2021 International Nuclear Atlantic Conference – INAC 2021
Virtual meeting, Brazil, November 29 – December 2, 2021

ISBN: 978-65-594-1256-3

Counting Time Optimization for Gamma-Ray Spectrometry Analysis: A Review of Published Scientific Papers

M.B. Nisti[1], M.F. Máduar[1], C.H.R. Saueia[1] and F. Cavalcante [1]

[1]*mbnisti@ipen.br*, [1]*mmaduar@ipen.br* [1]*chsaueia@ipen.br*, [1]*fcavalcante@ipen.br*
Instituto de Pesquisas Energéticas e Nucleares, IPEN/CNEN

1. Introduction

Gamma-ray spectrometry is a powerful, non-destructive, fast, and reliable analytical measurement technique, widely employed for determining the contents of gamma-ray emitter radionuclides. As a result of applying this technique, a count vs. energy spectrum is obtained for each sample.

Good practices for their acquisition must be established for ensure high-quality spectrum, including physical setup, electronic settings, system calibration with certified standards, use of recommended gamma-ray energies and intensities, counting conditions, corrections for unwanted sources of radiation and proficiency testing for validation [1, 2, 3].

Adequate counting time is a very important step in using gamma-ray spectrometry; this parameter setup depends on each particular application, such as, sample type, radionuclides to be determined, radiation background of laboratory, counting geometry, detector features, shielding and associated electronics.

Several circumstances must be considered in order to optimize the system utilization with minimum counting time and acceptable detection limits for analysis of gamma-ray emitters.

Acceptable minimum detectable activity (MDA) value in the analysis as well as quantifying low radionuclide concentration needs to be considered as a criterion to define counting time for gamma ray spectra acquisition. When the counting time is not correctly determined, it can generate high uncertainty in concentrations.

Three basic requirements can be considered for increasing the counting time: decrease the value of the MDA, improve the peak statistics and decrease the uncertainty of the measurement.

Increasing the counting time decreases the uncertainty resulting in a better statistic. However, depending on the laboratory infrastructure, large number of samples can restrict the measurement time; in this case there is a compromise: what is the minimum counting time to use in order to guarantee quality and reliable results with adequate uncertainty? This review of published scientific articles shows possibilities to contribute with appropriate alternatives for solving the problem.

The gamma spectrometry analysis laboratory can have a problem when large amount of sample needs to be analyzed in a short period of time. In this case, two criteria can be used to solve the problem: increase the laboratory's infrastructure or optimize the counting time in the measurement.

The increase in infrastructure is an alternative to solve the problem, preparing the laboratory to receive a larger amount of sample, thus allowing the analysis to be carried out in the desired time, but this alternative often has a high cost and need available time before starting the detection system operation: purchase, wait for the receipt, installation, adequate local inside the laboratory, calibration and validation of the new detection system.

Counting time optimization is the fastest and most economical alternative for solving the problem, being an important method for accomplishment of automation in routine measurements with acceptable detection limits and reduction in the measurement cost and time [4,5,6,7,8].

In this review, a collection of scientific papers published approaching studies on the counting time optimization using the gamma-ray spectrometry technique was carried out.

2. Methodology

This review has basically two objectives: to facilitate the search for papers on counting time optimization using the gamma-ray spectrometry technique and to collaborate in optimizing of productivity of a laboratory when processing large batches of samples. An important reason for the elaboration of this paper is that few papers justify the counting time that was used in the gamma-ray spectrometry analysis and the authors consider it a very important topic to be explored.

3. Results and Discussion

Summary of bibliographic review of scientific papers about counting time optimization for gamma-ray spectrometry analysis is presented below with the publication date in ascending order:

(a) ***Fast methodology for time counting optimization in gamma-ray spectrometry based on preset minimum detectable amounts*** **[4].** Nisti et al. (2009) evaluated the time counting optimization in gamma-ray spectrometry using four coaxial HPGe detectors with relative efficiency of 15% to 25%, for estimate the ‘‘a priori’’ counting times for a sequence of measurements required to meet the given MDA and estimated the counting times. The blank samples for background determination were prepared with deionized water; and samples with a mixed radioactive solution of ^{133}Ba, ^{134}Cs, ^{106}Ru, ^{137}Cs, ^{65}Zn, and ^{60}Co were prepared to check the values obtained from MDA, in four different counting geometries with volume of 100 mL, 850 mL, 1 L and 3 L; and seven different counting times were used: 1,000; 5,000; 10,000; 15,000; 50,000; 100,000 and 150,000 seconds, measured three times for all detectors. The counting time of 50,000 seconds was found to be sufficient to reach agreement between the preset and actual counting times; actual counting times obtained were in good agreement with the preset times for all the studied detection systems and counting geometries, and could be applied for a wide range of detection counting systems and sample geometries. Some care could be needed in applying of this proposed method as in relation to the composition and density of the samples.

(b) ***Determination of the optimal measurement counting time and detection limit for gamma-ray spectrometry analysis [5].*** Shweikani and Hasan (2015) evaluated the correlation between the minimum detectable activity and the measurement counting time in determination of the optimal measurement counting for gamma-ray spectrometry analysis. For this study it was used two HPGe detectors with relative efficiency of 60% (N-type) and 80% (P-type), thus determining the optimal measurement counting time. The background spectra were used to estimate MDA the artificial radionuclides ^{60}Co, ^{85}Sr, ^{131}I, ^{133}Ba, ^{134}Cs, ^{137}Cs, and ^{241}Am, and natural radionuclides ^{7}Be, ^{40}K, ^{224}Ra, ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{228}Ac, and ^{234}Th for the three different counting geometries with volume 1000 cm^3 (Marinelli beaker), 27 cm^3 and 125 cm^3 (two cylindrical containers) and different counting times were used in a range of the 1 until 40 h. The paper mentions for gamma spectrometry analysis of environmental samples with low radioactivity the measurement the parameters must be perfectly balanced to decrease the cost of analysis, such as: increasing the sample quantity, increasing the counting time, perfecting the sample geometry and efficient shielding; but these solutions have its limitations, and extend a certain parameter provides no additional benefit. The authors this paper the optimal counting time was considered value obtained in the derivative (MDA)´ is as low as -0.05 Bq kg^{-1} h^{-1} (almost constant level and approaches zero).

(c) ***Optimal measurement counting time and statistics in gamma spectrometry analysis: The time balance [6].*** Joel et al. (2017) determined the optimal counting times for a sequence of measurements required to meet the given MDA considering the spectrometer properties and counting physical geometry. The initial idea for this paper came by comparing two spectra for same sample counted in the same time but at two different moments, measures for 12 hours in the day and 2 hours in the night, but day spectrum does not fully cover the night spectrum. To establish the optimal measurement counting time, was used an HPGe detector with relative efficiency of 60%, were analyzed natural radionuclides (^{40}K, ^{212}Pb, ^{212}Bi, ^{214}Pb, ^{228}Ac, ^{232}Th, ^{214}Bi, ^{235}U and ^{226}Ra) and fourteen different counting times, were: 5 min; 10 min, 30 min, 45 min, 1 h, 2 h, 3 h, 4 h, 6 h, 12 h, 24 h, 36 h, 48 h and 72 h, were counted different reference samples with different activity concentration. The result of the analysis of the samples in the fourteen measurement times were: at the 5 min counting time, the only radionuclide above MDA was ^{212}Pb; 10 min counting times were ^{212}Bi and ^{40}K; 30 min were ^{214}Pb and ^{228}Ac; 45 minutes the ^{232}Th and ^{214}Bi are detected for the first time; and ^{235}U and ^{226}Ra were the last radionuclides to be detected in 3 hours, it was observed that the longer the counting time, the lower the uncertainty in the measurement. The authors emphasize that the estimation of results uncertainty is the most important issue in analytical techniques in gamma spectrometry, especially in low concentration samples, such as environmental samples. The conclusion of the paper: to detect natural radionuclides studied with uncertainty less than 1%, it to count was necessary with minimum duration of 24 hours.

(d) ***Measurement of naturally occurring radioactive material, ^{238}U and ^{232}Th: part 2-optimization of counting time [7].*** Naskar et al. (2017) studied the optimization of counting time for ray-gamma spectrometry technique in samples with low-level measurement of naturally occurring radioactive material (NORM), to perform the study were evaluated the standard deviation and counting error in the different counting time to determine the most significant counting time in samples with low-level radioactivity. The studied used an HPGe detector with relative efficiency of 50%, natural radionuclides in soil samples were analyzed used average of three photopeaks from ^{238}U and ^{232}Th series (^{238}U: photopeaks of daughter radionuclides ^{214}Pb and ^{214}Bi) and (^{232}Th: photopeaks daughter radionuclides ^{228}Ac and ^{208}Tl), and nine different counting times were used: 5,000; 10,000; 20,000; 30,000; 50,000; 75,000; 100,000; 125,000 and 150,000 seconds. This paper did a great research of bibliographic references of the application of the gamma spectrometry technique to verify the counting time used, there were 56 references, the authors observed that there was no agreement regarding the efficiency of the detector with the counting time used. Four soil samples collected and measured at all times for the determination of natural radionuclides (^{238}U and ^{232}Th), and two samples were prepared with activities of 2 and 5 Bq for validation and analysis of minimum counting time. To confirm the secular equilibrium in the samples, the activity of the three photopeaks was determined of ^{234}Th. In the paper, the appropriate time interval was 75,000 to 100,000 s, a regular trend was observed, with RCE values come closer to RSD, but for safety the chosen time was 100,000 s. Conclusion, the time required to stabilize the RSD values determines the minimum counting time for low-level NORM measurement. The authors recommend that the experimenter determine the minimum reasonable counting time considering the values of RCE and RSD in their own detection system.

(e) ***Optimization of the gamma-ray spectrometry counting time based on uncertainties of radionuclides concentration in samples [8].*** Nisti et al. (2020) established a method to optimize the counting time using the uncertainties in the concentration in acceptable levels, easy and fast method and with a good accuracy. Soil samples were sealed for about four weeks and using an HPGe detector with relative efficiency of 25% determined the concentrations of radionuclides^{210}Pb, ^{212}Pb, ^{214}Pb, ^{212}Bi, ^{214}Bi, and ^{228}Ac (in the range of 46.52 keV to 969 keV) and the preset times of 86,000 s and 150,000 s were used. Background spectra were collected with counting time of 500,000 s. For determination of the net peak area Multichannel Maestro software (Maestro, 1995) was used, and for assessment of concentrations and associated uncertainties WinnerGamma/Interwinner 6.01 (InterWinner, 2004) software was employed. The difference of uncertainties in the concentrations was determined through the comparison between the counting times for the same sample. The authors defined the value of 1% in the difference of uncertainties as the most appropriate, as it was not observed advantage in increasing the counting time; but each laboratory can establish the uncertainty differences for best performance of the detection system available.

The ratios obtained, resulting in uncertainty differences of 1% for each peak, were: 4 for 46.52 keV (^{210}Pb); 2 for 238.63 keV (^{212}Pb); 23 for 295.21 keV and 14 for 351.92 keV (^{214}Pb); 28 for 609.31 keV (^{214}Bi); 4 for 727.18 keV (^{212}Bi); 14 for 911.20 keV and 18 for 968.97 keV (^{228}Ac). When these values are obtained, the required measuring time has been achieved and the counting can immediately be stopped.

It is important to mention that the rates obtained in this paper change for each detector, requiring an individual study for each counting system.

4. Conclusions

This paper was a review of excellent published scientific papers on the study of counting time optimization for the gamma spectrometry technique. The scientific papers reviewed were concerned with obtaining quality and reliable results, used standards for validation and/or participation in proficiency tests.

The intention of this bibliographic review of published scientific papers was to disseminate the study of counting time optimization, as it is little discussed, thus helping the gamma-ray spectrometry laboratories to obtain a quick initial research on the subject.

The scientific papers presented different procedures for optimization of counting time in gamma spectrometry; determining the desired MDA or time required, deriving the adjustment function, counting several times the same sample for determining acceptable uncertainty, evaluating counting error and standard deviation and acceptable difference of uncertainty between measures. The different procedures for optimization of counting time can be used in order to help gamma-ray spectrometry laboratories routine, depending on the need or situation of each particular laboratory.

References

[1] M.M. Bé and V. P. Chechev, “Recommended standards for gamma ray intensities”, *Nucl. Instrum. Methods Phys*. vol. 728, pp.157–172, (2013). DOI: 10.1016/j.nima.2013.05.134

[2] R.J. Gehrke and J.R. Davidson, 2005. “Acquisition of quality g-ray spectra with HPGe spectrometers”. *Appl. Radiat. Isot*., vol. 62, pp. 479-499, (2005). DOI:10.1016/j.apradiso.2004.07.005.

[3] M. Oddone, L Giordani, F. Giacobbo, M. Mariani, S. Morandi, ”Practical considerations regarding high resolution gamma-spectrometry measurements of naturally occurring radioactive samples”. *J. Radioanal. Nucl. Chem*., vol. 277, pp. 579-585 (2008). DOI: 10.1007/s10967-007-7113-3

[4] M.B. Nisti, A.J.G. Santos, B.R.S. Pecequilo, M.F. Máduar, M.M. Alencar, S.R.D. Moreira, “Fast methodology for time counting optimization in gamma-ray spectrometry based on preset minimum detectable amounts”, *J. Radioanal. Nucl. Chem*, vol. 281**,** pp. 283-286 (2009). DOI: 10.1007/s10967-009-0102-y.

[5] R. Shweikani and M. Hasan, “Determination of the optimal measurement counting time and detection limit for gamma-ray spectrometry analysis”, *Accred. Qual. Assur.*, vol. 20, pp. 501–504 (2015). DOI: 10.1007/s00769-015-1169-5

[6] G. S. C. Joel, S. Penabei, N. M. Maurice, C. Gregoire, N. M. E. Jilbert, T. S. Didier, V. Werner, S. David, 2017. “Optimal measurement counting time and statistics in gamma spectrometry analysis: The time balance”, *Published by the American Institute of Physics* (2017). DOI: 10.1063/1.4969040.

[7] N. Naskar, S. Lahiri P. Chaudhuri, A. Srivastav, “Measurement of naturally occurring radioactive material, ^{238}U and ^{232}Th: part 2—optimization of counting time”, *J. Radioanal. Nucl. Chem*., vol.312, pp.161–171 (2017). DOI 10.1007/s10967-017-5205-2.

[8] M.B. Nisti, M.F. Máduar, C.H.R. Saueia, F. Cavalcante, B.P. Mazzilli, 2020. “Optimization of the gamma-ray spectrometry counting time based on uncertainties of radionuclides concentration in samples”, *Braz. J. Radiat. Sci*., vol. 08-02, pp. 01-12 (2020). ISSN: 2319-0612.