Erasmus Mundus Program

Activity Report

25/07/2019

Title: *Quantus* software: detector's calibration and efficiency curve

Abstract

Detectors need calibration before starting data acquisition in both energy and full width half maximum of the signal (FWHM). The first requires multiple known energy peaks so several gamma sources are used, while the second is related to the fact of having Gaussian distribution shapes in the spectrum centred in the peak energy instead of a sharp shape. All the spectra collected and calibration done bring the possibility of calculate the efficiency of the detector, which is a curve showing the relation between energy and efficiency. Thanks to that curve scientists are able to set the best conditions for the acquisition, as well as to determine the activity of an unknown source.

Quantus is CAEN high performance software to make Quantitative Spectrometry with Hexagon digital MCA. It permits working with many spectra at the same time. Since it allows setting parameters in the hardware, collecting data and analysing them, it is a useful tool for calibration purpose.

Purpose of the experiment

The main purpose of the lab session is to get in touch with *Quantus* software. In order to understand its operational way, a two-steps standard procedure in characterization of the NaI(Tl) solid state detector is followed.

The first goal is to find the appropriate calibration parameters in a NaI(Tl) detector using different radioisotopes, and the second one is to determine the activity of one unknown source using the efficiency calibration curve calculated from certification points.

Introduction

An inorganic scintillator detector of doped sodium iodine NaI(Tl) is calibrated in this experiment. NaI(Tl) it is an inorganic crystal which have the properties to emit light when ionizing radiation interacts in it.

Since photoelectron statistics plays a key role in the accurate determination of the energy of the radiation, the use of scintillation materials with a high light output is preferred for all spectroscopic applications. On the other side, fast light emission through fluorescence process improves the measurement of the ionizing radiation interaction time.

Efficient detection of Gamma rays requires the material of the scintillator to be high dense and with high atomic number. NaI(Tl) scintillation crystal meet the requirements of stopping power and optical transparency (refraction index ~ 1.9): its density of 3,67 g/cm^3 makes it very suitable to absorb penetrating radiation like Gamma rays.

Model	Material	Dimension	Decay Constant	Voltage Divider	Resolution for 137Cs
76B76/3M	NaI(Tl)	3” Ø x 3" h	0.23 ms	no	< 7,5 %

Table 1: Specifications of NaI(Tl) detector provided by the company. Available in Datasheet from https://www.caensys.com/inorganic_sd/

For nuclear detection at room temperature as it is the case, signal created in the detector depends not only to the energy of the incident photon but also to the position of the interaction. This can bring an incomplete charge collection caused by a deep-trapping or a ballistic deficit of charge carrier. The ballistic deficit of a signal happens when the time taken by the carrier to reach electrode is longer than the period during which the signal is formed, and it reduces electron efficiency. In signal analysis from a large-volume detector such as the NaI(Tl), the effect should be corrected adjusting the time of the trapezoidal tail collection (Figure 2).

The most relevant part of the lab session is the technique used for the analysis, with a new software called “*Quantus*”. *Quantus* is a Quantitative Spectrometry Software in development not yet accessible to public use. It is an intuitive plus widely useful tool that allows the scientist to set hardware parameters from the computer as well as to analyse the spectra from several samples and to import the results. For the calibration in energy and FWHM, this program represents a powerful improvement from other time-consuming methods. For further specifications please go to the company webpage.

The program is capable to compare data with the estimated value using the residuals method. In regression analysis, a parameter called "Residuals" shows how appropriate is the Gaussian fit of the photopeak, ranging this fit in units of deviation (σ) until a maximum 2σ. The limits of the Region Of Interest (ROI) can be adjusted once the Gaussian fit is set for having as low residuals as possible. If the residual is too high, there is an overlapping then further analysis is required.

The precision of the fitting with data in this session will be acceptable for residual value below 2σ, twice the standard deviation, as it is shown in the correspondent section *Measurements> FWHM calibration*.

- Theoretical bases and uncertainties

The radioactive decay is a physical phenomenon happening when an instable nucleus reaches a new state of equilibrium emitting particle or radiation. The *activity* ($A(t)$) is defined as the number of decay for second, in Bequerel units [Bq].

$$A(\tau) = A_0 \cdot e^{-\lambda\tau} \tag{1}$$

Where λ is the decay constant of the nuclide, A_0 is the activity of reference and τ is mean time of the element. The activity of a source changes with time; this is why a reference activity of the source is needed in order to calculate the activity today.

The *efficiency of the detector* for a given source is the ratio between the number of counts inside a ROI and the activity of the source.

$$\varepsilon = \frac{N_p}{A_{source}} \tag{2}$$

While the number of counts inside the ROI is given by the program, the activity of each source is estimated by the provider of the sample. The error associated to the statistic could be reduced if there are at least 10000 counts under each ROI, because

$$\Delta N = \frac{1}{\sqrt{N}} \tag{3}$$

hence there would be an error of 1%. Unfortunately, some of the sources used are not active enough for achieving this value.

But the efficiency of a detector is different for each source, as a function of the energy of the photopeak of the source. Then, several sources provide many efficiency points. It means there is a fitting curve for efficiency which follows the expression (4).

$$\ln(\varepsilon) = \sum a_k \cdot \ln(E)$$

(4)

where a_k are weight parameters calculated by *Quantus* with the aim of better fitting the curve to the experimental data.

A way of testing the calculation of the efficiency curve is to deduce the unknown activity of a source. From expression (2) it is easily noticed that the activity of the source in relation to the efficiency is

$$A_X = N_p \cdot \varepsilon \tag{5}$$

According to the laws of propagation of uncertainties, the uncertainty associated to the activity of the sample is

$$\Delta A_X = \Delta N_p \cdot \varepsilon \tag{6}$$

All these expressions are used in the following sections for calculating the efficiency of the detector and the activity of the source. Along next sections these formulae will be referred by their numbers (1-6).

Material and procedure

Energy calibration requires several gamma sources. Samples used here were Co-57, Co-60, Cs-137 and Na-22, some of them with very low emission rate. The detector used is NaI(Tl) solid scintillator, with low energy resolution which specifications are included in Table 1. It is connected to the *Hexagon* digital multichannel analyser (MCA), and this to the computer. Due to the importance of the software part of the experiment, images shown below will illustrate steps and results on the *Quantus* interface. The sequence for performing the spectrometry analysis is the following.

Figure 0: Scheme of the experimental setup: radioactive source (A) in front of the inorganic scintillator (B), which is connecter to the MCA Hexagon (C) *(see more here) supplied by the software* Quantus (D).

Firstly, the connexion of software with hardware is done manually, selecting the appropriate high voltage (HV) as shown in Figure 1.

In this case the signal of HV applied to the PMT must be positive. The voltage suggested by detector's provider is 800V. Since NaI(Tl) detector is not as sensitive as HPGe, the ramp or rise of the voltage can be relatively fast. The system shows the value of voltage and current.

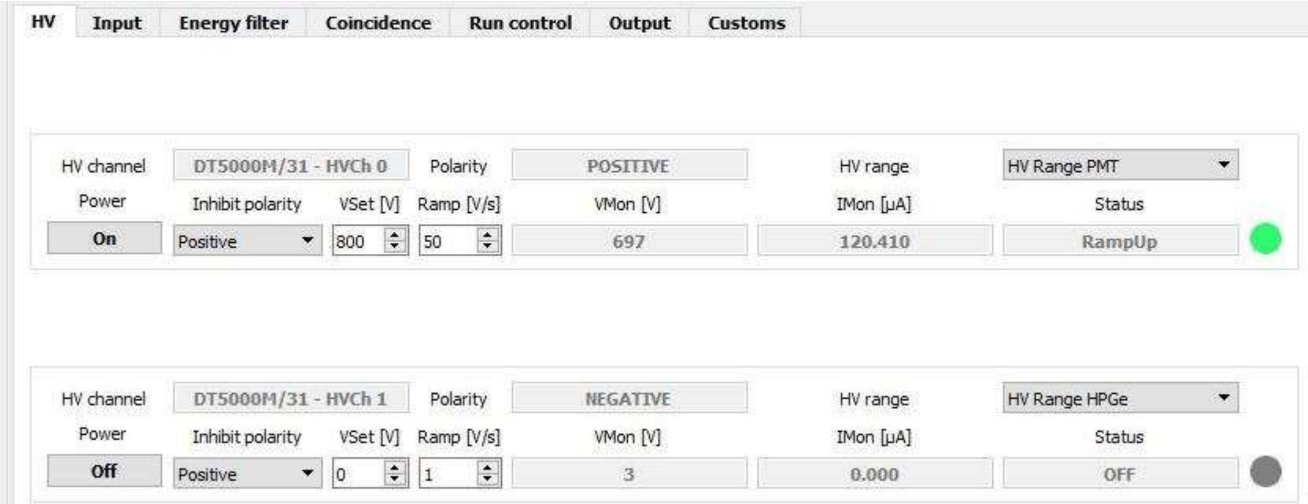


Figure 1: Interface settings of the HV and power supply. Quantus *allows the user to select the configuration of the hardware from the same software.*

The optimization of input parameters is done automatically (Figure 2), with values of 40% of DC-offset, threshold of 2 LSB (Least Significant Bit) and a trapezoid tail correction of 33.25 μs for the particular case of this experiment. The Figure 3 below shows the shape of the input wave and the optimized trapezoid

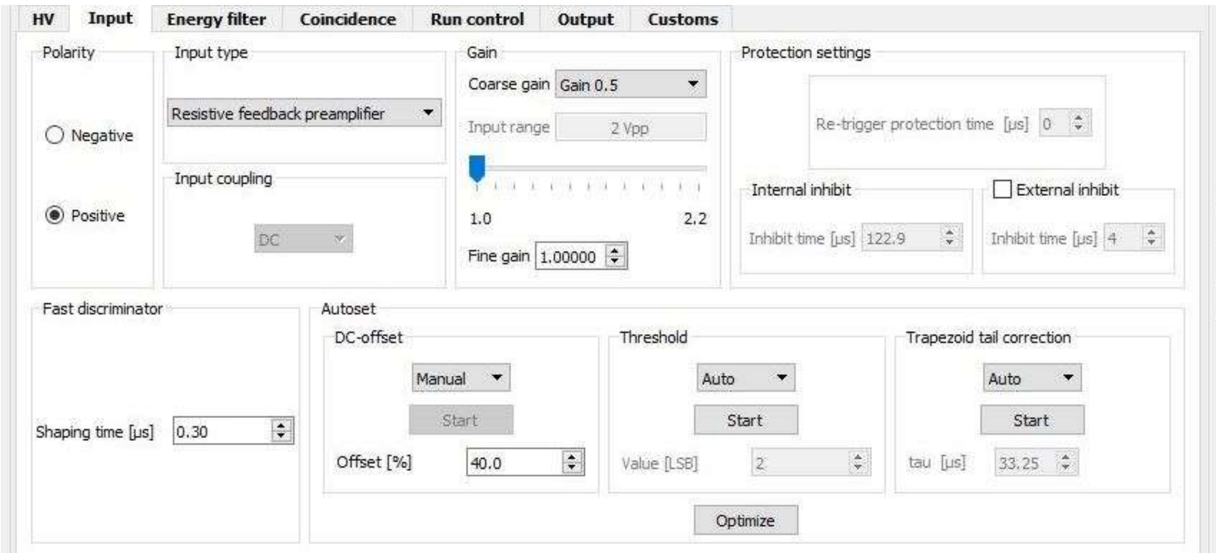


Figure 2: Input settings, including polarity of the signal, gain and fast discriminator. The software is able to autoset the most convenient values for the DC-offset, the threshold and the trapezoid tail correction.

Due to the ballistic effect of the NaI(Tl) detector mentioned in the first section, the plateau part of the trapezoid is not that flat, hence a change in time (µs) is required to give additional time to the device for collecting charge.

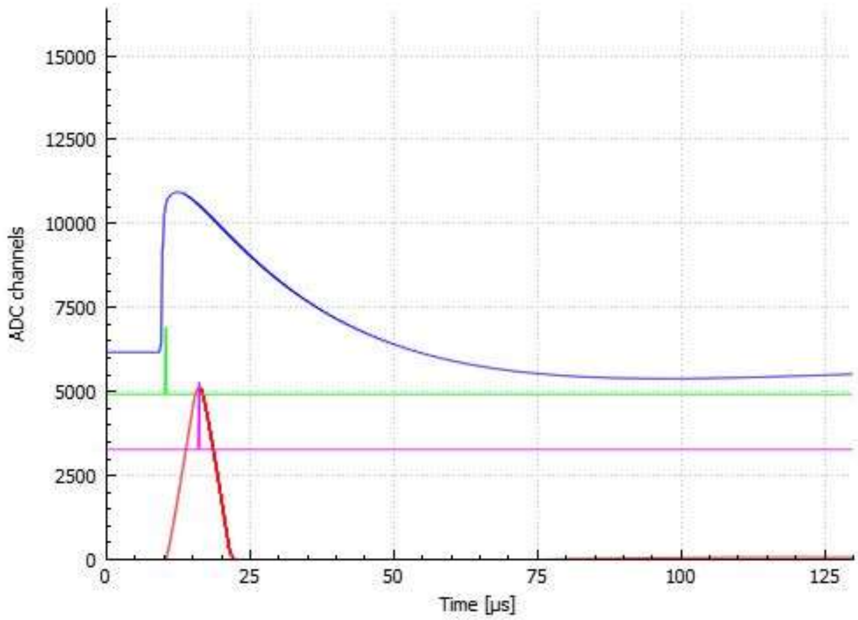


Figure 3: Optimized input signal (blue) and trapezoid discriminator (red).

Once the calibration of the hardware parameters is set, and the shape, amplitude and characteristics of the input signal is adequate to avoid undesired effects, measurements for energy calibration start.

Measurements

- Energy calibration

Different gamma sources are measured with the purpose of calibration. For the energy calibration good statistics are required, hence a reasonable number of counts. Due to low activity of the Co-60 source placed at first in front of the detector, counts are not very high.

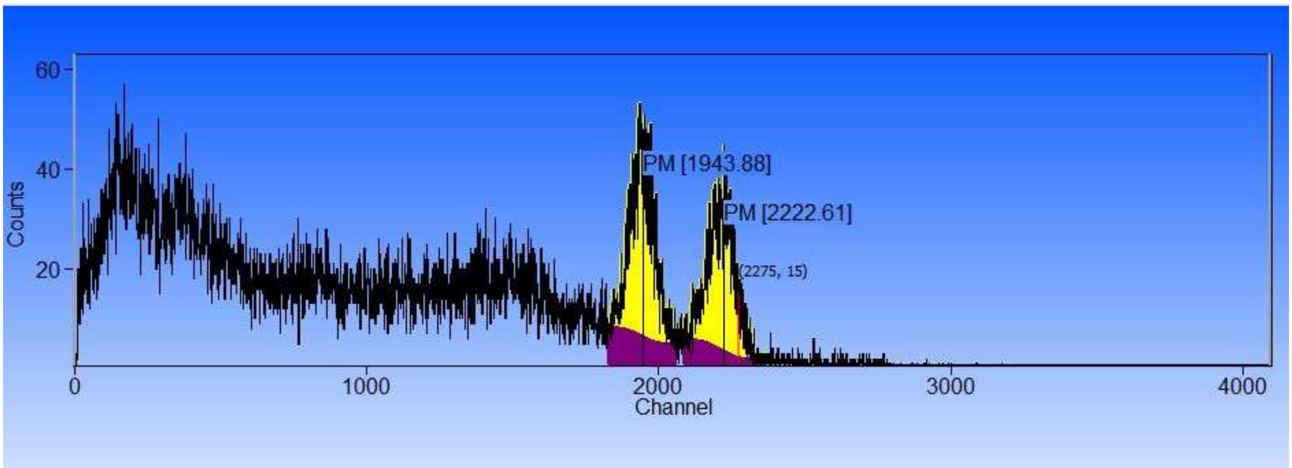


Figure 4: Spectrum of Co-60 with ROI over the photopeaks of its two gamma peaks: 1173 keV and 1332 keV respectively.

Centroid (keV) of Co-60	
ROI 1	1943.9 ± 7.7
ROI 2	2215.1 ± 8.4

Table 2: Centroids of the distribution in energy of Co-60 photopeaks.

The purple region is the estimation of the background on each peak according to the spectrum line before and after the photopeak.

Knowing that the most probable gamma emission energies of Co-60 are 1173 keV and 1332 keV the first calibration in energy is done. Now each ADC channel has its correspondently energy according to the match of the centroids of ROI regions with these theoretical values.

As it is shown in Figure 5, the software *Quantus* requires theoretical values for doing the energy calibration automatically. All data for the expected gamma rays emitted are taken from Lara Module.

Energy calibration could be finished with only the measuring of one source, even if this is not the most active one. Nevertheless, this calibration may improve with the addition of other photopeaks coming from several samples with known gamma energies, such as Cs-137 and Co-57, which expected energy are 661,64 keV and 122 keV respectively.

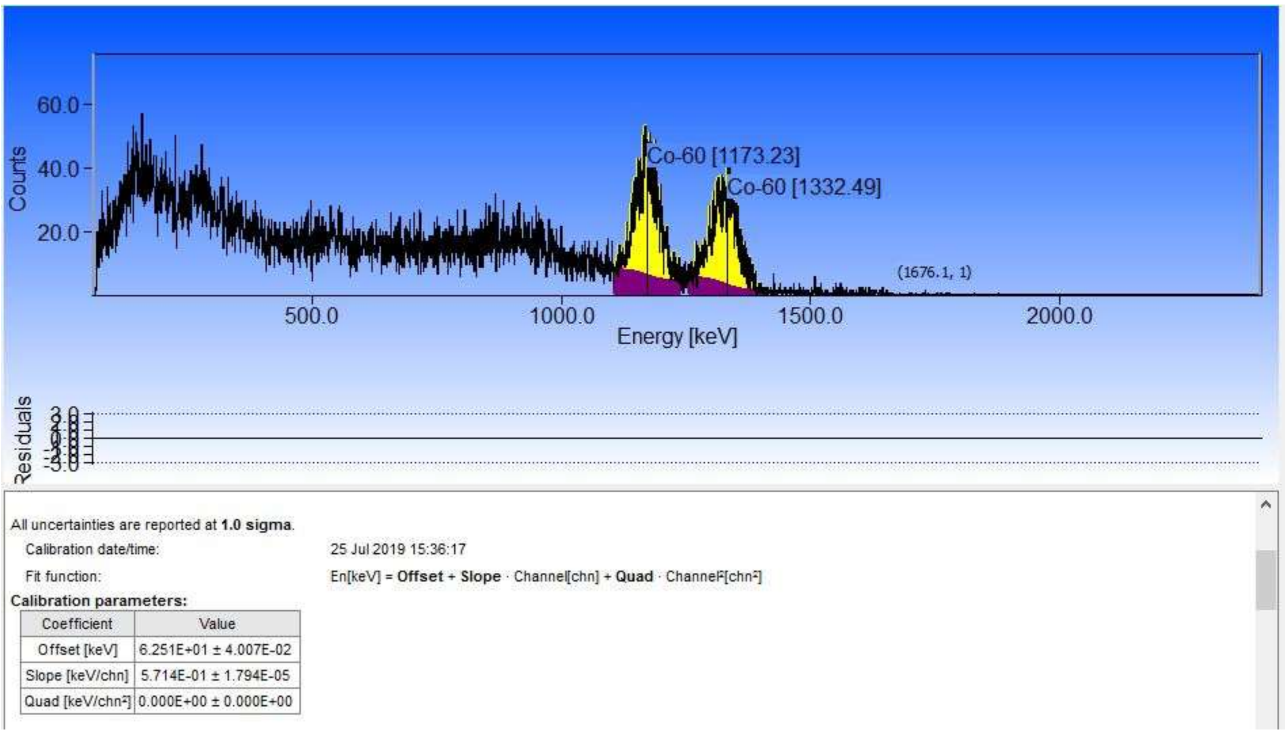


Figure 5: Calibration parameters are automatically set by the software once the user determines the ROI and the correspondent energy of the centroid.

- <u>FWHM calibration</u>

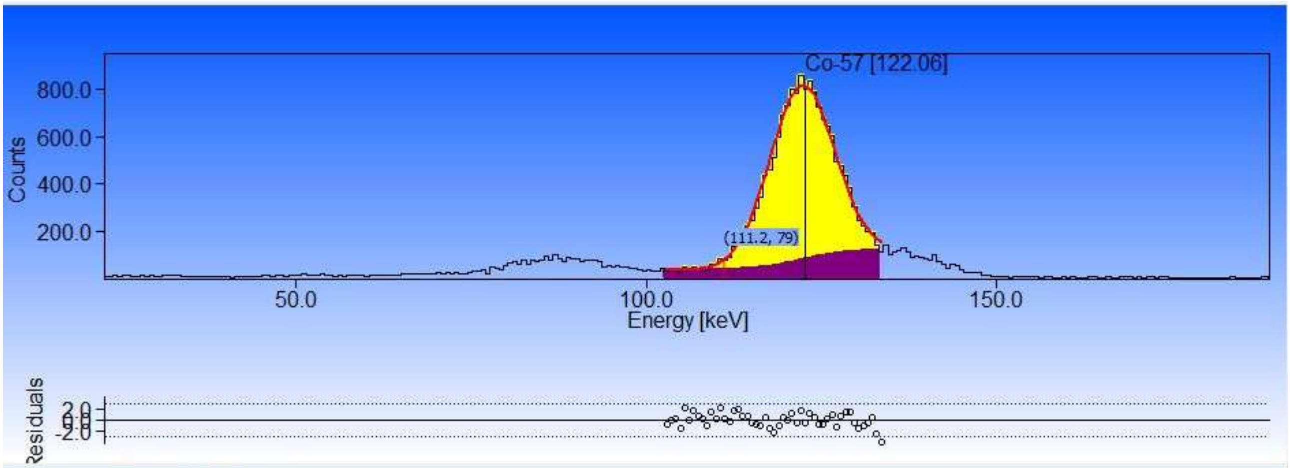
Photopeaks should be seen in the spectrum as sharp as possible, due to the monoenergetic gamma emitted. In real spectra a Gaussian shape appears instead. FWHM calibration is done thanks to the fit of all the ROIs from several samples. In order to calibrate the FWHM of the peaks, and also to add more points to the energy calibration, a sample of Co-57 is used. This sample has higher activity than the previous one, consequently it provides better statistics.

At this point residuals play a key role for the acceptance of the fitting as explained in section "Introduction". In Figure 6, residual points are presented under the spectrum, with values inside the $\pm 2\sigma$ range.

A third gamma source, Cs-137, is analysed again with the aim of checking the validity of the energy calibration and adding points for the FWHM calibration. As it is shown in Figure 7, the spectrum of Cs-137 has a peak corresponding to X-Ray emission, while the interesting photopeak has 662.1 keV. Residuals from the analysis of Cs-137 are checked in Figure 8.

Spectrum from Cs-137 presents a peak compatible with the value of energy registered on the library of *Quantus*, therefore the calibration in energy has been checked and seems to be correct.

A last Na-22 source with unknown activity is placed in front of the detector. It is used with calibration purpose too because its photopeak has 1274 keV, higher than previous sources so it adds a calibration point in the high energy side of the spectrum. The software is able to add all the spectra into a common plot, consequently the aim of this step is not only to improve the calibration but also to keep on learning about different uses of the software *Quantus*.



All uncertainties are reported at **1.0 sigma**.

Peak label	Related ROI	Position | Std.Dev. [chn]		Position | Std.Dev. [keV]		FWHM | Std.Dev. [chn]		FWHM | Std.Dev. [keV]		FWHM | Std.Dev. [%]		Area | Std.Dev. [cnts]		Continuum | Std.Dev. [cnts]		Reduced χ² [aunits]	Identified nuclide (NID)
PM [122.23]	ROI-1	211.31	0.09	122.23	1.00	16.93	0.18	10.79	0.16	8.82	0.15	13306.02	175.30	3216.11	155.34	1.53	Co-57 [122.06]
PM [1173.23]	ROI-2	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	Co-60 [1173.23]
PM [1332.49]	ROI-3	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	Co-60 [1332.49]

Overall reduced χ² = 1.53

Figure 6: Gaussian fit to the ROI on the 122 keV photopeak of Co-57.

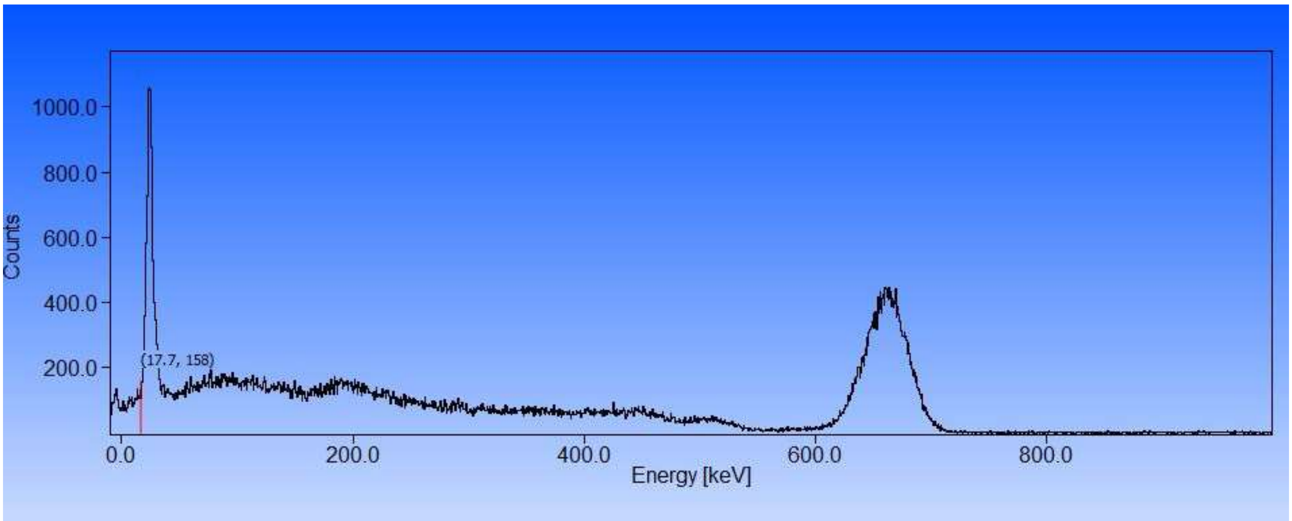


Figure 7: Spectrum of Cs137. The sharped peak corresponds to a X-Ray emission. The interesting part of this spectrum is the Gaussian distribution corresponding to 662.1 keV.

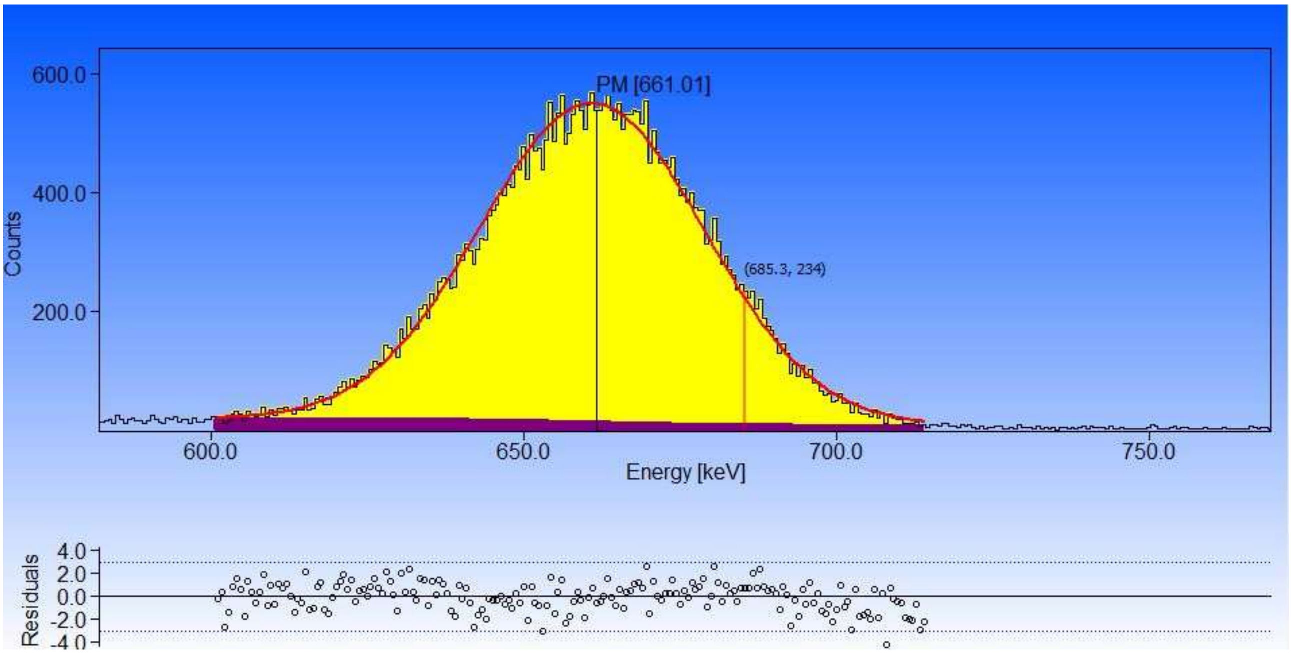


Figure 8: Resolution Cs-137. As in previous images, instead of a sharp shape there is a Gaussian.

Once the calibration in FWHM and energy are done, it is time to calculate the efficiency of the detector for a particular energy: the efficiency curve uses all the previous measurements and leads to obtain the unknown activity of a source.

- <u>Efficiency calibration curve</u>

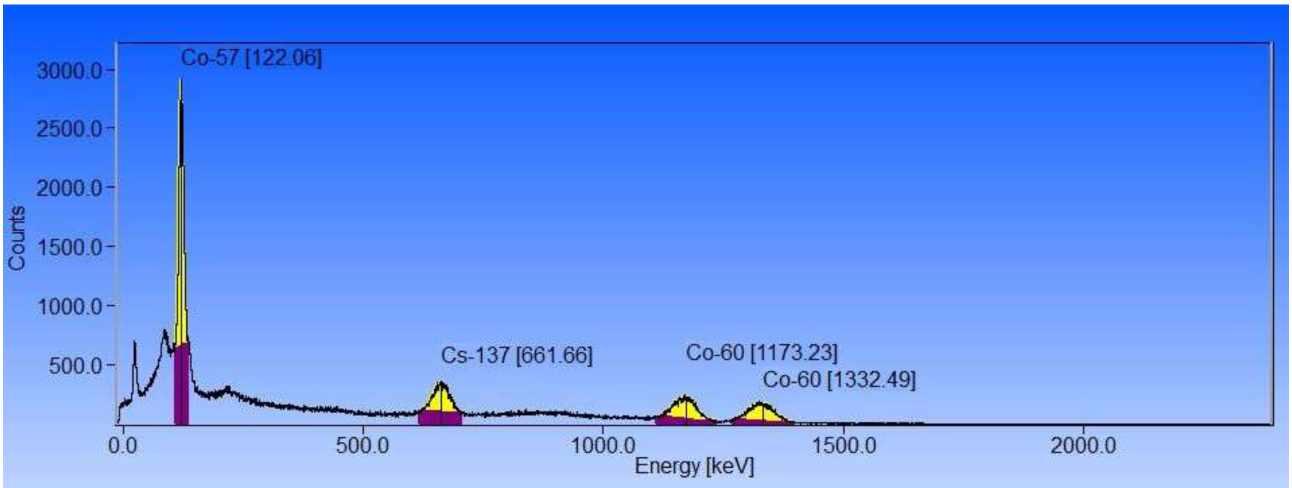


Figure 9: All the spectra together. The gamma energy of each source appears in the plot.

The efficiency of the detector is calculated using the expression (1). While the number of counts inside the ROI is given by the program, the activity of each source is estimated by the provider of the sample. The uncertainty associated to the statistics follows instead the definition in (2).

The analysis of the efficiency ratio for all the sources analysed (those presented in Figure 9) gives several efficiency points which follow a specific curve according to the detector characteristics. For calculating detector's efficiency the relative distance between detector and the source is constant, being 6 cm in the case of this experiment.

An aspect that must be taken into account while performing the experiment is that diverse samples have different activity rates, so the most active ones might cover the peaks coming from the less active.

Data analysis

Data from the samples such as energy of gammas emitted, their intensity and reference activity as well as the activity today is presented in the following table.

Nucleide	Type	Half life	Energy [keV]	Intensity [%]	Activity Ref [Bq]	Activity today [Bq]
Cs-137	γ	30.08	661.657 ± 0.003	85.1 ± 0.2	$(9.25 \pm 0.04) \cdot 10^3$	$(7.33 \pm 0.03) \cdot 10^3$
Co-60	γ	1925.28D	1173.228 ± 0.003	99.85 ± 0.03	$(3.700 \pm 0.004) \cdot 10^4$	$(1.645 \pm 0.002) \cdot 10^4$
Co-60	γ	1925.28D	1332.492 ± 0.004	99.983 ± 0.001	$(3.700 \pm 0.004) \cdot 10^4$	$(1.645 \pm 0.002) \cdot 10^4$
Co-57	γ	271.74D	122.061 ± 0.002	85.60 ± 0.17	$(1.850 \pm 0.004) \cdot 10^5$	$(1.811 \pm 0.004) \cdot 10^4$

Table 3: Relevant data from all the analysed sources. The activity of reference is given by the provider of the sample.

For each sample a certificate has been created, which includes efficiency of the detector to that source and its uncertainty. Results are presented in Figure 10, where the fitting curve for efficiency follows the expression (4).

Values for these variables a_k are shown in the Figure 10, and they provide the efficiency curve that can be observed in the bottom left of the image.

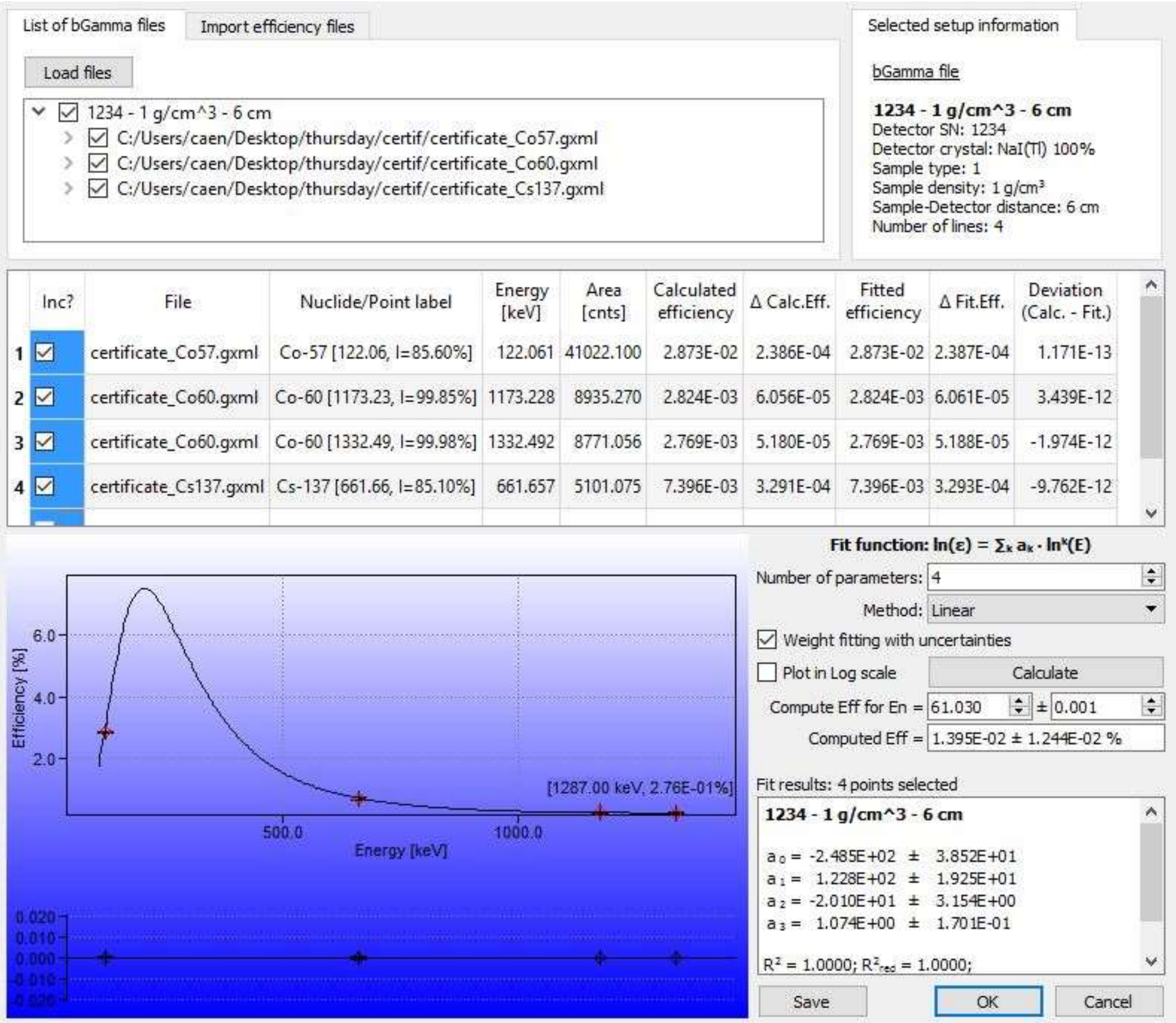


Figure 10: Efficiency curve and its parameters.

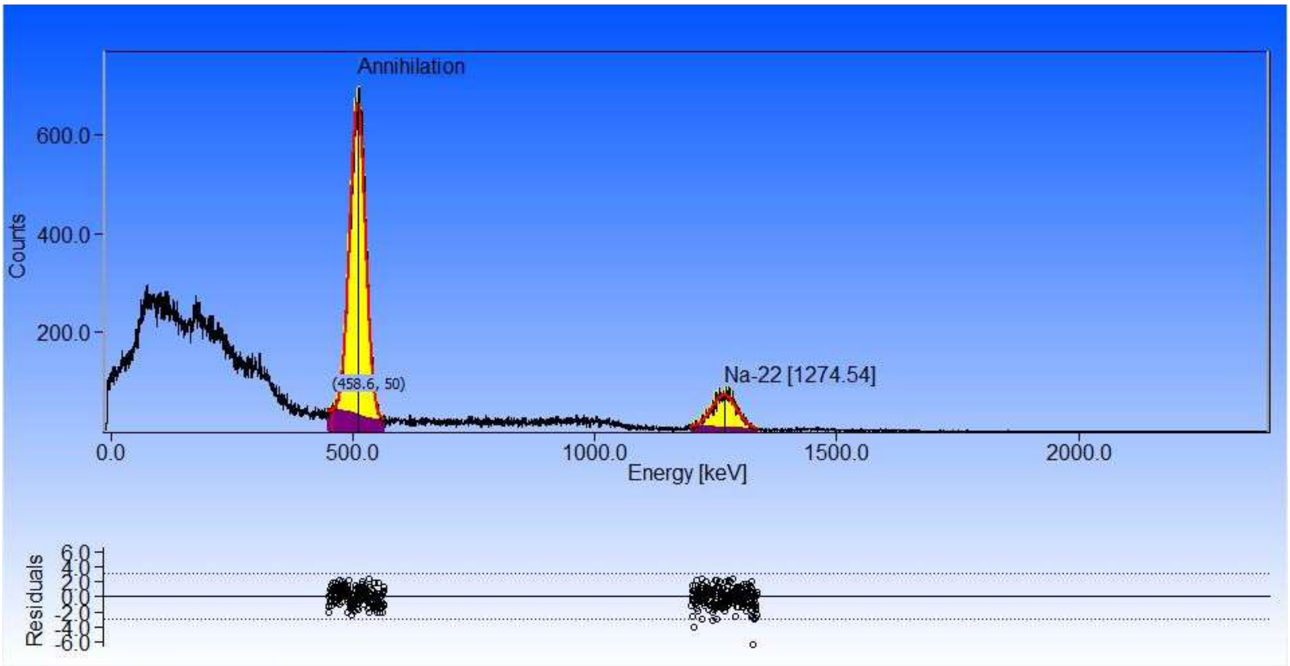
All efficiency calibration curve need to have a peak followed by a logarithmic decrease. The critical part lays at low energy, below the peak. This peak represents the most efficient parameters of the detector. Under that point there is a conflictive region due to absorption effects.

- <u>Determination of unknown activity of a sample</u>

The efficiency is related to the activity of the source as it is seen in equation (2). It means that once the efficiency of the detector for a fixed value of energy is known, the activity of the source responds to the equation (5).

With the aim of checking the validity of the efficiency value obtained throw Co-60, Co-57 and Cs-137 samples calibration, the activity of an unknown source is calculated.

A sample of Na-22 has been chosen, which gamma energy is 1274,55 keV. Substituting this value inside (5) the efficiency of the detector for that energy is obtained.



All uncertainties are reported at **1.0 sigma**.

Nuclide	Line labels	Reference energy [keV]	Reference intensity [%]	Line activity [µCi]	Nuclide activity [µCi]
Na-22	PM [1268.17]	1274.537 ± 0.007	99.940 ± 0.014	(1.090 ± 0.215)E+00	(1.090 ± 0.215)E+00

Figure 11: Characterisation of the source and activity.

The uncertainty associated to the activity of the sample is expressed by the formula (6), hence the result for the activity of the Na-22 sample is, as presented in Figure 11:

$$A_{22Na}^{exp} = (\ 1.09 \pm 0.22\ )\ \mu Ci$$
$$A_{22Na} = 1\ \mu Ci$$

Results & Conclusion

Reference activity of a Na-22 source has been established using the efficiency curve of the detector previously calculated with *Quantus* software. Accordingly to the satisfactory results, it can be said that perform of the experiment and the acquisition of measurements was a success.

The uncertainty is around a 20%, probably due to the fact that the efficiency curve needs more radiative samples to add different points all along the spectrum for calibration. The fact of having some sources very actives and others with very low activity leads to a variation in the number of counts per ROI for each sample. Since the purpose of the experiment is educative and the time for performing it was not too extended, this result has been considered as adequate by the experimented scientist presented at the lab.

Experimental and theoretical data are compatible in 2σ so the method for calculating the efficiency curve of the NaI(Tl) detector has been successfully performed. Library from *Quantus* is able to identify the kind of source and the annihilation peak associated to it, another proof of the good calibration of the spectrum.

As in many other experiments, energy calibration is improved with the addition of different samples with more than one gamma peak. In the spectrum presented in Figure 9 the centroid of Cs-137 might be affected by the Compton Effect from the Co-60 hence it is shift in energy. This is the reason why it is more efficient to use a single emission source with several gammas, like in the case of Europium which does not show this adverse effect.

The experiment performed is a standard procedure widely used to find the calibration of a detector for several radioisotopes, and as in this case it could be applied to determine the activity of any other source. It is remarkable to mention the 'wave-like' shape of the efficiency curve: a pure logarithmic shape is not expected since the most efficient parameters for the detector at a given energy are pointed out by the peak of the curve. In case of not having this peak, the addition of a low energy gamma source to the certificated samples is required.

Patricia Pérez Martín, (University of Catania, Italy)

Julgen Pellumaj, (University of Seville, Spain)

Iftikhar Ahmad, (University of Seville, Spain)