OPTIMIZATION OF GROSS ALPHA AND BETA DETERMINATION USING A LOW BACKGROUND GAS-FLOW PROPORTIONAL COUNTER

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Abstract

Gross alpha/beta measurement is one of the simplest radioanalytical procedures which are applied widely as a screening technique in the field of environmental monitoring. Due to the uncertainties of gross alpha/beta measurements this method is often the subject of discussions and debates. The self-absorption of alpha and beta particles in the sample is the main drawback of the method due to the substrate thickness. To account for this effect, the attenuation correction curves were determined from the measurement of standard sources prepared with different known thicknesses. The paper presents the results of an experimental test for determination of self-absorption corrections for gross alpha and beta counting using a low background proportional counting system, in case of increase salt content.

Keywords: *Environmental monitoring, Gross alpha/beta measurements, Self-absorption corrections*

Introduction

One of the main parts of environmental studies is represented by determination of the quality of drinking water, taking into consideration that it may contain radioactive substances which are harmful to human health. Radioactivity in trace amount is normally found in all kinds of drinking water, which may vary from place to place depending on the radiochemical composition of the soil and rock strata through which the raw water has passed. Several naturally occurring and man-made sources present in the environment result in radioactivity of water, and it is quite expensive as well as time consuming to identify individual radionuclides. Therefore, gross alpha and gross beta analyses are used as the first step of the radiological characterization of drinking waters, as screening methods in the field of radioprotection, being a relatively inexpensive and readily available [1].

Gross alpha and beta activity give no information regarding the isotopic composition of the sample, therefore cannot be used to estimate radiation exposure, but it is one of the radiological parameters that must be determined in order to decide whether further specific analyses of radionuclides in samples are required [1].

When the alpha and beta emissions exceed the guideline values, determination of the concentration of individual radionuclides is conducted. World Health Organization, WHO, guidelines for drinking water quality recommend levels of 0.5 Bq l^{-1} for gross alpha and 1 Bq l^{-1} for gross beta activities, respectively [2]. According to Romanian laws, the acceptable limits for radioactive substances in drinking water are 0.1 Bq l^{-1} for gross alpha activity and 1.0 Bq l^{-1} for gross beta activity, respectively [3].

If the measured values are below the reference levels given for radiations gross activity, the drinking water examined is acceptable for human consumption without any further action with respect to its radioactivity.

Gross alpha and beta activities measurements can be made using several sample preparation techniques and counting methods. The U.S. Environmental Protection Agency, EPA, Method 900.0 is approved for these measurements due to its simplicity and quantitative recovery. The method is based upon evaporation to dryness of a liquid sample, followed by activity measurement of the residue deposited on a planchet, by using a proportional detector [4]. This method sensitivity is strongly affected by the total dissolved solids in water and the sample chemical composition as well. The method requires that the sample aliquant be limited to what will produce 5 mg/cm^2 of solids on the planchet. Thus, for a 50.8 mm diameter planchet (~20 cm^2), an aliquot containing 100 mg of nonvolatile dissolved solids is the maximum test-source mass [5].

For the gross alpha detection in water samples with dissolved solids content greater than 500 mg/*l*, the co-precipitation method is recommended to be used. This method co-precipitates alpha-emitting radionuclides using barium sulfate and iron (III) hydroxide where the solid is collected onto a filter for subsequent counting.

Both sample preparation methods are primarily directed for use in drinking water samples [5]. Besides these standard methods, some alternative techniques, either based on liquid scintillation counting or the combination of alpha and gamma-ray spectrometry have been developed in order to overcome the drawbacks of the standard methods [1].

Materials and methods

The equipment used for this research work is a low background gross alpha and beta counter, model MPC 9300. It is a counting system that provides true ultra-low background performance for applications with low sample throughput requirements. Simultaneous or separate alpha plus beta counting modes provide high efficiency detection in a single counting cycle [6].

Counting geometry, sample self-absorption and counting efficiency

Geometry plays an important role in counting efficiency, so it is important to match the geometry of the calibration source to that of the samples to be counted.

One aspect to be considered for the counting geometry is the distance between the sample and the active volume of the detector. Alpha and beta particles emitted at small angles from thin samples placed in deep planchets will lose energy in interaction with the planchet walls [6].

Another important aspect in the efficiency calibration process is the homogeneity of the source. The calibration sources should be prepared in a manner that reduces the non-uniformity of the material. Any deviation from this requirement can lead to biased results and contribute to the overall uncertainty of the laboratory results. Source uniformity is related to the physical nature of the source material. Uniformity of source material relative to its thickness, density (which can be influenced by water content), and homogeneity is important. If sources are deposited in a non-uniform manner, the absorption characteristics will vary from source to source, and acceptable reproducibility may not be achieved [7].

Experimental tests and results

Gross alpha counting efficiency

Alpha particles are characterized by an intense, high rate of energy loss while passing through matter, meaning that the attenuation or self-absorption corrections may be necessary for alpha counting. Attenuation corrections should be made whenever the test-source matrix differs from that of the calibration source. In order to determine the change in counting efficiency as a function of source thickness or mass, a self-absorption curve must be developed [7].

In the present paper the absorption curve for gross alpha-particle measurements was constructed using as reference material Am-241. Calibration sources containing a known amount of the radionuclide of interest were prepared by varying the thicknesses (masses) and counted. A known amount of radioactive solution containing the reference radionuclide was pipetted into a 50.8 mm diameter stainless steel planchet. The planchet was evaporated using an infrared lamp. After evaporation, the counting efficiency (ε) was determined by counting the source to accumulate sufficient net counts so that the statistical uncertainty is satisfactory and dividing the resultant net count rate (cps) by the alpha emission rate of the source:

$$\varepsilon = \frac{Measured\ Net\ Count\ Rate\ (cps)}{Corrected\ Activity\ of\ Source\ (Bq)}$$

The absorption curve is constructed by counting planchets containing varying mass of material but with a known amount of added radioactivity. For this purpose a saline solution containing $CaCl_2$ was prepared in a separate vial, the concentration of salts in the solution being around 0.3 g/ml. Small amounts of this saline solution were added repeatedly to the planchets over the radioactive solution. After each addition the sample was homogenized, dried under the infrared lamp, weighed to determine the masses of the dry residues and counted to determine the counting efficiency. Thus, the dependence of the efficiency on the mass of salts in the planchets was determined. The ″zero thickness″ source and one of the sources mass could be seen in Fig. 1.

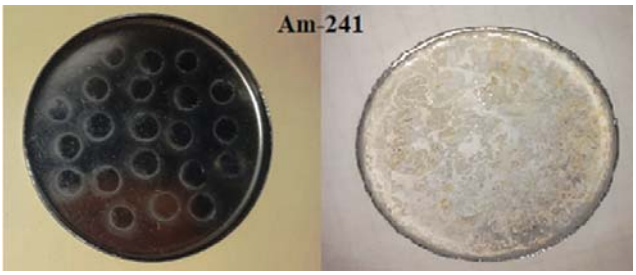


Fig. 1. Calibration sources (zero thickness and thick sources)

The self-absorption curve was generated by determining the counting efficiency as a function of mass density of the source (in g/cm^2), as one can see in Fig. 2.

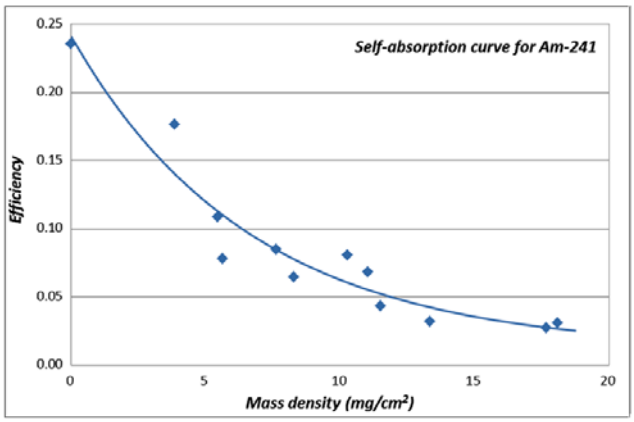


Fig. 2. Gas-flow proportional counter self-absorption curve for Am^{241}

The study illustrates the rapid reduction in efficiency as sample mass density increases. An important consideration in the analysis of charged particle emitting radionuclides consists in the energy absorption. When the sample is concentrated by evaporation, detection is limited by the energy absorbed of the remaining dissolved solids. It can be seen from Fig. 3 that the optimal amount of total dissolved solids on the planchet for obtaining a minimum value of the minimum detectable activity concentration, in a given measurement time, is approximately 7.5 mg/cm^2.

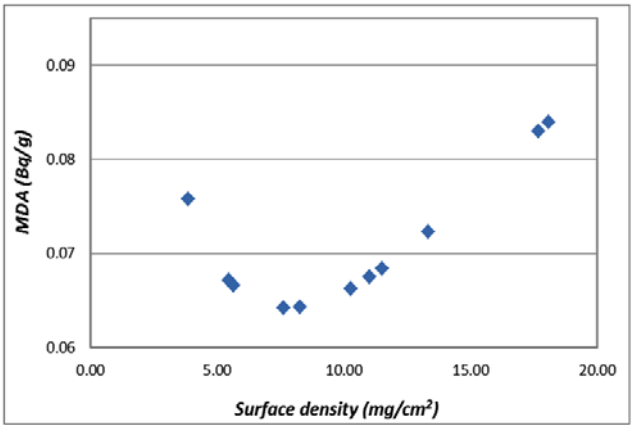


Fig. 3. Minimum detectable activity concentration

Gross beta counting efficiency

Radioactive decay by beta particle emission is generally accompanied by one or more gamma-ray emissions; the latter normally is much easier to identify and quantify. Beta-particle counting typically is more difficult, because of the additional source preparation and associated complications resulting from the effects of backscatter, scattering, and absorption in the source material. In cases where finite test-source thicknesses are unavoidable, beta-source measurements can be adjusted to account for self-absorption [7].

For gross beta-particle measurements, the absorption curves were constructed in the same way as for alpha particle. The reference material was Sr/Y^{90} and the planchet used for source preparation being of 29 mm in diameter and having a height of 7 mm. It was chosen to determine the counting efficiency using the mentioned geometry because is most commonly used for gross beta measurements in the Radiation Protection Laboratory at the RATEN ICN Pitesti. The self-absorption curve for Sr/Y^{90} was generated by determining the counting efficiency as a function of mass density source, as is illustrated in Fig. 4.

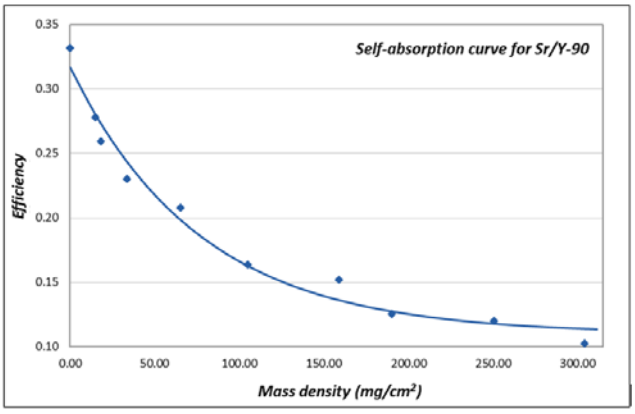


Fig. 4. Gas-flow proportional counter self-absorption curve for Sr/Y^{90}

Self-absorption of beta particles is not as pronounced as was for alpha particles, because the charge and mass of beta particles are significantly smaller.

For sufficiently thick sources, the number of beta particles interacting with the detector will reach a limit and the count rate becomes independent of the source thickness. To study the dependence of the net counting rate on the concentration of the activity, four calibration sources containing about 3 g of radioactive talcum powder spiked with Sr/Y^{-90} were prepared.

The sources were prepared as follows: in 4 glass vials were placed known amounts of talcum powder, over which different amounts of radioactive solution were added, resulting in sources with different activity concentrations. The vials were dried on the sand bath and under the infrared lamp, and after total evaporation these were vigorously mixed to homogenize the talcum powder. Known amounts of radioactive powder from each glass vial were transferred to four planchets so that the sources had the same thickness, the planchets being almost full. Each source was counted using the gas flow proportional counter system for a time range long enough to assure a satisfactory statistical uncertainty.

The dependence of the net counting rate on the activity concentration is represented in Fig.5.

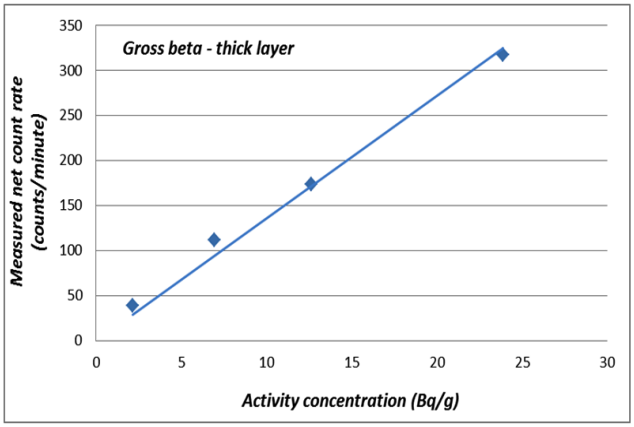


Fig. 5. Dependence of the net counting rate on the activity concentration

A good way for assessing the analytical methods performances of a laboratory is participating in proficiency tests. Proficiency tests represent an effective way to ensure the quality of measurements and to improve measurement methods. Hereby, the RATEN ICN's Radiation Protection Laboratory was able to check and validate the gross alpha and beta determination methods by regularly participation in the ALMERA Proficiency Tests organized by the International Atomic Energy Agency, IAEA, on the determination of natural and anthropogenic radionuclides in different types of samples (water, fish, soil, shrimps, etc). The gross alpha/beta measurements for the most recent proficiency test were performed using the gas-flow proportional counter. The samples for which it had to be performed the gross alpha/beta determination consisted in two spiked water samples and one fish sample. The reported results and the evaluation from proficiency test are presented in the table below.

Table 1 ALMERA Proficiency Test results – gross alpha/beta, 2020

Sample Code	Sample Type	Analyte	Robust Mean	Robust SD	Reported Value	Reported Uncertainty	Z-Score	Z-Score Evaluation
1	Spiked water	Gross beta	170	27	152.7	22.9	0.64	A
2	Spiked water	Gross alpha	40	14	29.7	4.5	0.74	A
2	Spiked water	Gross beta	38	7	40.3	6	0.33	A
4	Fish	Gross alpha	169	126	297	89	1.02	A
4	Fish	Gross beta	597	186	488	146	0.59	A

Conclusions

Sample residue thickness and uniformity are crucial for accurate gross alpha/beta counting using a gas-flow proportional counter. The self-absorption of the alpha and beta in the sample matrix was analyzed for a typical gas-flow proportional counter in case of increase salt content. In both cases, gross alpha and beta, the counting efficiency data were fitted with empirical exponential law functions. Correctness of the used methods was confirmed by the results and the evaluation from the most recent proficiency test organized by the IAEA-ALMERA network in 2020.

References

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