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COMPLEX GAMMA RAY SPECTRA ANALYSIS*

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Abstract. The gamma ray spectrometry laboratory of the University of Bucharest was designed for the analysis of low-level radioactivity samples. High efficiency shielded HPGe detectors are used. Efficiency calibration is achieved by complementing experimental calibration with Monte Carlo computation carried out using GESPECOR, also applied for the evaluation of coincidence summing corrections. Low detection and decision limits are challenging, and the method to respond to this situation is presented. Using a blank sample in the same experimental conditions as the actual sample turns out benefic for precise peak subtraction. Procedures for peak finding were developed, which include analysis of the peak area/FWHM ratios, hypergeometric alert, in order to establish confidence limits concerning the actual existence of a certain peak in the spectrum. The method is oriented to numeric solutions rather than developing complex fitting functions that might not be appropriate. Development of a software insuring retrospective predictability and asking for human intervention in controversial situations is in progress.

Key words: HPGe detectors, coincidence summing effects, low activity sample analysis

1. INTRODUCTION

Gamma-ray spectrometry with germanium detectors [1, 2] is a well-established analysis tool applied in many fields of investigation. The task, currently met in many gamma-ray spectrometry laboratories, of achieving the lowest detection limit in the shortest measurement time, supported by the technological progress in manufacturing big high purity Ge crystals, resulted in a widespread use of high efficiency Ge detectors.

In this work we present the characteristics of two high efficiency HPGe based gamma-spectrometry systems recently installed in the Atomic and Nuclear Physics Department of the Faculty of Physics. Tests of the systems are presented, the

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magnitude of the coincidence summing effects is illustrated, the method for removing these effects is demonstrated, and low background spectra analysis is presented.

2. THE DETECTION SYSTEMS AND THE ASSOCIATED PARAMETERS

Two high-efficiency HPGe detectors have been recently acquired by the Department of Atomic and Nuclear Physics of the Faculty of Physics - University of Bucharest. They are meant to serve for high precision and low background measurements of different kinds of samples among which there are environmental samples, geological samples and various samples measured in view of specific purposes.

The first detector is a p-type HPGe detector, model GEM60P4-83, manufactured by ORTEC, with a relative efficiency of 70% for the photons with energy 1332 keV emitted by ^{60}Co. The dimensions of the crystal are: diameter 6.98 cm, length 8.07 cm. The Ge dead layer has a thickness equal to 0.7 mm. The distance from end cap to detector is 4 mm. The end cap is made from Al, with a thickness of 1 mm.

The nominal parameters of the detector are: Resolution (FWHM) at 1.33 MeV: 1.86 keV; Peak to Compton Ratio (^{60}Co): 76:1; Peak Shape (FWTM / FWHM) at 1.33 MeV: 1.9; Peak Shape (FWFM/FWHM) at 1.33 MeV: 3.7; Resolution (FWHM) at 0.122 MeV: 0.87. In the above notations FWTM, FWFM and FWHM represent the full width at 1/10, 1/5 and 1/2 respectively, from the height of the peak (background corrected). The procedures that should be applied for the measurement of these parameters are detailed in the appropriate standard [3].

The second detector is an n-type HPGe detector, model MGX45P4-83, also manufactured by ORTEC, with a relative efficiency of 47% at the energy of 1332 keV (^{60}Co). The crystal has the dimensions: diameter 61.5 mm, length 73.4 mm. The crystal has rounded edges with a nominal radius of curvature equal to 8 mm. The inner hole has a diameter of 9.3 mm and a depth of 65.1 mm, ending with a spherical shape of 5 mm radius. The dead layer on the entrance face due to boron implantation has a thickness of 0.3 microns; the inner contact is obtained by Li diffusion and has a thickness of 0.7 mm. The detector cup is made from 0.03 mm Al + 0.03 mm mylar on the entrance face, 0.8 mm Al on the sides and 3 mm Al on the back of the crystal. The entrance window is made from Be, with the thickness of 0.5 mm. The Be window can be protected by a standard plastic cap. The side part of the end cap is made from Al, with the thickness of 1 mm. The distance from the crystal to the entrance window is equal to 4 mm.

The parameters of the detector, as specified in the manufacturer's sheets, are: Resolution (FWHM) at 1.33 MeV: 2.00 keV; Peak to Compton Ratio (^{60}Co): 63:1; Peak Shape (FWTM / FWHM) at 1.33 MeV: 1.9; Peak Shape (FWFM/FWHM) at 1.33 MeV: 2.8; Resolution (FWHM) at 5.9 keV (^{55}Fe) 640 eV.

In both systems the technique of digital signal processing is applied with DSPEC-Plus modules, connected to PC. Spectrum acquisition, display and analysis is controlled by GammaVision 32 or Maestro 32 software.

3. BEHAVIOR AS A FUNCTION OF COUNTING RATE

The two systems are primarily intended for the measurement of low level sources. However, as they are the only HPGe detectors available in the Department of Atomic and Nuclear Physics, they will be of general use, including possibly the analysis of high activity sources. In view of this potential application, a study of the behavior of the systems as a function of count rate was carried out. This study was focused on the measurement of resolution, Peak-to Compton and count rate in the peak of 1332 keV for a fixed position ^{60}Co source measured alone or in the presence of sources of ^{133}Ba of increasing activity, placed near detector. It is important to mention that the spectrum of ^{133}Ba is confined to lower energies than the parts of the spectrum that are analyzed for resolution, Peak-to-Compton and count rate in the peaks of ^{60}Co.

DSPEC-Plus has two procedures to deal with high count rates. The first, based on the Gedcke-Hale method [4], is of general use and introduces a difference between the real time and the live time of measurement as a function of the dead time (implicitly, as a function of count rate). The method takes into account the fraction of time when the system is blocked by processing a pulse and extends the real time (the clock time) with the amount required to compensate for the interval when the system was not able to process pulses. The ratio of the number of counts to the live time (instead of the real time) gives the corrected count rate, taking into account most of the effects of the dead time. Another procedure available in DSPEC-Plus does not correct the acquisition time, but the count rate. This method is called Zero Dead Time (ZDT) method and is preferred when the counting rate has a strong variation during the measurement [5], because in this case the classical Gedcke-Hale method is not appropriate. In view of the fact that in our measurement the count rate was expected to be constant (^{133}Ba and ^{60}Co have a long half-life in comparison with the counting intervals), we selected the Gedcke-Hale method for dealing with dead time corrections.

In all the measurements the parameters of the amplifier and converter were kept constant, no attempt to optimize them in function of the count rate was undertaken. The acquisition time in each measurement was long enough in order to obtain a good statistical uncertainty of the quantities of interest.

All the measurements were done with the same cylindrical Pb shielding, of inner radius and thickness 5 cm. The compact shield decreases the contribution of radon distributed inside the shield to the background, but, due to scattering and photoelectric effect in Pb, does increase the total efficiency of the system.

A first measurement was done with a ^{60}Co point source placed at a distance of 5 mm from the end cap of the detectors. Then, keeping the ^{60}Co source in the same position, 5 sources of ^{133}Ba with increasing activity were successively placed on the end cap, gradually increasing the input count rate. In what follows the results obtained with the n-type detector will be presented, but the results obtained with the p-type detector are qualitatively similar.

In Fig. 1 the part of the spectrum including the 1173 and 1332 keV peaks of ^{60}Co obtained in the absence of the ^{133}Ba source is presented. The dead time is about 0.3%. In this case the resolution was 1.975 keV and the Peak-to-Compton ratio approximately 54. The resolution is in good agreement with the manufacturer's value, but the Peak-to-Compton ratio is worse. We consider that the contribution of the scattering in the compact shield surrounding the detector and the source increases the count rate in the Compton region, and also, increases the effect of coincidence losses from the 1 332 keV peak, being at least partially responsible for the lower Peak-to-Compton ratio in our measurements.

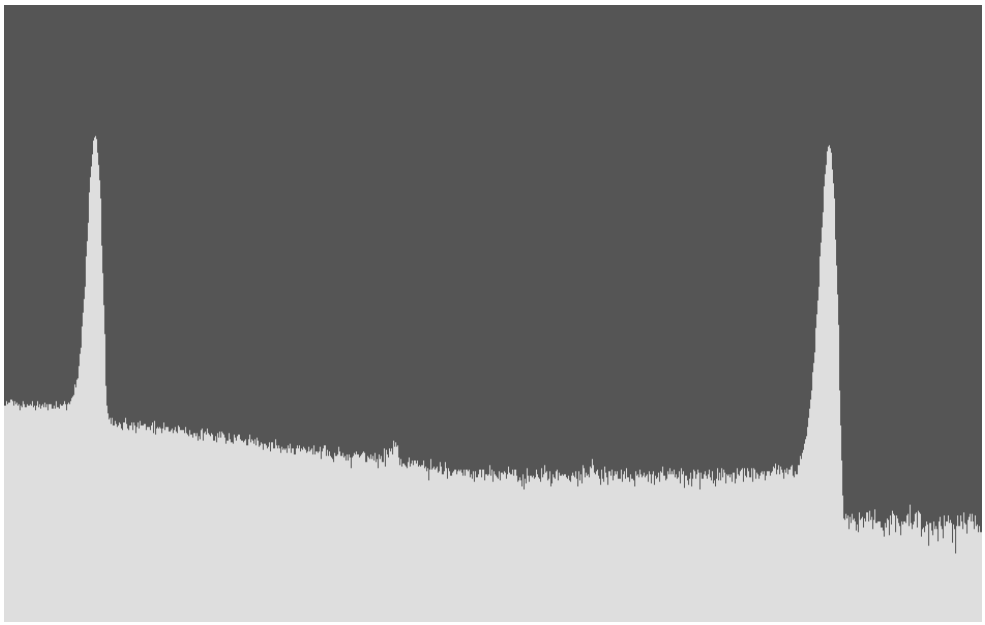

Fig. 1 – The high energy part of the ^{60}Co spectrum measured with the n-type detector in the absence of ^{133}Ba source. The dead time is about 0.3%.

When the ^{133}Ba sources were added, the dead time successively increased from 28% to more than 91%. In these conditions the spectra get worse, the resolution increased, the Peak-to-Compton ratio decreased and the count rate in the peaks of ^{60}Co decreased. The spectra displaying the same part of the ^{60}Co spectrum for dead time values equal to 47 and 91% are presented in Figs. 2 and 3. Besides

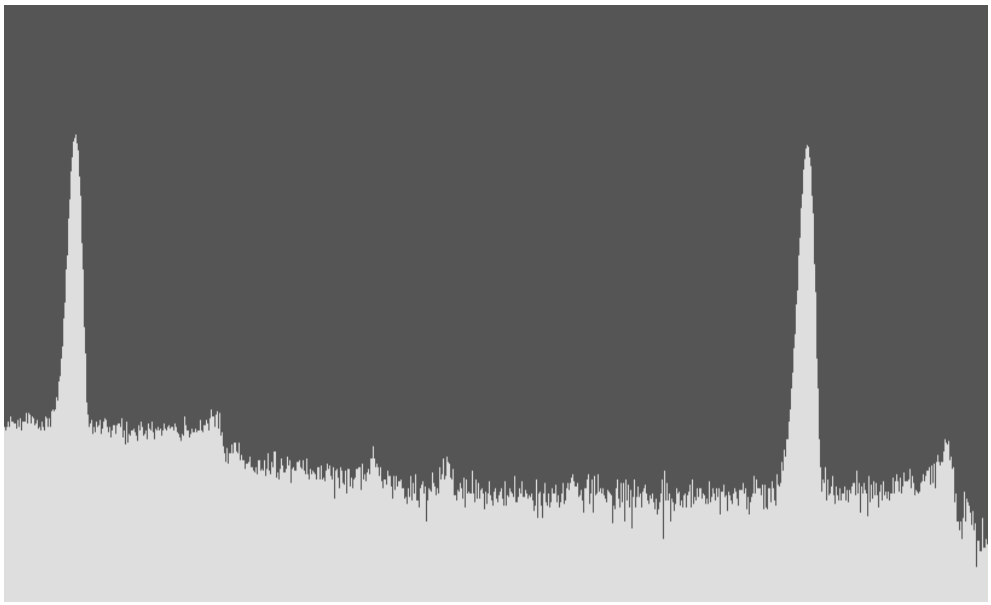

Fig. 2 – The same part of the spectrum of ^{60}Co as in Fig. 1 in the presence of a ^{133}Ba source producing a dead time of 47%.

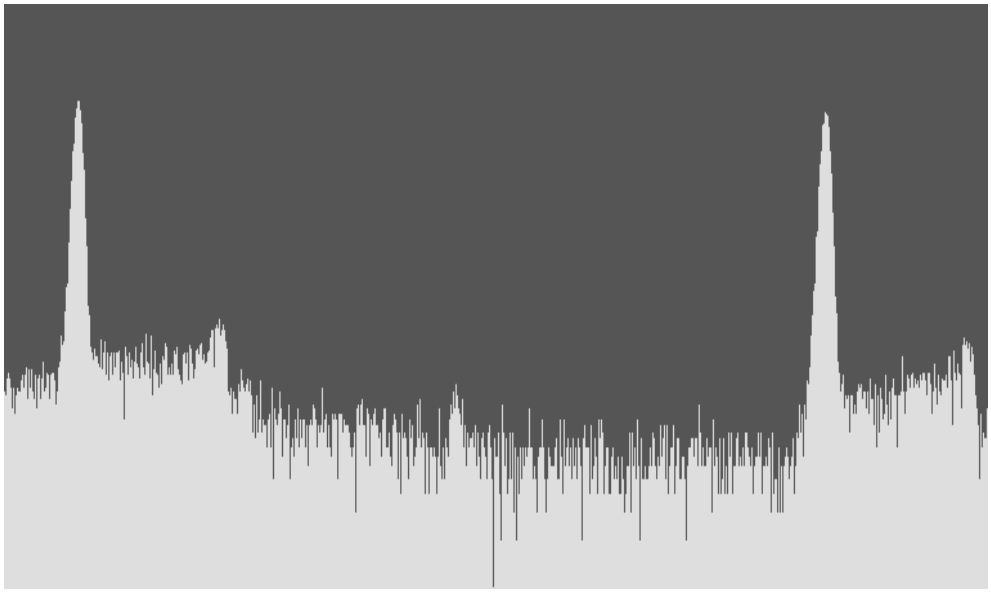

Fig. 3– The same part of the spectrum of ^{60}Co as in Fig. 1 in the presence of a ^{133}Ba source producing a dead time of 91%

the fact that peak resolution is deteriorated, in the case of high count rate (i.e. also high dead time) pile-up effects or random coincidences occur. This can be clearly observed as a systematic change of the spectra in the high energy part of the 1 173 and 1 332 keV peaks of ^{60}Co as a function of dead time (Figs. 1-3). The specific shape of the spectra on the high energy part of the main peaks of ^{60}Co is a result of summing the energy deposited in the detector by the ^{60}Co photons with the energy deposited by the photons emitted by ^{133}Ba, mainly the KX-ray photons with the energy equal to about 31 and 35 keV.

In spite of the high count rate, the resolution and the Peak-to-Compton ratio did not change drastically. The corresponding values are presented in Table 1.

Table 1

Resolution, Peak-to-Compton ratio and peak count rate in the peak of 1332 keV for the n-type detector as a function of dead time

Dead time [%]	Resolution [keV]	Peak-to-Compton Ratio	Count rate [s^{-1}]
0.3	1.975	54.0	2.928
27.6	2.008	51.5	2.098
47.9	2.013	49.9	1.501
61.5	2.062	47.6	1.127
91.3	2.124	36.8	0.244

4. COINCIDENCE SUMMING EFFECTS

From the point of view of laboratory throughput the benefits of using high efficiency detectors are evident. There are however drawbacks associated with these detectors, namely the presence of high coincidence summing effects in close to detector measurement configurations. In order to take full advantage of the positive features of high efficiency detectors a thorough understanding of these effects, including the evaluation of coincidence summing corrections, is required.

Coincidence summing effects are of two origins. Random coincidences occur in the case when just by chance two photons emitted by different nuclei happen to interact with the detector so closely in time that the detector cannot resolve them into two different signals. Random coincidences are more and more important as the count rate increases, because the distribution of time intervals between two successive decays of different nuclides is displaced towards shorter time intervals in this case.

True coincidence summing effects are produced when two or more than two photons emitted in the decay of the same nuclide happen to interact with the detector. Normally the time interval between the emissions of all the photons along the same decay path is much shorter than the resolving time of the detector system

and consequently the detector will deliver a single signal, proportional with the summed energy deposited by all these photons together. True coincidence summing effects do not depend on source activity or count rate, but depend on the decay scheme of the nuclide. The evaluation of these effects requires a specific combination of decay data of the nuclide with the probability of photon interactions in the detector [6–8].

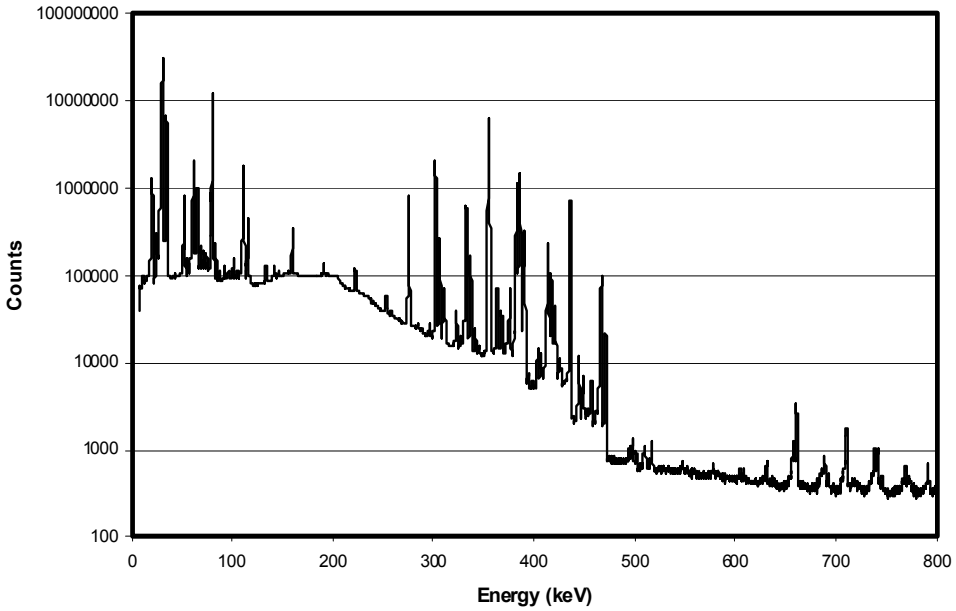

Fig. 4 – Spectrum of a ^{133}Ba point source measured on the end cap of the n-type detector.

As an exemplification of the magnitude of coincidence summing effects in Fig. 4 the spectrum of a ^{133}Ba point source placed on the end cap of the n-type detector is displayed. In the decay of ^{133}Ba 9 gamma rays with energies equal to 53, 79, 81, 160, 223, 276, 302, 356 and 383 keV are emitted; also X-rays with the energies equal to 31 and 35 keV are emitted. Correspondingly in the spectrum of ^{133}Ba measured in the absence of coincidence summing effects (e.g. with a low efficiency detector) only 11 peaks, with the energies given above, are expected. All the other peaks from the spectrum displayed in Fig. 4 (except the 661 keV peak of ^{137}Cs) are the result of coincidence summing effects. Most of the peaks are due to true coincidence summing, but random summing is also present (the dead time was 27% in this measurement). For example the peaks from the higher energy part of the spectrum are due to random summing, e.g. the peak at the energy of 712 keV is the result of summing two 356 keV photons emitted by two different nuclides (712=356+356 keV). Note that although the count rate in the 356 keV peak is very

high, the count rate in the 712 keV peak is low; from the count rate in this peak the order of magnitude of the random coincidences sum peaks can be inferred. It is clear that, even if random summing contributes also to the peaks that can be attributed to true coincidence summing, this contribution is much smaller than the contribution of true coincidence summing.

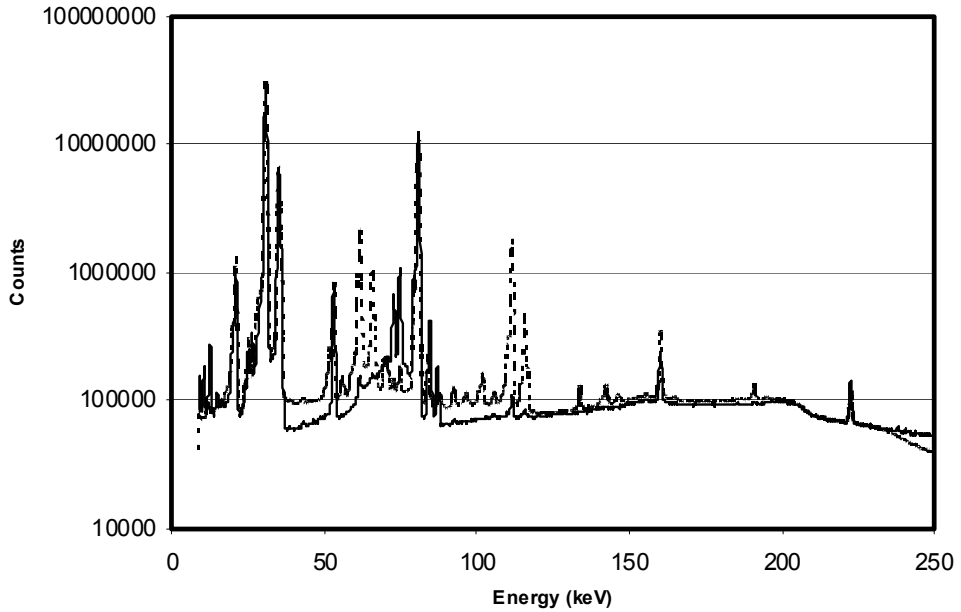

Fig. 5 – The first part of the spectra of the ^{133}Ba source measured on the detector end cap (dashed line) and at 15 cm distance (full line) with the n-type detector. The spectra were normalized to give equal number of counts in the 356 keV peak.

A convenient way of evidencing the magnitude of the coincidence summing effects is to display the spectra of the same point source measured close to the detector and far from the detector. In the absence of coincidence summing effects the spectra should look similar, with the count rate in the peaks proportional with the solid angle. If a normalization factor is applied in such a way as to provide equal count rates in a selected peak in the two spectra, then the two normalized spectra should be practically identical (minor differences might be present due to background contribution). The deviation from this expectation is entirely the result of coincidence summing effects. In Figs. 5 and 6 two energy ranges from the spectra of the ^{133}Ba source located at 15 cm from the detector (full line) and directly on the end cap (dashed line) are displayed. The two spectra have been normalized at the 356 keV peak. The spectrum of the source measured close to the detector contains many peaks that are absent in the other spectrum (or have a much

smaller count rate). It resembles the spectra from ref. [9], where the detailed explanation of the origin of the peaks is presented. Briefly speaking, the peaks at 132, 134, 304, 357 and 437 keV are due to sum peaks involving only gamma photons, while all the other pure sum peaks are due to summing of at least one X-Ray photon with other photons. Due to the high efficiency of the detector, coincidence summing effects are not completely negligible even for the source located at 15 cm (note e.g. the presence of the 437 keV sum peak).

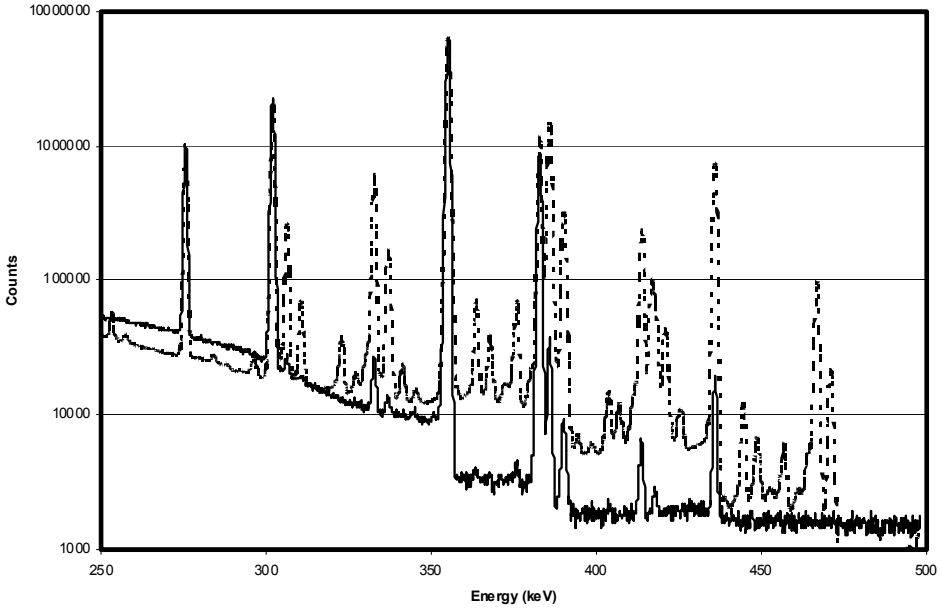

Fig. 6 – Same spectra as in Fig. 5 for the energy range 250–500 keV.

In the case of the p-type detector summing with X-Rays is strongly suppressed. In Fig. 7 the spectrum measured with the p-type detector equivalent with the spectrum presented in Fig. 4 is displayed. The only prominent pure sum peak is at the energy 437 keV; please note that the background is higher than in Fig. 4, so small peaks are not easily observed.

In Figs. 4–7 the presence of pure sum peaks, that result from coincidence summing and do not correspond to any single photon emitted by the source, is clearly observed. It is important to mention that the correct identification of all these peaks, including sum peaks with X-Rays, is important in spectrum analysis, in order to avoid falsely attributing a sum peak to another nuclide. The other effect of coincidence summing, namely coincidence losses from the peaks, is also important in spectrum analysis for obtaining an unbiased activity of the nuclide.

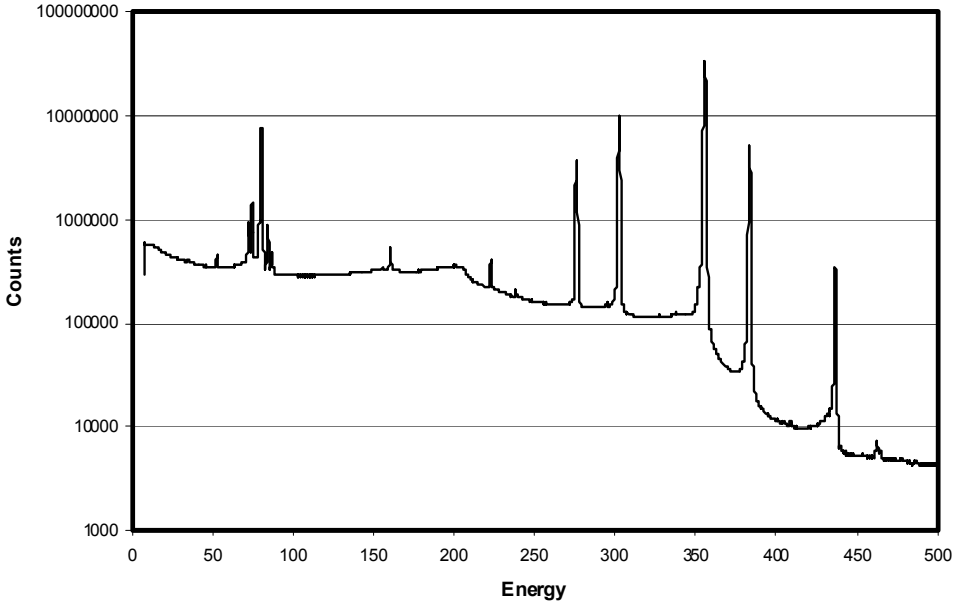

Fig. 7 – ^{133}Ba spectrum measured with the p-type detector. The source is located directly on the detector end cap.

5. DEVELOPMENT OF A GAMMA RAY SPECTRA DECISION TOOL

The first applications of the gamma-spectrometry systems presented above were the analysis of the detection limits for charcoal filters from Cernavoda Nuclear Power Plant and the assessment of environmental radiation dose for archaeometry samples. In the analysis of the spectra by standard tools some inconveniences were found in these applications.

The application of luminescence dating in archaeometry is based on the following idea. In various materials irradiation produces excited states that can be observed by analyzing the luminescence signal, either thermally or optically stimulated. The intensity of the signal detected with a TL/OSL analyzer is proportional with the dose accumulated in the crystals of the sample from the moment the luminescent information was reset (for example by burning an ancient brick in an oven in the roman period) to the moment the analysis is performed. If the environmental radiation dose rate delivered to the sample is known, then dividing the paleodose by the dose rate gives the age of the object, after considering a few correction factors depending on the nature of the artifact and the region where it was found [10]. The dose rate of interest, mainly delivered by the natural radioactive series of U-238 and Th-232 and by K-40, can be obtained by

analyzing the gamma spectra from the environment of the sample. In view of this application soil samples collected from the places of interest for archaeometry were analyzed.

We systematically considered blank samples for measuring activities in the exact same conditions as for the soil samples: same matrix, same sealant, same acquisition time, etc. Soil samples need to be sealed at least a couple of weeks before measurement, in order to avoid non-equilibrium in the decay chain of U-238 which can be provoked by partial loss of Rn. Some sealants contain β+ emitters, which influence the 511 keV peak; the matrix itself has a specific activity per mass unit, and it's the main reason for using identical blank samples, for they have the same mass & mass distribution within the volume. The subtraction of peak areas from the blank sample is very important, because when we approach the detection limit due to the very small count rates, it can lead to change the decision, or at least to proceed with increasing acquisition time; this way we minimize the chances of obtaining false positives/negatives (type I or II errors).

In order to use and develop this custom method for analysis and obtain more accurate results for different regions of interest in the spectra, we decided to develop in-house analysis software, which would not be based on fitting the curves, but would rather use a numerical approach. The main reason for this choice is that fitting functions do not necessarily provide the best results when getting close to the detection and decision limits. On the other hand, while having a decent statistic result for the number of counts cumulated in a peak, the most natural way to get closer to the real activity while comparing with calibration results seems to be the use of the exact number of events recorded in the concerned region. The standard region concerned in our approach is the area above the Full Width at Half Maximum, which has the advantage of including counts that have much greater chances of belonging to the peak we analyze than any other extension (for example considering 2 or 3 σ).

But the most important feature of this software is probably the procedure which leads to asking for a human decision in any controversial situation that cannot be solved by computing the standard way. There are quite a few such situations to describe, but they can be summarized as belonging to one of the three following cases:

– anomalous background;
– anomalous peak width and shape (without recognizing a multiplet);
– black swan type events.

A black swan type event is an event that is first a surprise to the observer, but which has a major impact, and after the fact, the event can be rationalized by hindsight, as if it had been expected [11].

The program is called GADETOOL, and a simplified screen shot of the main window is shown in Fig.8.

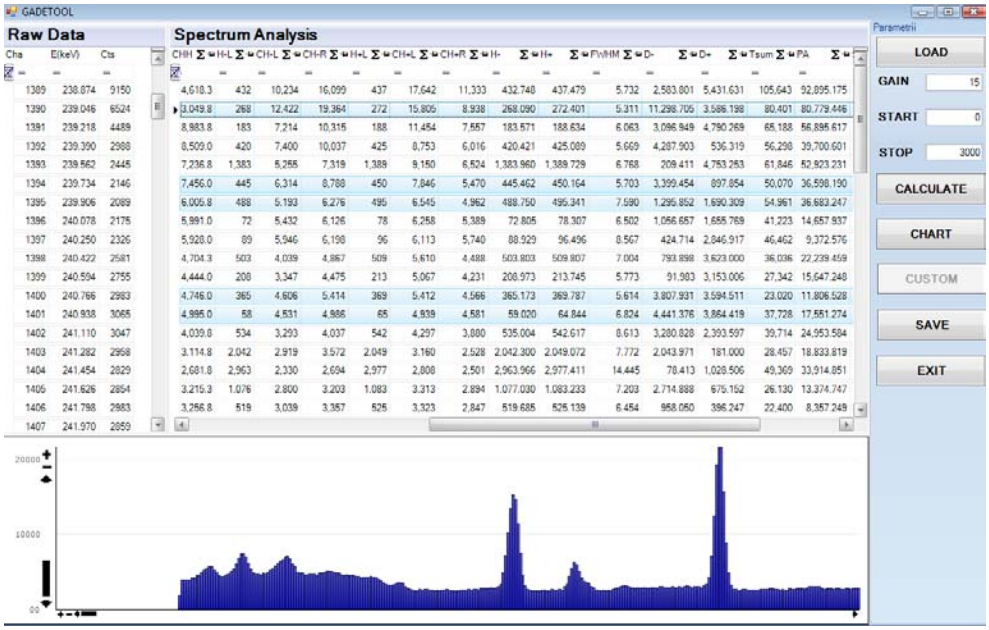

Fig. 8 – Main window of the GADETOOL (version β) peak analyzer.

The first application was to test GADETOOL on calibration spectra, of which manual analysis had been performed previously, so we could check for the accuracy of the results.

In the analyzing procedure, a manual check of the whole spectra is performed after GADETOOL calculations, no matter the results reported (i.e. even if no human decision is asked at all). This ensures first of all checking the results, and also the possibility of learning how to make this tool more valuable. As accurate analysis of the spectra is needed for minimizing the associated uncertainty, work directed towards developing GADETOOL in order to become a powerful and more versatile analysis and decision program for difficult spectra is in progress.

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