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Optimal Measurement Counting Time and Statistics in Gamma Spectrometry Analysis: The Time Balance

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Abstract. The optimal measurement counting time for gamma-ray spectrometry analysis using HPGe detectors was determined in our laboratory by comparing twelve hours measurement counting time at day and twelve hours measurement counting time at night. The day spectrum does not fully cover the night spectrum for the same sample. It is observed that the perturbation come to the sun-light. After several investigations became clearer: to remove all effects of radiation from outside (earth, the sun, and universe) our system, it is necessary to measure the background for 24, 48 or 72 hours. In the same way, the samples have to be measured for 24, 48 or 72 hours to be safe to be purified the measurement (equality of day and night measurement). It is also possible to not use the background of the winter in summer. Depend on to the energy of radionuclide we seek, it is clear that the most important steps of a gamma spectrometry measurement are the preparation of the sample and the calibration of the detector.

1. INTRODUCTION

Radioactive decay occurs randomly in time, so the measurement of the number of events detected in a given time period is never exact but represents an average value with some uncertainty reliable to the sample, the detector, the geometry of the experiment and the technics used to analyzed sample [1]. Better average values can be obtained by acquiring data over longer time periods. But, since this is not always possible, it is necessary to be able to estimate the accuracy of any given average in accordance with the nuclear methods used.

Nuclear counting statistics at high count rate are assessed on a γ-ray spectrometer set-up. Our typical gamma spectrometry system consists of a High Purity Germanium (HPGe) detector, liquid nitrogen cooling system, preamplifier, detector bias supply, linear amplifier, analog-to-digital converter (ADC), multichannel storage of the spectrum, and data readout devices. Although the system is powerful enough for background measurements, it is important, nowadays, to have a great statistical in short time measurement: which is a challenge for scientists.

Knowing the optimal measurement counting times is practical when determining detection limits for long-lived radionuclides, such as to satisfy laboratory accreditation technical requirements. For the analysis of environmental samples with low radioactivity, a relatively long counting time is required, e.g., up to 1–2 days, to obtain accurate and precise results [1-3]. Therefore, to decrease the cost of gamma-ray spectrometric analysis, the various

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measurement parameters should be optimally balanced. For example, increasing sample mass, extending counting time, and improving sample geometry will increase the net count N, the best parameter in experimental gamma spectrometry measurement. Another important factor is efficient shielding as it lowers the spectral background noise. However, any of these solutions have its limitations and overextending a given parameter provides no additional benefit. Better average values can be obtained by acquiring data over longer time periods. But, since this is not always possible, it is necessary to be able to estimate the accuracy of any given average [4].

The purpose of this study was to determine the average time for gamma spectrometry measurement. This idea came to us by comparing the spectra of measuring radioactivity lasts for 12 hours in the day that does not fully covered the twelve hours night spectra for the same sample with 24 hours background measurement. In this study, "a priori" counting times as a function of the preset (MDA) are proposed for routine use. Initially, sample counting time was determined. This time is related to sample composition, radionuclide being analyzed, background radiation, the geometry of detection flask and detection system (detector, shielding, and associated electronics). Time counting optimization is also described, as an additional step in routine gamma-ray spectrometric measurements and automated spectral analysis which are used to control and assess the quality of the measurement and analysis [5-8].

Time counting optimization in gamma-ray spectrometry was established using HPGe detectors and counting geometries. Some samples were prepared and measured for fourteen different counting times, ranging from 300.00 to 259,200.00 s. The obtained results were compared to the reference values from solutions with well-known activity concentrations samples, for minimum detectable amount (MDA) calculation [6, 8]. A counting time of 86,400 s was found to be generally sufficient to reach the agreement between the preset and actual counting times.

2. MATERIAL AND METHOD

Our challenge is to obtain the "best" spectroscopy data possible in every measurement situation. "Best" is a combination of statistical (number of counts) and spectral quality (peak, width, and position) over a wide range of counting rates [7].

The optimization of the throughput of a laboratory, when the sample counting geometries and sample measurement times are related, is deeply important. The aim of the present work is to determine the counting times for a sequence of measurements required to meet the given minimum detectable activity and to express the counting times in terms of the spectrometer properties and counting physical geometry.

As the gamma emission is a random process and due to the interaction of gamma rays with the material of the detector, the efficiency of gamma spectrometry is not absolute, so the aim of the efficiency calibration of the system is the determination of a factor corresponding to each gamma line that normalizes their activity concentration to its absolute value. Typically, a standard or reference mixed gamma source with multiple energy transitions, in a similar geometry to the measured samples, should be used to produce an absolute efficiency calibration curve along with the necessary attenuation corrections [9, 10]. These reference materials should be traceable to a well-known international reference material manufacturing by organization like NIST and IAEA. However, occasionally these kinds of sources might not be available for every laboratory [3]. As a result, an alternative calibration procedure can be employed, namely relative efficiency calibration.

Our typical gamma spectrometry system consists of a Germanium (Ge) detector, liquid nitrogen cooling system, preamplifier, detector bias supply, linear amplifier, the analogue-to-digital converter (ADC), multichannel storage of the spectrum, and data readout devices. The detector is often housed within a shield to reduce the background caused by sources other than the sample. The shield is constructed of a dense material (such as lead) that will absorb a large portion of background gamma rays. The shielding is usually crafted in such a way as to minimize backscattering. The lead shielding material is usually graded with a two-part thin metal shield such as tin and copper to reduce the effects of x-rays generated by the interaction of ambient photons with the lead. The sample is positioned within the shield at some distance from the detector [11, 12]. The distance will depend on a number of parameters, such as expected count rate and geometry of the sample container. The measurements take many aspects and are explain in the following paragraph.

Firstly, 12 hours background measurement at day was compared to 12 hours night background; the spectra were not fully covered and this remark may give some hypothesis. 12 hours background measurement is subtracted to twelve hours sample measurement, all at daylight and the same experiment was done during 12 hours at night. The spectra are not fully covered. The decision to substrate 48 hours background is applied to all other measurement.

Using this hypothetical system, photons emitted from the sample interact with the Ge crystal to produce a pulse. The amplitude (or height) of the pulse is proportional to the energy of the photon absorbed by the Ge. Each pulse is amplified (or magnified), shaped, and sorted according to pulse height to produce a histogram (counts per unit energy) of the incident photons. This histogram is called a spectrum. As the counts accumulate, peaks develop that can be identified by energy and thus the nuclide identities of the spectrum are also identified assuming that the system has been calibrated. In general, the goal of the gamma spectrometry is to derive nuclide specific gamma emission rates of the sample (in activity units, such as Becquerel [Bq] or decays per second) from the spectral data.

Despite the apparent simplicity with which gamma-ray measurements are made (little sample preparation is required), there are a number of correction factors to the raw counting data that must be considered:

- The loss of pulses due to pulse pile-up (at high count rates).
- Coincidence summing (both random and cascading).
- The decay of the source during counting.
- The decay of the source from some previous reference time.
- Attenuation of photons as a result of interactions with the sample.
- Emission rate (or yield) of the specific photon energy.

Activities and uncertainties calculations were made using the formula proposed by W. F. Bakr and Y. Y. Ebaid, 2011 and R. Shweikani M. and Hasan, 2015 [9, 13]. MDA calculation is based on the equation proposed by M. B. Nisti et al., 2009 [8]. The gamma-ray spectrometric measurements were carried out using a hyper-pure germanium detector of planner configuration CANBERRA model BEGE6530, with relative efficiency 60% and resolution 1.95 keV (FWHM) at 1.33 MeV Co-60 gamma line. The gamma energy transitions of 295.2 keV (19.4%) and 351.9 keV (37.1%) of ^{214}Pb and 609.3 keV (46.1%) and 1120.0 keV (15.0%) of ^{214}Bi was used for ^{226}Ra determination [14].

3. RESULT AND DISCUSSION

Detection limits for a peak are typically defined by setting a threshold that the counts in the peak must exceed to claim detection. In general, the threshold is chosen so that there is a 95% probability of the counts exceeding that threshold if the peak is truly present in the spectrum. That defines the 95% confidence limit. This choice implies that there is a 5% probability of not detecting the peak, even though it is considered to be 1% in this study and the confidence limit is defined to be 99% [15, 16].

"TABLE 1" and "**FIGURE 1**" present the percent contribution of the relative standard uncertainty of each element to the total uncertainty of the activity calculation. To evaluate the effect of counting statistics on the total uncertainty, different IAEA reference samples with different activity concentration contents of ^{226}Ra were counted and the percent contribution of the net counts to the activity standard uncertainty was represented. The activity concentration of ^{226}Ra in the selected samples is illustrated in "**TABLE 1**". "**FIGURE 1**" illustrates the percent contribution of relative uncertainty of net count to the activity uncertainty of each set time measurement.

"TABLE 1" summarizing the results from a series of fourteen consecutive time measurements of some natural radionuclides under the influence of the time measurement and counting statistic. First, on the table are sampling time measurement beginning at five minutes up to 72 hours. Next are the results of activities concentration of ^{212}Pb with an error related to the experimental measurement. It is the only radionuclide detected with five minutes time measurement, but the related error is higher than 20%. In this case, this value cannot be considered when determining radioactivity in gamma spectrometry. After two hours measurement, the result shows is more reliable to the reality with an error lesser than 3%. Next two columns are activity concentration related to ^{212}Bi and ^{40}K. They are both detected after ten minutes measurement but the relative error related to the obtained values are 27 and 33 in term of percentage, respectively. After 24 hours measurement, the obtained values, and relative errors make sense for these two radionuclides.

Next two columns are ^{214}Pb and ^{228}Ac. To detect these two radionuclides in the laboratory, we need a minimum of thirty minute time measurement, but the result shows 15.19±17% and 52.15±11%. The measurement needs time to obtain a good result, in this case, is two hours. After these two radionuclides, the attention is showed to ^{232}Th and ^{214}Bi which are first time detected after 45 minutes measurement. After twelve hours measurement, the results show a relative error of 4% <5% (acceptable rates of the mean activity value) for both radionuclides.

The last two columns show the results of ^{235}U and ^{226}Ra. These two radionuclides need a minimum of 3 hours to be detected at the first time. To improve the results obtained after 3 hours measurement, it is recommended to count during 12 and 24 hours.

TABLE 1: Specific activity related to different commonly used radionuclides in Gamma Spectrometry. Measurement for different time (Called Sample Time or ST*) ranged from 300 to 259,200s.

ST*	Radionuclides detected Specific activity (Bq/Unit)								
	^{212}Pb	^{212}Bi	^{40}K	^{214}Pb	^{228}Ac	^{232}Th	^{214}Bi	^{235}U	^{226}Ra
5min	34.61±6.7	ND	ND	ND	ND	ND	ND	ND	ND
10min	39.61±5.94	53.34±14.6	151.35±49.6	ND	ND	ND	ND	ND	ND
30min	49.20±3.61	51.06±13.6	209.52±32.69	15.19±2.61	52.15±5.50	ND	ND	ND	ND
45min	42.96±2.31	50.41±11.8	202.11±26.72	13.85±2.98	55.90±4.92	7.78±7.50	24.12±3.04	ND	ND
1h	39.01±2.23	46.0±14.25	170.16±21.15	10.66±1.42	51.27±3.67	37.29±4.55	15.82±2.37	ND	ND
2h	53.85±1.45	76.95±12.63	176.89±16.08	16.70±1.41	57.21±2.66	38.92±3.69	14.63±1.90	ND	ND
3h	51.32±1.17	53.38±11.05	179.20±13.40	16.53±0.96	58.46±2.18	43.19±3.38	20.18±1.58	0.935±0.98	14.90±7.24
4h	51.28±1.05	62.97±10.04	184.37±12.56	19.22±0.88	61.47±2.14	46.00±3.20	18.06±1.38	1.11±2.30	21.22±37.75
6h	52.61±0.88	80.67±9.68	223.09±10.85	16.41±0.67	51.86±1.66	43.62±2.70	17.58±1.06	0.97±0.85	15.43±6.73
12h	50.95±0.61	62.99±5.83	182.83±7.25	14.35±0.43	47.70±0.77	37.75±1.86	18.38±0.77	1.38±0.36	21.93±4.20
24h	50.03±0.52	61.00±3.14	190.70±6.20	16.40±0.35	52.85±0.69	38.17±1.75	17.49±0.66	1.41±0.31	22.11±2.41
36h	49.95±0.42	59.44±2.22	189.99±4.82	15.92±0.25	48.91±0.54	39.57±1.56	16.91±0.40	1.55±0.19	20.76±1.54
48h	49.58±0.39	61.03±2.01	191.18±4.51	16.22±0.23	57.46±0.52	41.20±1.48	18.84±0.39	1.54±0.14	20.96±1.18
72h	49.92±0.33	60.80±1.96	190.79±4.42	16.04±0.19	51.25±0.47	40.51±1.40	18.05±0.36	1.53±0.12	21.03±1.09

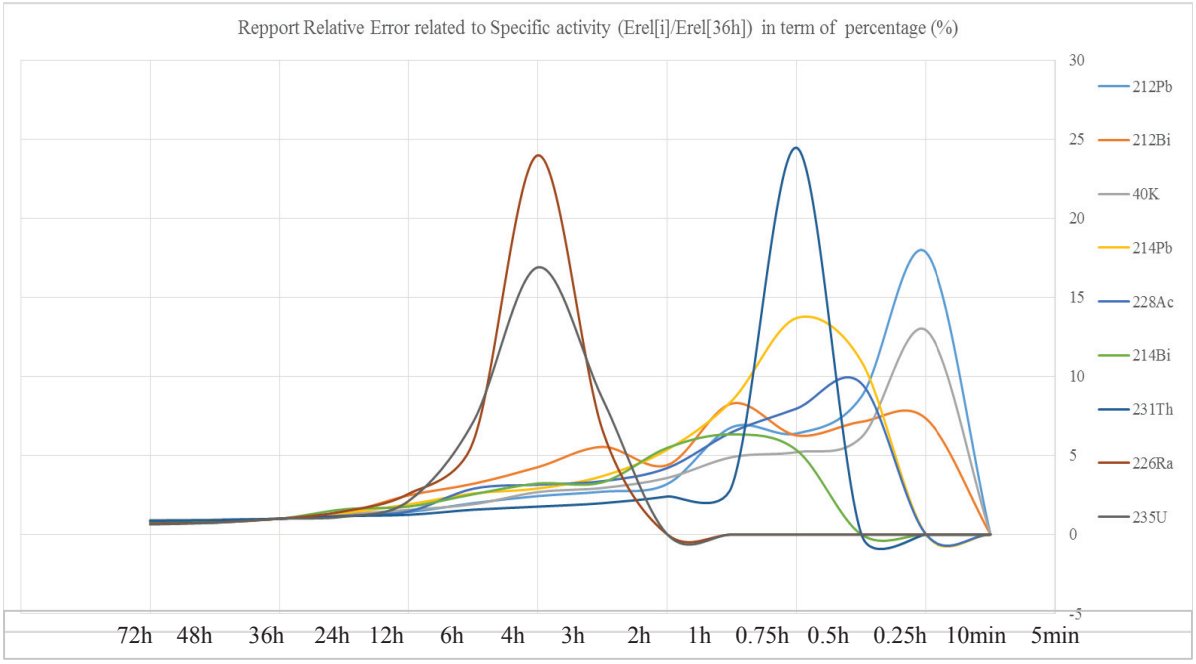

FIGURE 1. Report of Relative Error related to Specific activity (Erel[i]/Erel[36h]) in term of percentage (%) estimated in nine radionuclides with fourteen set count times.

Clearly, it is important to maximize the peak-to-background ratio, the net counting rate in the peak, and the counting time to achieve the lowest detection limits. The estimation of results uncertainty is the most important issue in analytical techniques in gamma spectrometry especially in the determination of radioactivity in

environmental samples in which their activity concentration is mainly low: the radionuclides analysed in this work [4, 6, 11, and 17].

It is observed that daylight spectrum and night spectrum are not fully covered each other: This is certainly due to the perturbation of the Sunlight, which are slowly varied between the day and night but not significantly. In Astrophysics, some researchers proved that this variation is negligible, but it is possible, according to this study, to observe the little contribution of Sunlight to background measurement in gamma spectrometry [20, 21].

CONCLUSION

To detect Uranium, Thorium and their respective daughters and Potassium series with a relative related error less than 1%, it was found that it is necessary to count during a minimum of 24 Hours (86,400 s). This result is in accordance with the literature with planar geometry detector. These results conduct us to make the following three guidelines for selecting the detector best suited for an application:

1. The more detector material available (germanium semiconductor), the higher the full-energy peak efficiency.
2. The smaller the distance between the detector and the source material, the higher the full-energy peak efficiency. The efficiency of the detector inversely increase with the distance between sample and detector.
3. While better resolution gives a better MDA, the resolution contributes only as the square root of the MDA value, whereas the MDA is proportional to the full-energy peak efficiency [11, 16, 18, and 19].

It could be shown that variable counting losses of more than 95% may be corrected in real time by means of the virtual pulse generator principle and that the corrections are complete, without bias and uniquely determined by counting statistics by enhancing counting time in gamma spectrometry.

The proposed methodology has the goal of optimizing the throughput of a laboratory when processing large batches of samples. The actual counting times obtained were in good agreement with the preset times for all the studied detection systems and counting geometries, suggesting that this methodology could be applied to a wide range of detection counting systems and sample geometries. Furthermore, a counting time of 50,000 s was found to be generally sufficient to reach the agreement between the preset and actual counting times. Some care could be needed in applying these results when the composition and density of samples are significantly different from those used in the study reported here [19].

The conclusion after several investigations became clearer: to remove all effects of radiation from outside (earth, sun and universe) our system, it is necessary to measure the background for 24, 48 or 72 hours. In the same way, the samples have to be measures for 24, 48 or 72 hours to be safe to purify the measurement (equality of day and night measurement). It is also possible to not use the background of the winter in summer. Depend to the energy of radionuclide we seek, it is clear that the most important steps of a gamma spectrometry measurement are the preparation of the sample and the calibration of the detector [20, 21].

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