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PROPOSAL OF A METHODOLOGY TO MEASURE NEUTRON ACTIVATION USING GAMMA-GAMMA COINCIDENCE SPECTROSCOPY

Guilherme S. Zahn[1], Frederico A. Genezini[2] and Cibele B. Zamboni[1]

[1] Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
gzahn@ipen.br
czamboni@ipen.br

[2] Centro Regional de Ciências Nucleares (CRCN / CNEN - PE)
Av. Professor Luiz Freire, 1
50740-540 Recife, PE
fredzini@cnen.gov.br

ABSTRACT

Neutron Activation Analysis is a very useful analytical technique that allows the quantification of several elements at once with small amounts of sample material. On the other hand, the results obtained are strongly dependent on the fit of the gamma-ray peaks, and under certain conditions this can be influenced by factors like the Compton continuum spectrum, x-rays from the shielding and background radiation. Even if the use of well-designed multilayer shields greatly reduces the last two factors, it may increase the first as Compton scattering from the shielding may also hit the detector, resulting in problems, especially on the low-energy range of the spectra ($E\gamma < 200$keV).

In this work the viability of the use of a simple gamma-gamma coincidence assembly to virtually eliminate the background interference without increasing the secondary effects is assessed. The results show that even this simple system greatly reduces the background radiation contribution, and that the secondary effects remain roughly constant; on the other hand, the detection efficiency is greatly reduced and the determination of the absolute detection efficiency gets overcomplicated and somehow unreliable. As a test for this system, the comparative quantification of a gamma-ray source with very low activity is performed, and the results are compatible with the expected values.

1. INTRODUCTION

Neutron Activation Analysis (NAA) is a very well established and widely used analytical technique to measure trace elements in several different types of samples [1,2]. Among the advantages of this technique is the fact that it allows the determination of concentrations of multiple elements at once using small amounts of sample material. Roughly speaking, the NAA technique can be divided in two lines: *instrumental* (or *comparative*) NAA, where the sample is analyzed together with a standard reference material and the concentrations are determined comparatively; and *absolute* (or *parametric*), where the sample is analyzed alone and the concentrations are determined from the absolute gamma-ray intensities. Both lines are strongly dependent on the fit of the gamma ray peaks, which can be influenced by both secondary effects originated in the detection system (as the Compton continuum, x-rays and

the 511keV annihilation peak) and gamma-ray peaks from the background. Also, the second line (*absolute* NAA) is strongly dependent on a good knowledge of the detection efficiency of the system – this is not true for the *instrumental* NAA, though, as the quantification is made comparatively.

The most usual way to reduce the background radiation contribution consists in arranging the detector and sample inside a Lead shielding; this, on the other hand, results in a great increase of some secondary effects, mainly with the presence of the Lead x-rays (72keV<E<85keV [3]), but also increasing the Compton background as radiation from the sample may scatter in the shielding and get back to the detector. In order to decrease the first problem, multilayer shields are available where lighter elements are placed inside the Lead shielding in order to absorb the Lead x-rays, emitting lower-energy x-rays that fall below the effective energy range of the Ge detector used, but even these expensive shields may increase the Compton continuum and the 511keV annihilation peak.

An alternative way to reduce the background, without increasing the secondary effects, is the use of a gamma-gamma coincidence system. In this type of assembly, the source is placed in front of two Ge detectors, and an event is counted only if "seen" by both detectors simultaneously (i.e., in a small time interval Δt). As background radiation comes from all sides while the sample radiation comes from a point close to both detectors, the probability that two gamma rays from a background event will be recorded is much lower than the probability that two gamma rays from a sample event will do it, so the ratio between the background counts and the sample counts should decrease accordingly.

2. EXPERIMENTAL PROCEDURE

For this work, two Ge detectors with active volumes of $89cm^3$ and $76cm^3$ were placed making a 90^o angle; collimators were fitted to both detectors in order to avoid Compton interference from one detector on the other, and the samples were placed in the crossing of both detectors' symmetry axis, at a distance of 3cm from the face of each (v. Fig 1a). The output from each detector went into a spectroscopy amplifier; from there, the signal from the $76cm^3$ detector went into a Single Channel Analyzer (SCA) with the energy window fully open, then to a Pulse Stretcher and into the gate input of a Multichannel Analyzer (MCA); the signal from the $89cm^3$ detector went into a delay module and then to the linear input of the MCA (v. Fig 1b) - the delay time was adjusted using an oscilloscope and a delay of 4μs was chosen. This very simple coincidence system doesn't discriminate the coincidences according to the energy recorded in the $76cm^3$ detector and has no account for accidental coincidences.

Standard sources of ^{152}Eu, ^{133}Ba and ^{60}Co were used to calibrate both detectors in energy and efficiency (in *single* – i.e, non-gated – mode); Table 1 shows the calculated activity of these samples. Acquisition in coincidence mode was also made using these same three sources. All the peak areas were evaluated using the IDeFix software [4], which models the peak as a Gaussian peak with corrections as an exponential tail and a step function.

The analysis of the coincidence acquisition system's performance was made focusing in the following parameters:

- ***Secondary effects:*** In the fits of the gamma peaks, the ratio between the background continuum and the peak area for the singles and coincidence acquisition was used to analyze the Compton effect contribution;
- ***Background radiation:*** The singles and coincidence count rates of background peaks were compared to the count rates of peaks from the standard sources close to them in energy, in order to analyze the efficiency of the coincidence method in reducing the background.
- ***Detection efficiency:*** The detection efficiency of the coincidence system was calculated for each gamma peak from the standard sources and compared to the detection efficiency of the 89cm^3 detector;

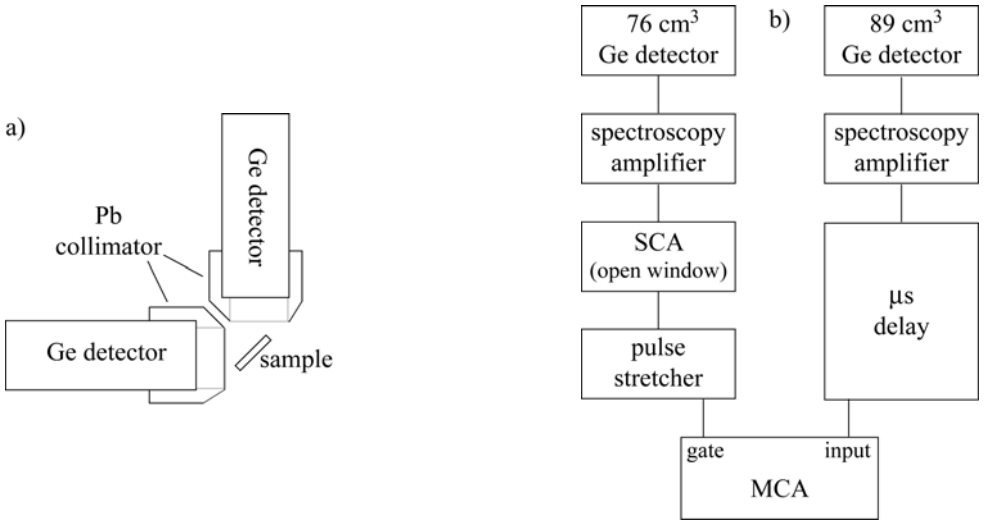


Figure 1. Experimental setup used in this work;
a) the detector assembly;
b) scheme of the electronics setup.

Table 1. Radioactive Sources Used in the Calibration

Source	Activity (kBq)
^{152}Eu	72.6 (13)
^{133}Ba	19.9 (26)
^{60}Co	55.9 (8)

Finally, two tests were performed in order to check for the method's usability in comparative activity analyses; first, a second ^{60}Co source was analyzed using both the 89cm^3 detector in singles mode and with the two detector in coincidence mode and its activity was calculated in both methods by comparing the peak areas with the peak areas obtained for the ^{60}Co source with known activity; also, another ^{133}Ba source with very low activity was analyzed using the coincidence system, and its activity was calculated comparatively using the known activity of

the first ^{133}Ba source and the weighted average of the ratios between the areas of the individual peaks.

3. RESULTS AND DISCUSSION

The individual efficiency calibration curves for the two detectors used in this work are shown in Fig. 2.

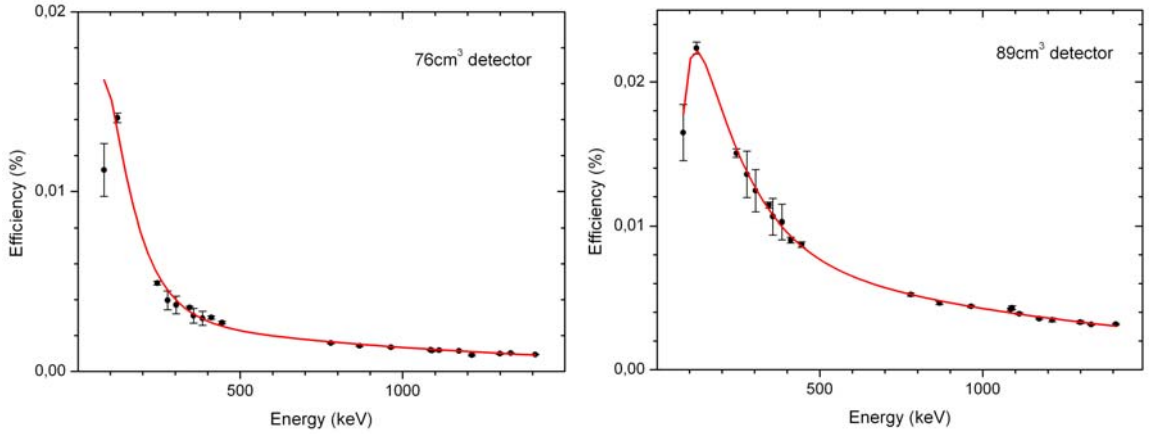


Figure 2. Efficiency calibration curve for the two detectors used in this work.

2.1. Compton Background

When fitting the gamma-ray peaks, the continuum background that appears even after the background spectrum has been subtracted from the "real" spectrum is a consequence of many secondary effects, but mainly of the Compton scattering, either inside the detector itself (so that the gamma ray deposits less than its total energy in the detector and the event is recorded with a "visible energy" that's less than the real energy) or in the surrounding materials – as Lead shields, other detectors and so on (and, once again, the recorded energy is less than the real energy of the original gamma-ray). In order to quantify this "Compton background", we define a "signal to noise ratio" for each peak fitted in both the singles spectrum from the 89cm^3 detector (S/N_S) and the coincidence spectrum (S/N_C) as the ratio of the peak area (A_X) and the integrated background under the peak:

$$(S/N)_X = A_X / (2 \cdot FWHM_X \cdot \overline{B}_X). \quad (1)$$

where $FWHM_X$ is the peak's full width half maximum and B_X is the fitted average background. Figure 3 shows the ratio of the coincidence and singles "signal to noise ratios" as a function of the energy; this is clearly constant, and the weighted average of 1.2(3) shows

that there's no noticeable difference in the continuum background between the singles and coincidence systems.

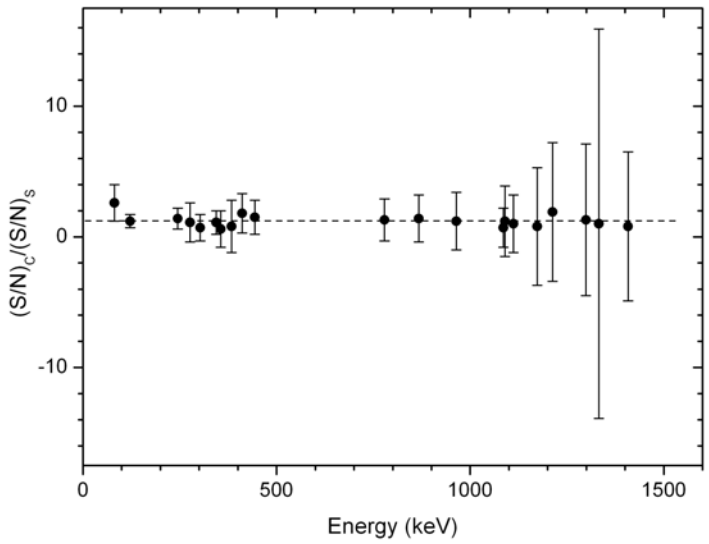


Figure 3. Ratio of the coincidence $(S/N)_C$ and singles $(S/N)_S$ *signal to noise ratio*.

2.2. Background Radiation

The usual way to subtract the background radiation in a singles acquisition consists in acquiring a "sourceless" spectrum and subtracting it from the spectrum taken with a radioactive source. This process is usually efficient, but may be troublesome when the peaks of interest are small and close in energy to strong background peaks as the 1460keV peak from ^{40}K or the 351keV peak from ^{214}Pb, the fit becomes complicated and frequently the area of the interest peak is "masked" by the statistical fluctuations in the much larger area being subtracted; other frequent problem is that even small variations in the gain of the acquisition system will make this subtraction quite problematic. Table 2 shows the comparison of four background peaks (from naturally-occurring isotopes as ^{40}K, ^{208}Tl, from the ^{232}Th decay chain, and ^{214}Pb and ^{214}Bi, both from the ^{238}U decay chain), all fitted together with peaks from the calibration sources in order to allow a comparison, for both the singles spectrum from the 89cm^3 detector (without background subtraction) and the coincidence system; these results show that the ratio between the area of the interest peak and the area of the background peak was better in the coincidence system for all the peaks, with the enhancement varying roughly between less than 2% and more than 400%, depending on the case.

Table 2. Comparison of the counts per second (cps) for four background peaks to the cps of peaks from the decay of ^{152}Eu for both the singles (S) and coincidence (C) systems.

Transitions (keV)	cps(Eu)/cps(BG) *singles*	cps(Eu)/cps(BG) *coincidence*	Difference (%)
1460 (^{40}K) 1408 (^{152}Eu)	33.3 (12)	33.9 (21)	1.87 (13)
583 (^{208}Tl) 586 (^{152}Eu)	1.37 (13)	3.2 (5)	135 (24)
609 (^{214}Bi) 586 (^{152}Eu)	1.80 (19)	10 (4)	440 (170)
351 (^{214}Pb) 344 (^{152}Eu)	65.1 (26)	88 (7)	35 (3)

2.3. Detection Efficiency

Figure 4 shows the relative detection efficiency of the coincidence system (i.e., the detection efficiency of the coincidence system divided by the detection efficiency of the 89cm^3 detector in single mode); these results show that the detection efficiency for a system as the one tested in this work isn't a simple function of the energy of the gamma-ray, as it also depends on the decay scheme of the nucleus being studied and on the response function of the other detector. Moreover, the lower relative efficiency for the ^{133}Ba peaks seem to suggest that there may be a relation between this efficiency and the activity of the source, as the ^{133}Ba source was also the weakest of the ones used here; this could be due to the contribution of accidental coincidences, which should depend on the activity of the source. Therefore, the absolute determination of sample activities can't be performed at this point using this system.

2.4. Activity Determination

The results obtained with the second ^{60}Co source by both methods is shown in Table 3, and they show that both methods agree in the determination of the source activity, although the coincidence method gives a higher uncertainty, probably due to the lower counting statistics.

The low-activity ^{133}Ba source was a calibrated one, and according to the certificate, its present activity would be 42.8(3)Bq. The experimental determination of this activity was also performed using both methods, but in the singles spectroscopy no peak from the decay of ^{133}Ba could be identified, mainly due to the contamination of the 356keV peak (the most intense in this decay) by the 351keV background peak from ^{214}Pb. In the coincidence mode, though, the 356keV could be analyzed and the calculated activity in this method was 44(11)Bq, in perfect agreement with the calculated value; it should also be noted that the 25% uncertainty was a consequence both of a 21% uncertainty from the fit of the 356keV peak (137 counts in 140400s of acquisition) and the 13% uncertainty from the activity of the known ^{133}Ba source.

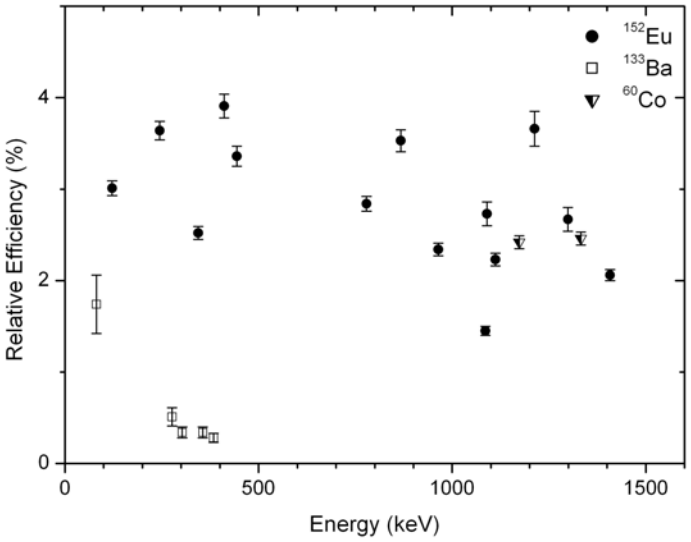


Figure 4. Detection efficiency of the coincidence system relative to the detection efficiency of the 89cm^3 detector in singles mode.

Table 3. Determination of the activity of the unknown ^{60}Co source.

Transition (keV)	Activity (kBq) *singles*	Activity (kBq) *coincidence*
1173	41.2 (7)	34.9 (14)
1332	41.8 (7)	34.5 (14)
Weighted Average	41.5 (7)	34.7 (11)

3. CONCLUSIONS

This work tested the performance of a simple coincidence system in the determination of radioactivity. The coincidence system proved to reduce the influence of the background radiation in gamma-ray analyses without any influence, either positive or negative, on the Compton continuum. The absolute detection efficiency of this coincidence system was shown to be very hard to determine, so that absolute analyses can't be performed; but tests with two different sources proved that the system is able to determine the comparative activity of two sources with the same isotope, so that it can be used for instrumental NAA, for instance; also, this work proved that the system can be useful to determine the activity of sources with low activity, due to the very low background. The next step in this work will be to do some accidental coincidence compensation in order to "clean up" even more the spectrum.

REFERENCES

1. L. C. Oliveira, C. B. Zamboni, F. A. Genezini, A. M. G. Figueiredo, G. S. Zahn, “Use of thermal neutrons to perform clinical analyses in blood and urine samples” *Journal of Radioanalytical and Nuclear Chemistry*, **263**, pp. 783-786 (2005).
2. J. C. Wasserman, A. M. G. Figueiredo, F. Pellegatti, E. V. Silva Filho, “Elemental composition of sediment cores from a mangrove environment using neutron activation analysis” *Journal of Geochemical Exploration*, **72**, pp. 129-146 (2001).
3. A. C. Thompson, D. Vaughan, *X-Ray Data Booklet*, Lawrence Berkeley National Laboratory, Berkeley, USA (2001).
4. P. Gouffon, *Manual do Programa IdeFix*, Instituto de Física da USP, São Paulo, Brazil (1987).