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sIGAle, a New Code for Automatically Determining Radionuclide Activities Using CdZnTe Spectrometry

I. Espagnon, A. C. Simon, F. Lamadie and C. Mahé

Abstract — **The sIGAle code is developed by CEA LIST at the request of CEA DEN to analyse spectra measured with CdZnTe detectors. sIGAle is a specific automatic analysis software which allows a complete analysis of the spectrum, with automatic calibration, identification of radionuclides present in the spectrum, determination of fluence rates for each significant peak and, if the transfer function is known, calculation of activity for each radionuclide. The sIGAle code takes into account the asymmetric peaks with tails specific to spectra measured with CdZnTe detector and provides a reproducible evaluation of the peak areas, which was the major requirement of CEA DEN. The sIGAle code has been tested by CEA DEN in the field and in laboratories. This paper presents a detailed description of the general principle of the sIGAle code and experimental results.**

Index Terms—**CZT detector, gamma activities, gamma spectrometry, isotope identification, sIGAle code**

I. Introduction

In gamma spectrometry today, semiconductor detectors using cadmium telluride (CdTe and CdZnTe) are an attractive alternative to high-purity germanium (HPGe) diodes, especially in dismantling domain. Indeed, they present a reduced size, a weaker cost, and above all they are operable at ambient temperature without cooling system. Despite these qualities, processing of CdZnTe spectra is subject to a major difficulty: the photopeaks can be strongly dissymmetrical, which can strongly affect the results [1]. At the request of CEA DEN, a specific automatic analysis software code, called sIGAle (System of Identification of Gamma Activities at Low rEsolution), has thus been developed by CEA LIST to take into account this type of spectrum, with the robustness of the spectrum processing as a major requirement.

The sIGAle code is based on the same general algorithm as another analysis code, called IGA [2] and previously developed by CEA LIST to determine actinide isotopic composition from HPGe spectra. In particular, the new code uses the same methods and modules and it benefits from a similar original approach. However, the specific characteristics of CdZnTe probes needed to be taken into consideration: their poor energy resolution compared to HPGe detectors, the peak asymmetry, the presence of significant escape peaks which can affect the main peak and a resolution-energy relationship presenting a different model to the one of the HPGe detectors. Further difficulties have been encountered, like the high variability of behaviour according to the different probes or a poor counting statistics and a high contribution of scattering in the field spectra. In consequence, a complete procedure for spectra processing had to be defined, including a preliminary phase for the energy calibration, the peak shape calibration and the calculation of efficiency curve with standard spectra.

The software has continuously been tested by CEA DEN in laboratories and in the field during its development, which has enabled to correct and improve it. In particular, it was used during test campaigns at CEA/Fontenay-aux Roses, at CEA/Cadarache, at the CEA/Marcoule Pilot Unit and on Phénix reactor with different sizes of probes and with spectra relating to different radionuclides; it gave satisfactory results to the end-users, especially on the radionuclide identification.

II. Processing of Standard Spectra

The aim of standard spectrum analysis is to determine energy calibration, width calibration, peak shape, escape peaks calibration and efficiency curve. The general algorithm of standard spectrum analysis is presented in Fig. 1. This spectrum has to be measured with a good counting statistics and must cover a large energy range. Standard spectrum is necessarily ^{152}Eu or ^{60}Co + ^{137}Cs spectrum.

A. Input data

The minimum data required from the user by the sIGAle code are the spectrum in ASCII format (including acquisition time), a rough energy calibration, the date of the spectrum measurement, the present radionuclides with, for each of them, source-detector distance, activity and the date corresponding to this activity.

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Calibration
Energy calibration, resolution calibration and tail calibration

First estimation of efficiency
Analysis of regions of interest without escape peaks

Escape peak calibration

Second estimation of efficiency
Analysis of regions of interest with escape peaks

Fig. 1. General algorithm of standard spectrum analysis.

B. *Tail calibration*

In the sIGAle code, the net peaks are modeled by a Gaussian and a tail function and background is modeled by a step function and a polynomial. To reduce as much as possible the number of parameters in peak fitting, a function with only two additional parameters *p1* and *p2* has been chosen for tail model [3]:

$$T(x) = p1.h.e^{\frac{(x-\mu)}{p2.\sigma}}.erfc\left[\frac{(x-\mu)}{\sqrt{2}.\sigma} + \frac{1}{\sqrt{2}.p2}\right] \quad (3)$$

The width parameter σ determines the FWHM (Full Width at Half Maximum) of any Gaussian through the relation FWHM = 2.35σ. The two parameters h and μ represent the height and the centroid of the Gaussian, respectively. The remaining two parameters *p1* and *p2* do not depend on channel x. Fig. 2 shows an example of peak shape for two values of *p1* and *p2*.

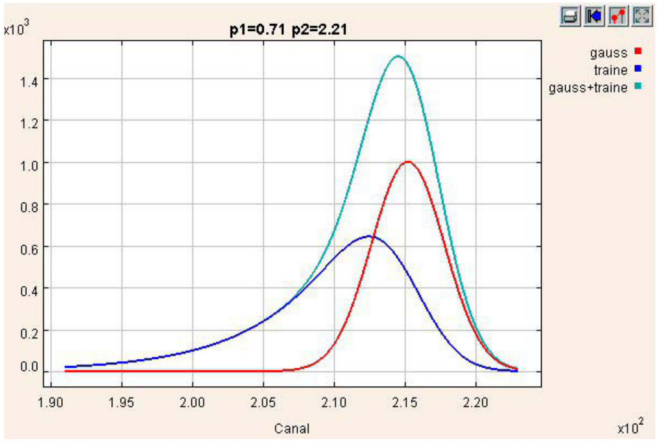


Fig. 2. Example of peak shape for two values of *p1* and *p2 (p1 = 0.71 and p2 = 2.21)*. Red component is the Gaussian, blue component is the tail function and green component is the sum of both.

The first step of standard spectrum analysis consists in determining the parameters *p1* and *p2* of the tail function (3) from a pre-defined peak: the peak at 344 keV for ^{152}Eu spectrum or the peak at 662 keV for ^{60}Co + ^{137}Cs spectrum. It has been checked that if these parameters are taken constant with energy, results are satisfactory. They are then fixed in the following process.

C. *Energy and resolution calibration*

The aim is to provide channel-energy and resolution-energy relationships:

$$i = a + b.E_i + cE_i^2 \quad (4)$$
$$\sigma_i = p + qE_i$$

i represents the channel number, E_i the energy at channel i and σ_i the width parameter at channel i. A second degree polynomial relationship is determined between the positions in channels of the fitted peaks and their energies. As for σ parameter, it grows linearly with the energy, which is different from HPGe detectors which have a linear relationship between σ^2 and the energy.

D. *Escape Peak Calibration*

Escape peaks arise whenever a fixed amount of energy may be lost from the detector with an important probability. The escape of the characteristic X-rays from CdZnTe following photoelectric absorption can be significant, especially for small detectors. Small peaks will then be found in the spectrum at 28 keV (Te escape), 24 keV (Cd escape) and 9 keV (Zn escape) below the photopeak, with the energy differences corresponding to the characteristic K X-ray energies for CdZnTe.

The ratios between the height of the photopeak and the heights of Cd, Te, and Zn escape peaks are determined from the peak at 344 keV for ^{152}Eu spectrum or from the peak at 662 keV for ^{60}Co + ^{137}Cs spectrum. For this fitting, *p1*, *p2*, σ and μ are fixed. Fig. 3 shows an example of deconvolution of a peak at 344 keV with its escape peaks. The calculated ratios are imposed constant with energy and remain fixed in the following process.

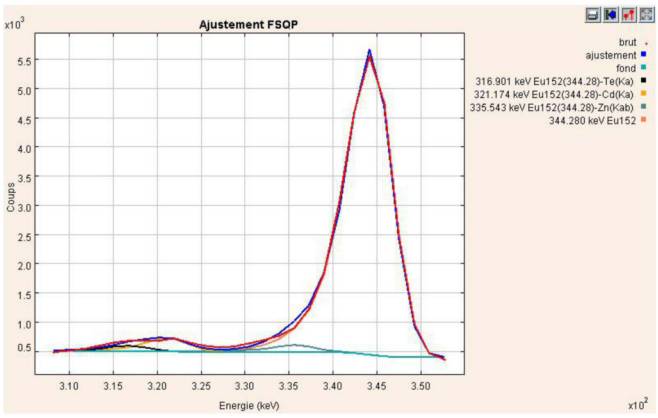


Fig. 3. Example of deconvolution of a peak at 344 keV with its Cd, Te and Zn escape peaks.

E. *Efficiency Curve*

The absolute efficiency *K(E)* of the detector is determined for a few peaks of energy E from measured net peak areas *S(E)* and photon fluence rates Φ*(E)*:

$$K(E) = \frac{S(E)}{t.\Phi(E)}, \quad \text{with} \qquad (5)$$
$$\Phi(E) = \frac{Act.I(E)}{4\pi d^2}$$

t is the acquisition time (s). $\Phi(E)$ is the total number of photons of energy E at the center of the detector ($s^{-1}.cm^{-2}$). Act is the activity (Bq) and $I(E)$ is the probability of photon emission of radionuclide at energy E. d is the source-detector distance (cm).

Efficiency curve is then obtained by fitting points with a log polynomial function:

$$\ln(K(E)) = a_0 + a_1 \ln(E) + \ldots + a_n \ln(E)^n \qquad (6)$$
$$with\ 1 \le n \le 5$$

F. Case with Several Standard Spectra

Several standard spectra can be used to establish efficiency curve (Fig. 4). Resolution calibration, peak shape and ratios of escape peaks are determined from the first standard spectrum and are fixed for other standard spectrum analysis.

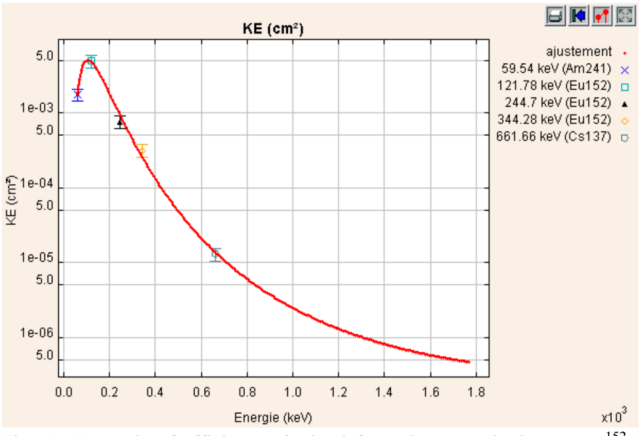


Fig. 4. Example of efficiency obtained from three standard spectra: ^{152}Eu, ^{137}Cs and ^{241}Am. Red curve is a log polynomial function.

III. Processing of Field Spectra

The aim of field spectrum analysis is to identify radionuclides present in the spectrum, to determine photon fluence rates for each significant peak and, if the transfer function is known, to calculate activity of each radionuclide. Fig. 5 presents the general algorithm of field spectrum analysis. The following chapters briefly present the principle of each of these modules.

A. Input Data

The minimum data required from the user by the sIGAle code for field spectrum analysis are the spectrum in ASCII format (including acquisition time), the name of the calibration file (result of standard spectrum analysis), the date of the spectrum measurement and the pre-defined radionuclide library to be used. Two radionuclide libraries are proposed, the first one for spectra of type "reactor", containing twenty radionuclides, and the second one for spectra of type "reprocessing", with nine radionuclides. The user can make his own library with one or more radionuclides.

Throughout spectrum processing, the software uses a database containing all the necessary information relative to the energy, the intensity and the natural width of all the X-ray and gamma lines of the radionuclides sought by the sIGAle code. This general nuclear and atomic database contains about three hundred radionuclides and elements. It has been created from LARA database. In the following process, the sIGAle code works with a reduced atomic and nuclear database produced from the radionuclide library.

Optional data may be specified by the user. If necessary, the user can introduce a rough energy calibration if it is too different from energy calibration defined by standard spectra. If experimental configuration contains a screen, the user can specify its type (among water, concrete and lead) and its thickness. To finish, the user can provide a transfer function so that the code calculates activities.

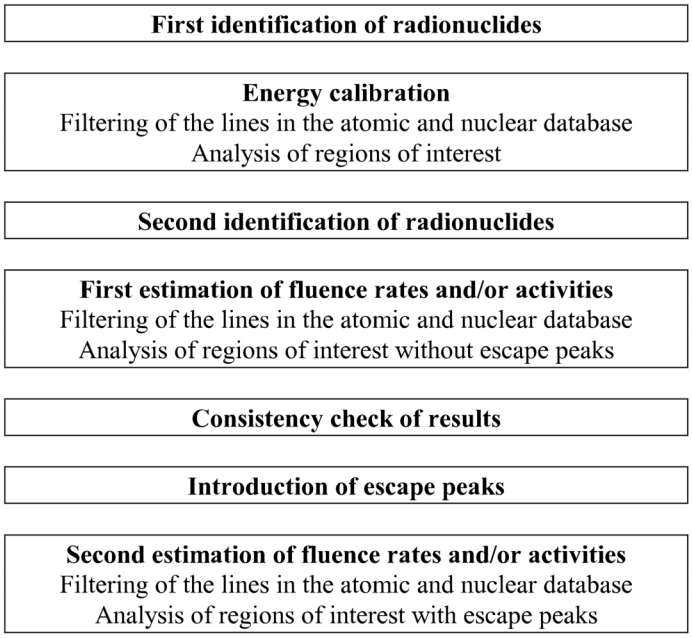


Fig. 5. General algorithm of field spectrum analysis.

B. Identification of Radionuclides

This identification of radionuclides is carried out twice, the second being more reliable because it is done after the energy calibration sequence.

First, it consists in a peak search in the spectrum. This stage leads to a list of peak energies found in the spectrum. Then the found peaks are associated with the lines contained in the atomic and nuclear database of the sIGAle software. A calculation of presence probability for each radionuclide according to the lines found for the different radionuclides and their intensity, weighted by efficiency determined with standard spectra, is finally performed on the basis of the determined associations. The radionuclide list used in the following process is composed of the radionuclides found by this module and the ones imposed by the user.

Fig. 6 presents results of peak search for a field spectrum which contains ^{60}Co and ^{51}Cr radionuclides. The sIGAle code found several peaks. Some of them are associated to ^{60}Co (1173 and 1332 keV) and to ^{51}Cr (320 keV). It found also Pb fluorescence peaks (74 and 84 keV), ^{60}Co backscattering (213 keV) and double escape peak of 1332 keV peak (310 keV).

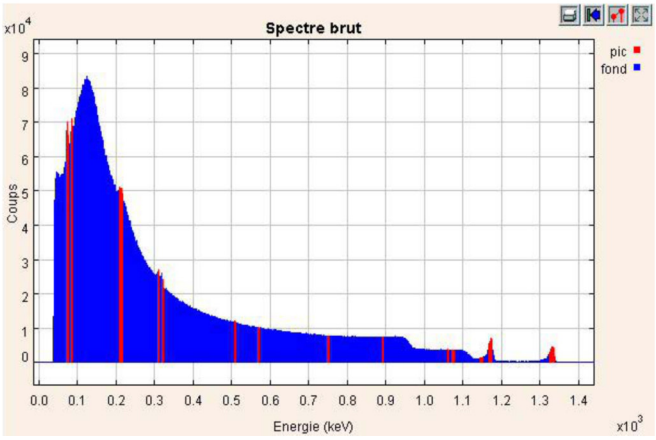


Fig. 6. Example of peak search. The red areas indicate the peaks found by the algorithm.

C. *Nuclear and atomic data filter module*

At the beginning of each of the following stages (energy calibration and determination of fluence rates and activities), the atomic and nuclear database is automatically filtered to only retain the lines considered as the most pertinent a priori, which are grouped in theoretical regions of interest, separated by distances depending on the spectrum resolution.

D. *Energy Calibration*

Energy calibration is the only calibration done for field spectra. All the other calibrations (resolution, tail parameters…) come from standard spectrum analysis.

E. *Photon fluence Rates and Activities*

Once the spectrum is properly calibrated and the list of radionuclides potentially present in the spectrum is established, the sIGAle code can carry out the estimation of peak areas to calculate fluence rates and/or activities.

Peak areas are estimated from deconvolutions. The fitting of each region is done by a sequential quadratic programming method, enabling numerous variables and constraints to be taken into consideration (Fig. 7). Deconvolutions thus include constraints between peak heights when they relate either to the same radionuclide or to two radionuclides in equilibrium.

Analyses of regions (and fluence rates or activities) are carried out twice, the first one without escape peaks and the second one with escape peaks. So, fluence rates or activities are calculated from these formulas:

$$\Phi(E) = \frac{S(E)}{t.K(E)} \qquad (7)$$

$$Act = \frac{\Phi(E)}{I(E).FT(E)}$$

$\Phi(E)$ is the photon fluence rate (cm^{-1}.cm^{-2}). $S(E)$ is the measured net area of the peak of energy E, t is acquisition time (s), $K(E)$ is the detector efficiency and $I(E)$ is the probability of photon emission of radionuclide at energy E. $FT(E)$ is the transfer function, given by the user and generally calculated from the MERCURE code.

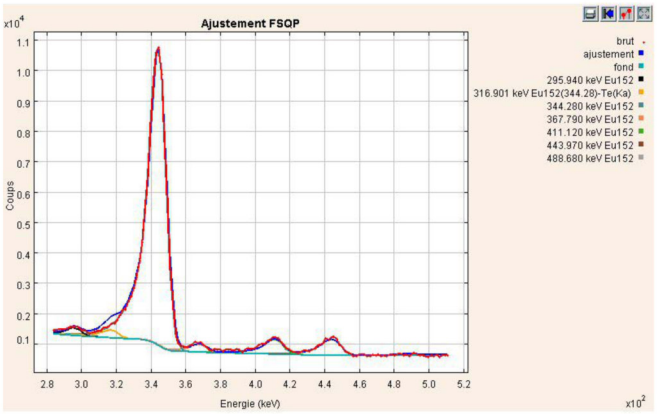


Fig. 7. Example of region of interest with several peaks of the same radionuclide ^{152}Eu. Constraints are applied.

F. *Consistency Check*

Consistency check is applied to first results obtained from analysis without escape peaks. This test is a comparison between the fitted areas in the regions of interest and the corresponding areas calculated from the detector efficiency $K(E)$ and the intensity $I(E)$ of the lines at energy E. This comparison is carried out individually line by line as well as globally through the sum of the fitted areas and the sum of the recalculated areas. If the difference is too high (>60%), the radionuclide is eliminated before the analysis with escape peaks.

G. *Output Files*

The application is integrated with a web browser, providing a very user-friendly interface. All the processing results are displayed in a highly-detailed manner in HTML pages.

IV. Experimental Results

Experiments have been carried out with standard sources in well-known experimental conditions. First tests have been performed and have shown that the quality of the standard spectra is primordial. The choice of the adequate standard radionuclides and the control of the experimental setup are also major points. For this latter point, an experimental bench has been designed and has been used to study the behaviour of the software according to several parameters.

Four types of experiments have been conducted:

- detection test: the counting time is gradually decreased in order to see the behaviour of the code at low statistics.
- SNR (signal to noise ratio) test: a second "disturbing" source is moved gradually closer from the detector, in order to evaluate the effect on the result for the first radionuclide.
- screen test: a screen of increasing thickness is placed between the source and the probe, in particular to estimate the impact on the radionuclide identification.
- volume test: several detectors with different volumes have been used to test the code.

Analysis of these spectra has been carried out with the sIGAle code and with manual method. The manual method consists in the analysis of the spectrum by the user, who marks manually the regions of interest using classic spectrometry software (GENIE 2000). The results of both methods are compared to the theoretical fluence rates. The theoretical fluence rates are calculated from the following formula:

$$\Phi_{thor.}(E) = \frac{Act.I(E)}{4.\pi.d^2} e^{-\rho(\mu/\rho).x} \quad (8)$$

Act is activity (Bq). *I(E)* is the probability of photon emission of radionuclide at energy *E*. *d* is the source-detector distance (cm) and *x* is the thickness (cm) of the screen put between source and detector. ρ is the screen density (g.cm^{-3}) and μ/ρ is the mass attenuation coefficient at energy *E* (cm^2.g^{-1}). This formula is correct only for point sources.

For all these tests, relative differences between fluence rates provided by the sIGAle code (or manual method) and theoretical fluence rates are presented to compare the different methods:

$$Fluence\ error = \frac{\Phi_{sIGAle\ (or\ manual)} - \Phi_{theor.}}{\Phi_{theor.}} \quad (9)$$

Evaluation of efficiency *K(E)* and parameters calibration have been carried out using standard point source of ^{152}Eu. Analyses of field spectra have been done with "reactor" library.

A. *Detection test*

The measurements have been performed with a 20 mm^3 probe. Energy resolution of this detector is 7 keV at 344 keV. Twenty-one field spectra have been measured with ^{152}Eu and eight with ^{137}Cs. For ^{152}Eu spectra, we look at the 121 keV peak for eight spectra, the 244 keV peak for six spectra and the 778 keV peak for the seven others. Automatic identification of radionuclides has found right radionuclide for twenty-seven spectra. For two spectra, module has not found any peak due to very bad statistics (around 10 counts in the largest peak). sIGAle results and manual results of fluence rates are compared to theoretical fluence rates and resulting relative errors are presented in Fig. 8 and Fig. 9.

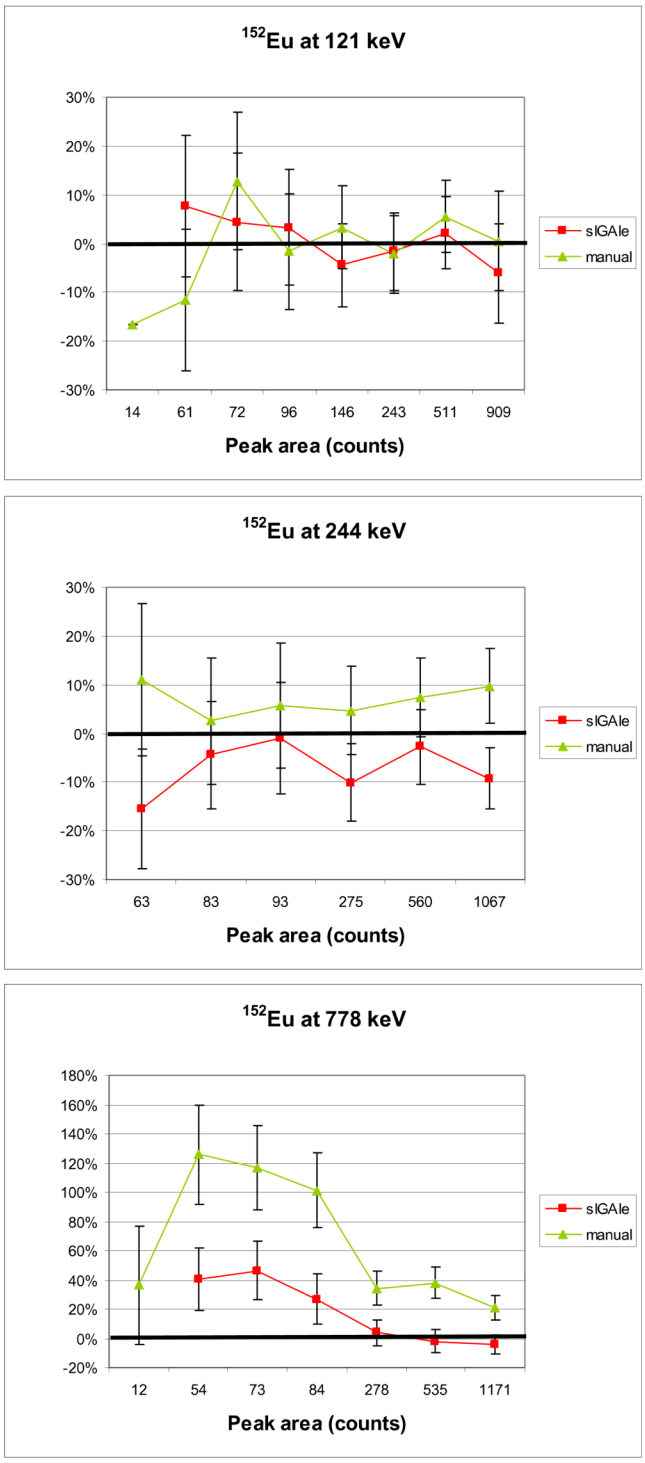


Fig. 8. Detection test. Comparison of sIGAle and manual fluence rates with theoretical fluence rates for ^{152}Eu peaks.

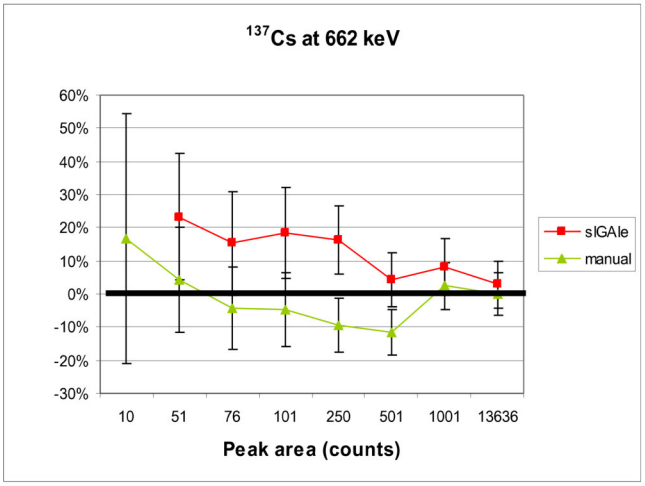


Fig. 9: Detection test. Comparison of sIGAle and manual fluence rates with theoretical fluence rates for ^{137}Cs peak

Manual method provides better results for the ^{137}Cs peak at 662 keV and the sIGAle code gives better results for the ^{152}Eu peak at 778 keV. For other ^{152}Eu peaks (at 121 keV and at 244 keV), sIGAle results and manual results are similar and relative errors (compared to the theoretical fluence rates) are under 10%. Better results could be obtained for 662 keV with the sIGAle code with a standard spectrum of ^{60}Co + ^{137}Cs instead of ^{152}Eu.

B. SNR test

The measurements have been performed with a 20 mm^3 probe. Field spectra are ^{152}Eu spectra. Energy resolution is 3.3 keV at 344 keV. This test consists in degrading signal to noise ratio for field spectra. To do this, an additional source of ^{137}Cs was used to raise Compton scattering in the spectrum. The ^{137}Cs was placed at variable positions to create different signal to noise ratios: the more the ^{137}Cs is close to detector, the more the signal to noise ratio is weak.

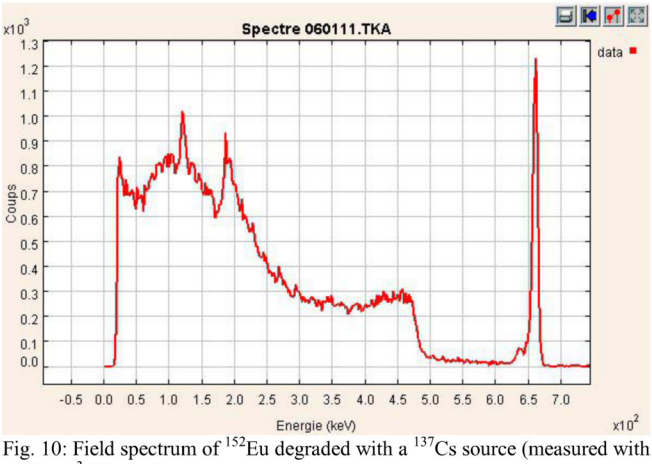
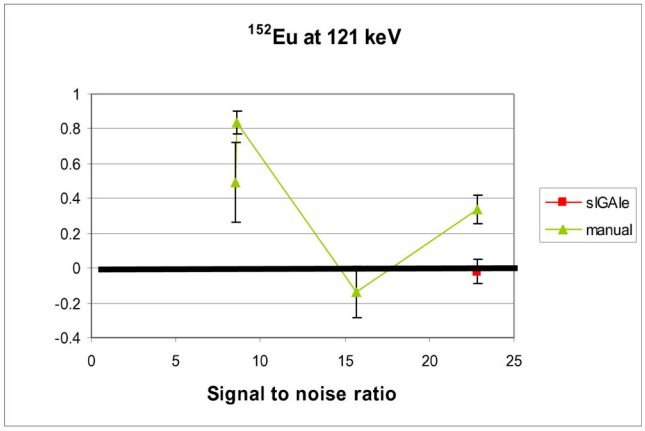


Fig. 10: Field spectrum of ^{152}Eu degraded with a ^{137}Cs source (measured with a 20 mm^3 detector).



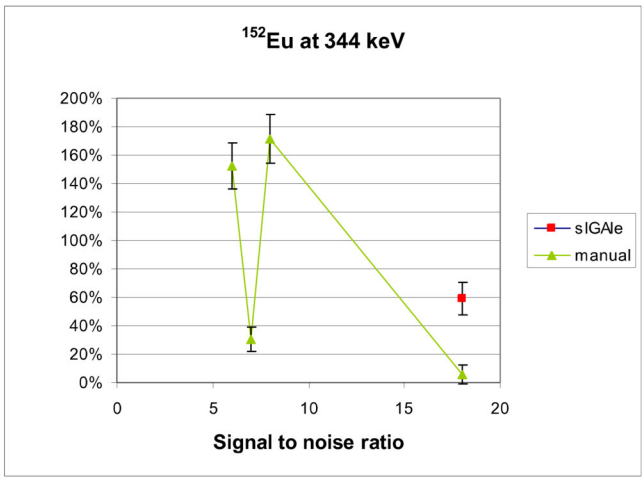
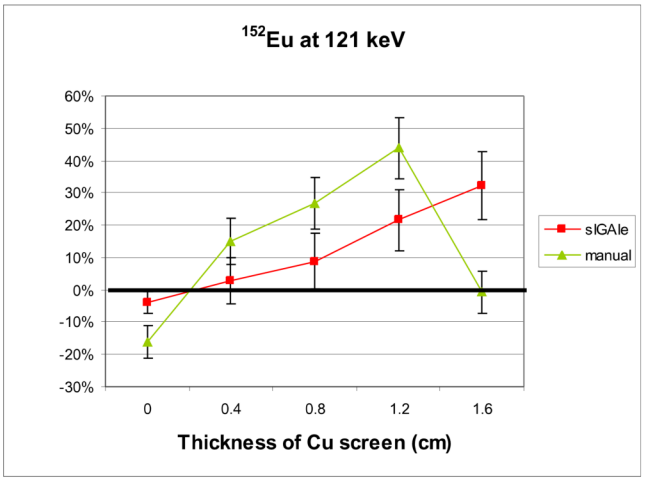


Fig. 11. SNR test. Comparison of sIGAle and manual fluence rates with theoretical fluence rates for ^{152}Eu peaks.

Eight spectra have been measured in these conditions. We look at the 121 keV peak for four spectra and the 344 keV peak for the four others. Fig. 10 is an example of spectrum in which we try to measure the peak at 344 keV (the signal to noise ratio is worth 8 in this case). Two difficulties are met by sIGAle code in SNR test. The first one concerns the radionuclide identification. When the software finds ^{152}Eu, it also finds often ^{154}Eu, which disturbs analysis. So, to obtain results of fluence rates for ^{152}Eu, a new library without ^{154}Eu has been created. Results (Fig. 11) have been obtained with this modification. The second difficulty concerns the signal to noise ratio. By default, the sIGAle code needs quite good ratios. The graphs show that the sIGAle code provides results for only two spectra against eight for manual method. Several processing failures occurred due to the too low signal to noise ratios.

C. Screen Test

The experimental device is the same as the one used for detection test, with the same standard spectrum. Five field spectra have been measured with ^{152}Eu. Four copper screens with different known thicknesses have been successively placed between source and detector.



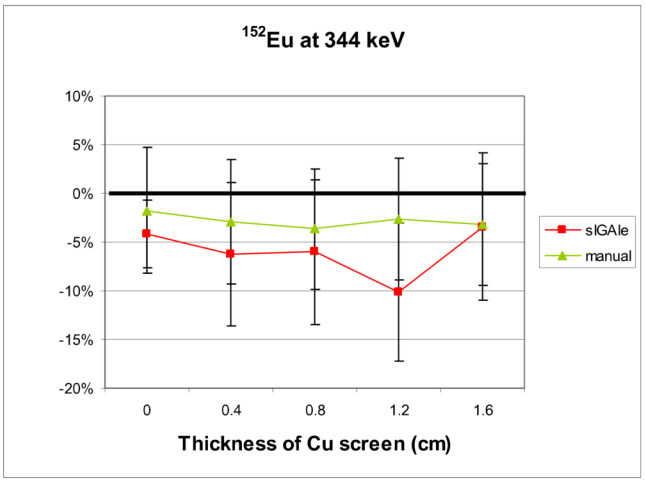


Fig. 12. Screen test. Comparison of sIGAle and manual fluence rates with theoretical fluence rates for ^{152}Eu peaks.

To do this test, the user did not supply information to the sIGAle code concerning screens. However, whatever screen was put, the sIGAle code finds ^{152}Eu (and sometimes ^{154}Eu). Indeed, the 121 keV peak was never totally removed by screen. Fig. 12 shows results of fluence rates for different thicknesses of screen. The fluence rates found for the peaks at 121 keV and at 344 keV are quite good.

D. Volume Test

The measurements have been performed with various volume detectors (5, 60, 500 and 1500 mm^3). Energy resolutions of these detectors vary between 6 and 20 keV at 344 keV. Field spectra are ^{137}Cs spectra. Each source was counted until at least 1000 counts were observed in the peak of interest.

Fig. 13 shows results of fluence rates for different detector volumes. The sIGAle code gives better results than manual method for detectors with large volumes.

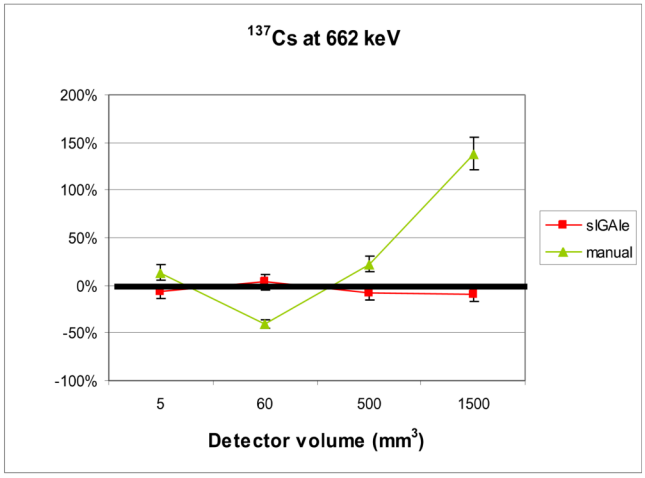


Fig. 13. Volume test. Comparison of sIGAle and manual fluence rates with theoretical fluence rates for ^{137}Cs peak.

V. Conclusion

The sIGAle code is an automatic tool of CdZnTe spectrum analysis. Its main functionalities are:

- creation of specific radionuclide libraries,
- automatic calibration based on the analysis of one or several standard spectra
- analysis of a field spectrum, including the identification of radionuclides present in the spectrum, the peak deconvolutions, the calculation of fluence rates for each peak and the determination of the apparent activities for these radionuclides if a transfer function has been previously provided.

The various tests carried out with known sources and in controlled experimental conditions allow to bring several preliminary conclusions on the behaviour of the sIGAle code by comparison with the manual spectrum treatment used currently by the CEA DEN. First, it seems that the automated area processing realized by the sIGAle code is more reproducible and more accurate at the same time than area processing made in a manual way with a classic gamma spectrometry software. This is illustrated by detection test and volume test. However, the sIGAle code is less competitive for degraded spectra when signal to noise ratio is weak (SNR test). To finish, the sIGAle code gives good results with Cu screen (until 1.6 cm), even without information concerning the latter. It would not be necessarily true for other screens and/or higher thicknesses.

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