{40}K	^{90}(Sr+Y)	^{137}Cs
Model 9300 PC-GFL	0. 458 ± 0.010	0.480 ± 0.010	0.440 ± 0.012	0.425 ± 0.010	0. 400 ± 0.013
Model S 5 XLB-G	0. 330 ± 0.003	0.339± 0.003	0.395± 0.010	0.369 ± 0.003	0.345± 0.021

4. MEASUREMENT OF SAMPLES

4.1. GAMMA-RAY SPECTROMETRY MEASUREMENTS

They were done in order to determine the content of gamma-emitters, like ^{241}Am, ^{40}K and ^{137}Cs. The total activity of the solid residuum was measured and the value was divided by 2, in order to obtain the activity concentration in Bq/L. The initial scan of the liquid sample was difficult to be done with a 2L volume of solution.

The results were the followings: ^{241}Am – (0.23 ± 0.03) Bq/L, ^{40}K – (0.79 ± 0.12) Bq/L and ^{137}Cs – under the MDA: 0.04Bq; uncertainties are given for a coverage factor, k =1. If all cited radionuclides in the list were present in the sample, it should be expected that the activities expressed as gross alpha and beta equivalent be different from the gamma-ray spectrometry result, as the equivalent gross activities express the contribution of all alpha or beta emitters, not only the respective radionuclide.

4.2. MEASUREMENT WITH THE INSTALLATIONS FOR GROSS ALPHA-BETA ACTIVITY

The reported measurements were performed with the equipment Model 9300 PC GFL.

The counting rates were calculated, after the extraction of the background, determined values. For a moment no special radon free system or fluxing nitrogen facility was used; however, the use of P10 flowing gas practically eliminates the radon from measurement. The superficial activities in equivalent radionuclides, representing the activity of the thin surface layer, were calculated by dividing the counting rates to the respective counting efficiencies and considering the emission probabilities of the respective radiations, according to their decay schemes.

4.3. APPLICATION OF SELFABSORPTION CORRECTION AND EVALUATION OF THE REAL ACTIVITIES

The surface density of the sample was 71.062 mg·cm^{-2}, what means that thick samples were measured and the selfabsorption correction had to be done. The treatment was different for the two types, alpha and beta radionuclides, due to the different type of selfabsorption in the dry mass of the residuum.

In the case of alpha evaluation, taking into account the maximum ranges for the radionuclides ^{239}Pu and ^{241}Am, it was considered that the sample corresponds to $n = 14.6$ and respectively $n = 13.9$ active emitting layers from the two radionuclides. The final calculation formula was:

$$A_{equivalent} = N_\alpha n/(2\varepsilon_\alpha s) \text{ [Bq/L]}. \quad (1)$$

In relation (1), $N_\alpha = 0.015$ s^{-1}; n is the number of absorbing layers; ε_α are taken from Table 1; s =1 is the emission probability for both radionuclides.

In the case of beta rays, it was considered an exponential expression for the self absorption of beta rays in the sample, with the attenuation linear coefficients equal with the reverse maximum range of mean energy beta rays of the three radionuclides ^{40}K, ^{137}Cs and ^{90}Sr. The total activity was calculated by amplifying the surface activity with the absorption correction, f, respectively f =1.593; 1.511; 1.507 for the three radionuclides. The final formula for calculation was:

$$A_{equivalent} = N_\beta f/(2\varepsilon_\beta \text{ s}) \text{ [Bq/L]}. \quad (2)$$

In relation (2), $N_\beta = 0.361$ s^{-1}; s is the emission probability: s = 0.893; 1.00; 2.00 for the three radionuclides.

4.4. UNCERTAINTY EVALUATION

The most important uncertainty was due to the selfabsorption corrections, as some simplifying assumptions were done, regarding the absorption formulae. The other uncertainties are due to counting statistics, background contribution, installations' efficiency uncertainty, mass recovery. The combined standard uncertainty (k = 1) was calculated by quadratic summation of all the components. Table 2 presents the uncertainty budget.

Table 2

Uncertainty budget

Component	Type of evaluation	Relative uncertainty, %
Equipment calibration	B	5.0
Selfabsorption:	B	18 for alpha; 17 for beta
Statistics	A	15
Background	B	10
Weighing	B	0.1
Solid mass recovery	B	0.5
Combined standard uncertainty, u_c	Quadratic summation	26 (α) 25 (β)

5. RESULTS OF THE LDPM IN THE PARTICIPATION AT THE PT, AQUATECK PROFICIENCY SCHEME. ROUND 408

The obtained values of activity concentration of solution in Bq L^{-1} were reported on line to the PT organizer. After the finalization of the entire PT, with the reception at the LGC Standards, UK, of the results from all participants, the evaluation of results was sent to us by the organizer. For the evaluation of the quality of the LDPM result, Table 3 presents the essential data according to the published report of LGC Standards, AQUACHECK Proficiency Scheme AQ 2757, IFIN-HH, Individual Report, Round 408, Issued Number 1, 15 July 2011 (http://portal.proficiencytestingschemes.com/Members/Results/Summary.aspx).

The *z*' score for acceptance of a result was applied

Table 3

Results of the participation of LDPM at the PT

Analyte	LDPM result [Bq L^{-1}]	Assigned value [Bq L^{-1}]	*z*′ score
Gross Alpha as ^{239}Pu	0.232 ±0.058	0.297 ±0.085	– 0.63
Gross Alpha as ^{241}Am	0.218 ±0.055	0.270 ±0.018	– 0.91
Gross Beta as ^{40}K	0.732 ±0.183	0.611 ±0.017	+0.99
Gross Beta as ^{137}Cs	0.682 ±0.170	0.957±0.126	–1.20
Gross Beta as ^{90}Sr	0.320 ±0.080	0.236±0.100	– 0.70

The discussion of the Table 3 results:

– All the reported concentration values passed the *z*′ score; the activity concentration value and the uncertainty of results were both realistically evaluated;

– The sample does not contain ^{239}Pu, although ^{239}Pu equivalent activity was calculated. The ^{241}Am activity determined by gross alpha and by gamma-ray spectrometry methods agrees within the limit of uncertainties.

– The sample really does not contain ^{137}Cs and maybe ^{90}Sr, although equivalent ^{137}Cs and ^{90}Sr activities were calculated. ^{137}Cs was not identified in the sample, and the ^{40}K equivalent activity determined by gross beta counting is in agreement with the gamma-ray spectrometry result; ^{90}Sr should be in equilibrium with its daughter ^{90}Y, and the influence in the counting rate should be due to both radionuclides; this value was not found.

Finally, we conclude that the sample contained only ^{241}Am and ^{40}K.

6. CONCLUSIONS

This paper presents a simple method of evaluation of the alpha- and beta-gross activity, based on the recovery of the entire radionuclidic content of a sample, the measurement using counters for gross alpha and beta activities, and the evaluation of corrections to be done.

It is recognized that the method is less precise than the standard one, but it can be applied in the case of the lack of advanced radiochemistry and when the results must be reported quickly.

The method is validated by the results obtained within the participation at the AQUACHECK 2011-PT, organized by the LGC Standards, UK.

The agreement between the equivalent gross alpha as ^{241}Am and gross beta as ^{40}K activities, determined by the two methods, confirmed the correctness of the applied selfabsorption correction.

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