Participation of the IFIN-HH Dosimetry Laboratory for Personnel and Environment (LDPM) at the Proficiency Test AQUATECK-2011

Ana Stochioiu* , Aurelian Luca, Maria Sahagia, Ion Tudor

Horia Hulubei National Institute for R&D in Physics and Nuclear Engineering, IFIN-HH, Bucharest. 30 Reactorului str. Magurele, Ilfov County, code 077125, Romania

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 notified 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.

2. Preparation and conditioning of samples

The sample received for analysis consisted of 2 L of 0.5% nitric acid solution. It was evaporated slowly, in standard conditions, in order to avoid the loss of radioactive particles from it, until a dry residuum was obtained on the evaporation porcelain vessel walls. It 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. 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 [5, 6].

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, Soft VISTA 2000 of L. Background counting rates are : alpha : 0.050±0.041cpm and minimum detectable activity is 0.010Bq; beta : 0.600±0.141cpm and minimum detectable activity (MDA) is 0.029Bq.

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 0.010Bq , beta 0.030Bq and gamma 1.12 Bq.

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 effciencies 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	45.8 ± 1.0	48.00 ±1.00	44.0±1.2	42.5 ± 1.0	40.0 ± 1.3
Model S 5 XLB-G	33.00 ±0.30	33.91±0.34	39.5±1.0	36.9 ± 0.34	34.5 ± 2.1

4. Measurement

4.1 Gamma-ray spectrometry measurements

They were done in order to determine the content of gamma-emitters, like 241Am, 40K and 137Cs. 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 results were the followings: ^{241}Am – (0.23±0.03) Bq/L, 40K – (0.79± 0.12) Bq/L and ^{137}Cs - under (MDA): 0.04Bq. It is 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 counting rates, after the extraction of the background, were determined for the equipment Model 9300 PC-GFL. 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 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 absorption 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:

$$A_{equivalent}=N_{\alpha}n/(2\varepsilon_{\alpha}\,s),\ \text{Bq/L} \qquad (1)$$

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:

$$A_{equivalent}=N_{\beta}f/(2\varepsilon_{\beta}\,s),\ \text{Bq/L} \qquad (2)$$

In relation (2), N_{β} = 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 absorption 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.

5. Results of the LDPM in the participation at the PT, AQUATECK Proficiency Scheme. Round 408

The obtained values of activity concentration of solution, 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 2 presents the essential data. The z' score for acceptance of a result was applied.

Table 2 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**

Discussion of the Table 2 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.

5. 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.

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