A new computer code for the determination of the isotope composition of actinides by X- and gamma ray spectrometry and its applications

Anne-Catherine Simon, Jean-Pierre Both, Isabelle Espagnon, Jean Lefevre,
Vincent Picaud, Alain Pluquet

HAL Id: cea-01682396

https://cea.hal.science/cea-01682396

Submitted on 4 Apr 2023

A NEW COMPUTER CODE FOR THE DETERMINATION OF THE ISOTOPE COMPOSITION OF ACTINIDES BY X- AND GAMMA RAY SPECTROMETRY AND ITS APPLICATIONS

Anne Catherine SIMON (CEA), France, anne-catherine.simon@cea.fr - **Jean-Pierre BOTH (CEA),** France, jean-pierre.both@cea.fr - **Isabelle ESPAGNON (CEA),** France, isabelle.espagnon@cea.fr – **Jean LEFEVRE (CEA),** France, jean.lefevre@cea.fr – **Vincent PICAUD (CEA),** France, vincent.picaud@cea.fr – **Alain PLUQUET (CEA),** France, alain.pluquet@cea.fr

Since 1998 CEA and COGEMA have been collaborating on new tools for isotope analysis by X- and gamma ray spectrometry with germanium detectors. The research program started with the determination of plutonium isotope composition, knowledge of which is essential in several branches of the nuclear industry. The newly developed application is characterised by an original approach and innovative gamma ray spectrometry techniques featuring:

- a generic design (for example, no distinction is made between the analysis of the different isotopes of plutonium),

- the systematic and automatic use of an atomic and nuclear database,

- an advanced constrained-optimisation algorithm expressing physical knowledge in terms of constraints thus allowing robust analysis of very complex regions of interest,

- the simultaneous determination of global efficiency curve and isotope ratios.

For a user, this application takes form of an entirely automatic tool that accepts spectra corresponding to any experimental setup (gain, energy range, resolution, screen etc.), without any detailed data on the setup being necessary. Moreover the user can control all intermediate results by the way of a user-friendly interface through an Internet browser.

As the performance of the application proved satisfactory for plutonium isotopes in terms of robustness and precision, it was decided to extend it first to uranium enrichment, then to uranium and plutonium mixtures, and more generally to any actinide and contaminant mixtures. The development and evolution of the enhanced tool proceeded smoothly due to its initial generic design. In addition, a specific peak-location module was developed. This can recognise isotopes in a spectrum, and orientate the treatment to different classes such as "plutonium spectrum", "uranium spectrum", or "mixture spectrum". The application, called IGA, can now recognise and analyse a spectrum with actinides such as plutonium, uranium, neptunium and curium, together or alone, and in the presence of contaminants.

The same set of algorithms has been also applied to another purpose, the characterisation of waste packages. A dedicated tool, called IGA/Sandra, has been developed for the CEA centre at Fontenay-aux-Roses. This tool is able to compute not only isotopic ratios and relative efficiency but also absolute activities, by normalisation with measured efficiency.

Another domain of application of IGA is analysis of spectra from CdTe, CZT and NaI detectors. The peak-location module already gives good results, since it is able to adapt itself to each type of detector and to find peaks in this type of spectrum.

1. INTRODUCTION

The IGP code (a French acronym standing for plutonium gamma isotopy) was initially developed by CEA in collaboration with COGEMA to meet the need for a new computer code for determining the isotope composition of plutonium.

The method employed is based on the use of the energy spectrum consisting of X and gamma rays emitted by the different radionuclides contained in the material to be analysed. In most of the earlier methods, measurement is based on the use of the part of the spectrum beyond 120 keV, referred to as the gamma region [1], or focuses on the part of the spectrum between 90 and 120 keV, referred to as the XK region, which is hard to analyse but can provide much information [2]. Unlike the existing applications, for IGP the

decision was made at the outset to take the broadest approach possible and not, for instance, to distinguish between specific regions, peaks or isotopes. All the regions of the spectrum are accordingly analysed in a similar manner, including the most complex ones, without any special processing for the different isotopes involved in the problem to be solved. This has not, however, been achieved at the expense of the physics of the problem, on the contrary all the physical data available are systematically used and the methods applied always feature the conceptualisation and intellectual approach that an expert might adopt. This has made it possible to subsequently extend its use to uranium, to mixtures of uranium and plutonium and, finally, to other actinides, possibly in the presence of contaminants, and create the present application, named IGA [3].

The approach was used again to develop another application, named IGA/Sandra, for the measurement of the activity of waste packages at the Sandra facility at the CEA centre at Fontenay-aux-Roses, featuring similar methods and using the same calculation core. The two tools have evolved in parallel, each benefiting from the improvements made to the other.

The general principles of the IGA computer code are first described, then the particularities of IGA/Sandra.

2. MEASUREMENT OF THE ISOTOPE COMPOSITION OF ACTINIDES

2.1. General

2.1.1. Principle

The essential problem IGA has to solve can be expressed as follows:

$$S_j \propto \sum_i m_i D_{ij} \varepsilon_j \tag{1}$$

where
- S_j is the **measured** net area of the peak of energy E_j,
- m_i is the **unknown** mass of isotope i,
- D_{ij} contains the atomic or nuclear data **known** for isotope i at energy E_j:

$$D_{ij} = \frac{I_{ij}}{T_i A_i} \tag{2}$$

where I_{ij} is the probability of photon emission of isotope i at energy E_j, while T_i and A_i are its half life and its mass number respectively,

- ε_j is the unknown global efficiency of detection at energy E_j, including phenomena such as self attenuation of photons in the material measured, their attenuation in screens, or the efficiency of the detector,

- the sum is for the isotopes i present in the sample as it is possible for a number of them to contribute to the same peak at energy E_j.

Unlike in the case of activity measurement, **absolute** detection efficiency in not supplied to the code, and cannot be determined by it intrinsically. Therefore, only **relative** variables are accessible, whether for efficiency ε_j or sought masses m_i, which are determined relative to an isotope taken for reference. This implies the following equation:

$$S_j = \sum_i f_i D_{ij} \varepsilon_{rj} \tag{3}$$

where
- f_i is the ratio of isotope i relative to a reference isotope,

- ε_{rj} is the overall relative efficiency of detection, for the same reference isotope, at energy E_j.

First, the global approach used with the IGA code to obtain these ratios is described, then the general principles of the main modules are briefly explained.

2.1.2. Main IGA algorithm

Figure 1 shows the general sequence of the processing of a spectrum by IGA.
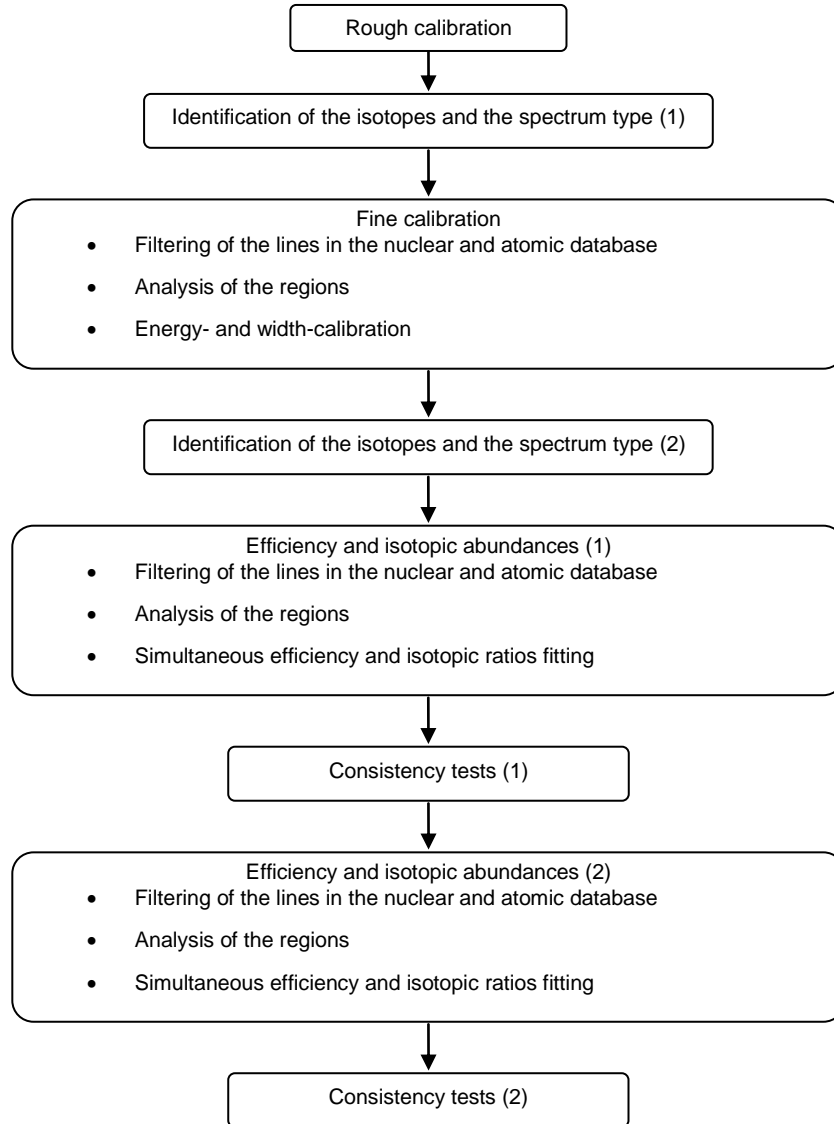

Figure 1 – IGA general algorithm

Actual isotope composition determination is preceded by a preliminary stage in which the radionuclides present in the spectrum are identified in order to select the isotopes that IGA will use, and to determine the type or "nature" of the spectrum in question, i.e. whether it corresponds to plutonium, uranium etc. This isotope identification stage is carried out twice, the second being more reliable because it is carried out after a fine calibration sequence.

This fine calibration sequence that follows the first isotope and spectrum identification step is highly important, involving spectrum shape and energy self-calibration. The next phase, which is also carried out twice, corresponds to the fundamental role of the application as it consists of calculation of the relative efficiency and of the isotopic abundances sought. As can be seen in Figure 1, these three stages have common modules, as those for filtering the nuclear and atomic database and analysing the regions, and ones that are specific, as the channel-energy and resolution-energy fitting for the self-calibration stage, and the determination of efficiency and isotope composition for the other two stages.

Furthermore, consistency checks of different types are carried out after each isotope determination.

2.1.3. Data and properties

The code requires very little data input by the user to carry out the processing, that necessary being restricted to the energy of the first channel, the energy of the last channel and the resolution relative to a user-selected energy. Moreover, no special experiment setup is required, and the user remains completely free to choose the gain, the resolution and the energy range, even though the results do of course depend on the quality of the spectrum acquired.

Furthermore, throughout spectrum processing, the application uses a single 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 IGA code. These data could, in principle, be selected by the user from any of the existing libraries (for instance [4], [5], [6] or [7]). However, considerable discrepancies can at times be observed between in energy data and above all the line intensities of the different tables, and some of which may in addition prove to be incomplete, particularly for X-rays. The repercussions of such situations can be substantial, and it has been found to be preferable to use a specific database that is as comprehensive, consistent and accurate as possible. This general nuclear and atomic database covers some 4000 lines, relating to around 80 radionuclides and elements. Of these 80 radionuclides and elements, about 40 have been placed in a restricted software working library. If necessary, the user can however modify this isotope list to create its own restricted library.

Lastly, the application is integrated with a web browser, providing a very user-friendly interface. All the processing results, and those of all the intermediate calculation, are displayed in a highly-detailed manner in HTML pages. In commercial use, a more conventional interface can, of course, be used and the results can be output in more summarised form as text files.

2.2. The modules

2.2.1. Spectrum type and isotope identification module

This module has some particularities that enable it to automatically adapt itself to different experiment configurations. It first seeks to locate peaks in the spectrum, and can automatically account for spectrum statistics and resolution. It then associates the peaks that it has found in the spectrum with X-ray and gamma lines recorded in the restricted database mentioned above. A calculation of the probability of the presence of each of the radionuclides in the database is then performed on the basis of the associations made. The isotopes found to be present then allow classifying the spectrum in one of the four categories currently considered, as follows:

- the Pu category, consisting of the isotopes of plutonium (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu) plus the various isotopes and elements commonly encountered in this type of spectrum (^{241}Am, ^{237}U, Pu, Pb etc),
- the U family, consisting, in a similar manner, of the isotopes of uranium (^{234}U, ^{235}U, ^{236}U and ^{238}U) and their daughters (^{231}Th, ^{234}Th etc), plus a few others (Cd, Pb, U etc),
- the UPu family, corresponding to the sum of the two preceding ones,
- the "unknown" family, for spectra containing neither plutonium nor uranium.

The list of the radionuclides used in the subsequent processing is thus established, in addition to the radionuclides found in the spectrum, of all the different isotopes associated with the nature of the spectrum determined, to which radionuclides can be added by the user.

By elimination of radionuclides, this stage enables an initial selection to be made of lines in the nuclear and atomic database so that the subsequent processing can be simplified by limiting the unknowns to those that are pertinent. Other ways of selecting lines from the database are then applied by the filter module, as described below.

2.2.2. Nuclear and atomic data filter module

At the beginning of each of the following stages (fine calibration and the two stages consisting of simultaneous determination of the efficiency and the isotopic ratios), the database is automatically filtered to only retain the lines considered *a priori* to be the most pertinent, grouped in regions of interest. To do so, a succession of filters is applied:

- elimination of the lines that relate to the radionuclides not selected by the spectrum type and isotope identification module, as described above,
- elimination of the lines of which the weighted intensity is low in absolute terms,
- elimination of the lines of low weighted intensity relative to other lines of the same energy,
- elimination of the lines of low weighted intensity relative to others in the same region, applying a double criterion of distance and relative height.

Finally, the remaining lines are grouped in regions, separated by distances depending on the spectrum resolution. In the case of the fine calibration stage, the regions containing more than two distinct energy lines are in addition deleted to avoid disturbing the self-calibration by the analysis of excessively complex regions at a stage when little is known about the spectrum.

This process of selection of lines and determination of regions of interest depends on the following factors conjointly:

- strong presuppositions such as default isotopic abundances,
- the context, i.e. the current stage of processing (fine calibration stage, stages of determination of the isotopic ratios and the efficiency) as both the needs and the available data differ accordingly,
- the spectrum itself, via its energy range and its resolution.

2.2.3. Region analysis module

The purpose of the region analysis module is to adjust the content of the channels of each of the regions of interest selected by the database filter module beforehand, in order to determine the variables needed in the current stage (heights, positions, standard deviations etc.). This operation is performed in three steps:

- Pre-fitting tests: these consist in checking that the peak is present in the region to be analysed, and calculation of the limit of detection after subtraction of the estimated background noise using the orthogonal polynomial method [8, 9]. If these two tests are not passed, the region is not adjusted.
- Fitting of the counting signal in the region.
- Pre-fitting tests: these consist in calculation of criteria of pertinence of the fitted peaks (limit of detection and χ^2 fit quality) for selection of the peaks that can subsequently be used.

The content of the channels is fitted by a model that can allow for both a background and the peaks corresponding to the lines selected for the region. The peaks are fitted by simple models that are Gaussian for the gamma peaks and Voigt functions calculated by fast Fourier transform for the X-ray peaks. The background for the entire region complies with a model consisting of a polynomial function, to which is added a "step" term under each of the peaks in the region.

The complete fitting of the region is carried out by a sequential quadratic programming method, enabling numerous variables and constraints to be taken into consideration. The latter include constraints between peak heights, when they relate to the same isotope or are linked by an equilibrium relationship. In the same manner, the background of a region can be fitted in accordance with certain rules. Finally, an estimation of the efficiency can, depending on the current step of the processing, be taken into account in the fitting of a region, thus adding constraints concerning the parameters to be determined.

Depending on the current stage, all or only some of the parameters of the model are fitted. Thus, in the self-calibration stage, all the parameters are free, the parameters of interest in this phase being the widths of the peaks and their positions in channels. In the subsequent stages, these two parameters remain fixed and the parameter of interest is then peak height. Any region of interest, even the previously mentioned XK region which is particularly dense and difficult to analyse, is therefore deconvoluted. In certain cases, if the peaks of

a region are too close and not sufficiently constrained to be satisfactorily fitted, the regions are automatically coupled to provide additional information.

2.2.4. Shape and energy calibration curves

The final goal of the self-calibration stage is to provide channel-energy and resolution-energy relationships. Owing to the special mode of selection of the lines as a function of the radionuclides effectively present in the spectrum, the fine calibration ensures high adaptability to the spectrum analysed and has proved to be unaffected by the presence or the absence of a particular isotope.

A second-degree polynomial relationship is then determined between the positions in channels of the peaks fitted in the analysis of the regions and their energies in the database. In the same manner, the parameters of a straight-line equation are calculated that links the variances resulting from the fitting of the peaks to the energies.

2.2.5. Simultaneous determination of efficiency and of isotopic ratios

The principle of this module is based on equation (3), in which both the isotopic ratios and the efficiency are unknown. The approach is then to describe the efficiency in two different ways, one given by the net areas measured in the spectrum by deconvolution of the previous regions involving the isotopic ratios sought, and the other driven by a parametric model as a function of the energy, which involves three terms, self-attenuation in the Pu and/or U matrix $\varepsilon_{self-att}(E)$, attenuation in a screen $\varepsilon_{att}(E)$, and a polynomial function approximately giving the efficiency of the detector $\varepsilon_{det}(E)$:

$$\varepsilon_i = \frac{S_i}{\sum_{j=1}^{nr} f_j D_{i,j}}$$

$$\varepsilon_i = f(P,E) = \varepsilon_{att}(E).\varepsilon_{self-att}(E).\varepsilon_{det}(E)$$

(4)

The difference between the two types of description is then globally minimised for all the peaks accepted, by fitting of unknown parameters isotopic abundances f_i and efficiency parameters P. In this optimisation, one of the isotopic ratio remains fixed and is thus used as a reference point for all the other isotopic ratios, as well as for the resulting relative efficiency curve.

This minimisation is, here again, established by a sequential quadratic programming method and also uses all the physical data available, as in the case of the equilibria between isotopes. The initial fitting values first consist of default values, then of values determined in the first estimation.

Figure 2, for instance, shows a global efficiency curve obtained by this type of analysis.
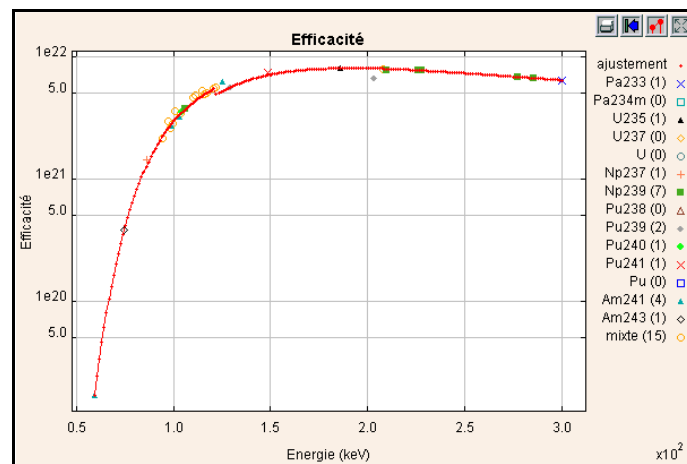

Figure 2 – Example of an efficiency curve obtained after optimisation
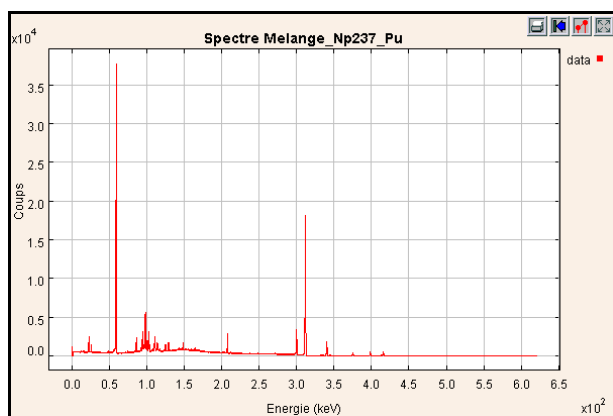
2.2.6. Consistency analysis

A number of different types of consistency tests are made on the results after each isotopy determination:

- Comparison of the energies of the peaks found in the spectrum with the energies of the lines analysed in all the regions; a peak thus found that is not taken into account in the processing can be representative of a radionuclide missing from the restricted database, which may be a sign of an error in the processing if some of the peaks of the radionuclide interfere significantly in the regions analysed.

- Comparison of the areas of fitted peaks of each radionuclide with the areas subsequently calculated on the basis of the relative efficiency and isotopic abundances results. A certain number of rules, such as, for instance, the absence of the main line of a radionuclide, make it possible at this level to identify a suspicious isotopic ratio result and to warn the user. Furthermore, this first-cycle analysis result affects the subsequent processing; among other things, the suspicious isotopic ratio is not re-used in the second cycle but is replaced with a isotopic ratio calculated from a detection limit.

- Comparison of the spectrum with the reconstructed spectrum based on the isotopic abundances results and the efficiency found, in order to locate and identify any differences between the two spectra.
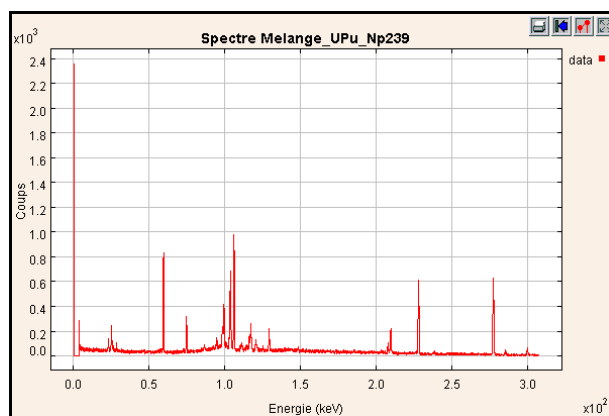
In addition to the degree of confidence given by the software itself in the results, these three types of tests provide the user with the ability to check the results supplied by the application in considerable depth.

2.3. Examples

Due to the generic and self-adjusting nature of its algorithms, the IGA application is currently capable of equally well handling the spectra of uranium, plutonium, or mixtures of uranium and plutonium, despite the presence of contaminants and other actinides, and even of analysing spectra from which both uranium and plutonium are absent. Examples of untypical spectra are shown in Figures 3a and 3b, for plutonium mixed with a substantial amount of ^{237}Np/^{233}Pa with an energy range of 0-600 keV and for a mixture of uranium and plutonium with a substantial amount of ^{243}Am/^{239}Np with an energy range of 0-300 keV respectively.

(a)   (b)

Figure 3 – Spectra (a) of plutonium with a substantial amount of ^{237}Np/^{233}Pa
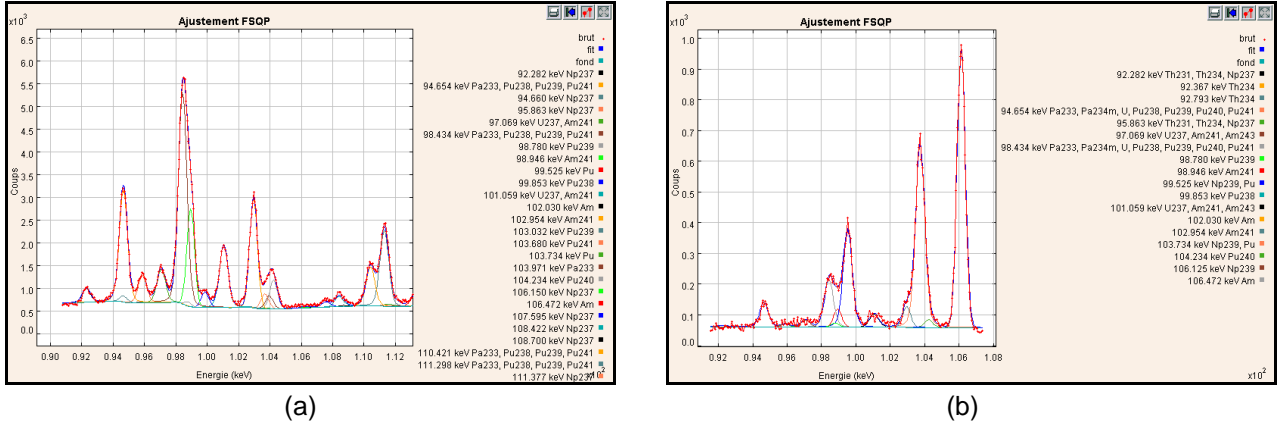and (b) of a mixture of uranium and plutonium with a substantial amount of ^{243}Am/^{239}Np

Figure 4 – Examples of deconvolution of a complex region by the region analysis module, for spectra (a) with Pu-[237]Np/[233]Pa and (b) with UPu-[243]Am/[239]Np

Figures 4a and 4b clearly show the adaptability of the IGA analysis module, with these two extremely different configurations. The application thus has the advantage of being very versatile, not only in terms of the experiment configuration (gain, energy range etc.) but also the radionuclides present in the spectra.

This original approach was accordingly extended to the development of the IGA/Sandra application, intended for measurement of the activity of waste packages, as described in the following section.

3. MEASUREMENT OF THE ACTIVITY OF WASTE PACKAGES

The IGA/Sandra application was specially developed for the Sandra facility at the CEA centre at Fontenay-aux-Roses with the purpose of measuring the activity of medium and low level waste prior to sending to the sites of the French national waste disposal agency ANDRA. In this case the conditions of the experiment were unfavourable, as a result of the count statistics frequently being low and the gain being high, although they had the advantage of being known and fixed. An example of a spectrum to be analysed is shown in Figure 5.
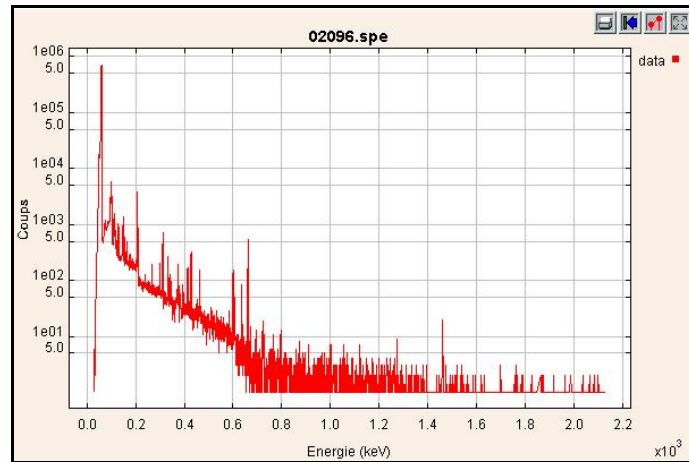

Figure 5 – Example of a waste drum spectrum

The principle of the IGA/Sandra application is to write activity $\mathcal{A}_i$ of an isotope i using its mass:

$$\mathcal{A}_i = \lambda_i N_i = \frac{\ln 2 . \mathcal{N} . m_i}{A_i . T_i} \tag{5}$$

where

- λ_i is the decay constant of isotope i ,

- N_i is the number of atoms of isotope i,
- $\mathcal{N}$ is the Avogadro number,
- m_i is the mass of isotope i,
- A_i is the molar mass of isotope i,
- T is the half life of isotope i in seconds.

The determination of masses is the goal of the spectrum analysis made by the IGA application, subject only to a factor of proportionality. The two issues are thus very similar, and it is for this reason that the IGA/Sandra application makes use of the great majority of the IGA functionality in its calculation modules. However, unlike in the case of the measurement of the isotope composition, in which only the relative proportions of the isotopes are sought, here an absolute measurement of the activity of the different radionuclides has to be made. Additional data, such as duration of acquisition and above all the absolute efficiency curve, are therefore necessary. The transition of the isotopic abundances into activity is therefore performed by normalisation of the relative efficiency curve determined by the IGA calculation kernel with the absolute efficiency curve supplied by the user. An example of normalisation is given in Figure 6.
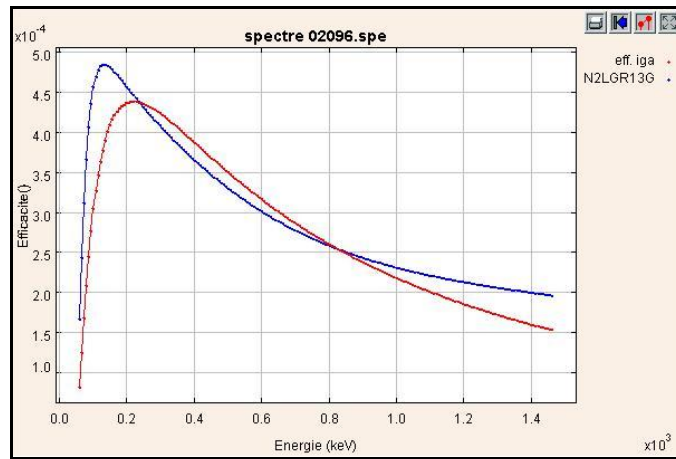

Figure 6 – Example of normalisation of the IGA relative efficiency curve
with the user's absolute efficiency curve

Figure 7 gives the general algorithm used by IGA/Sandra for determining the activity sought.
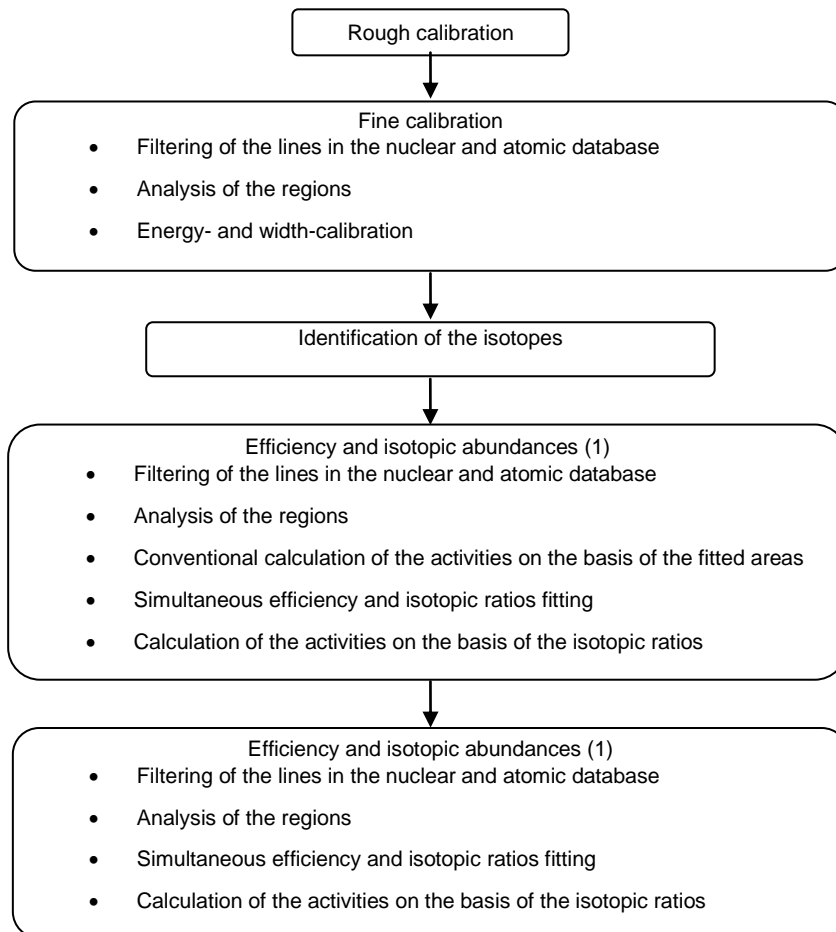
Figure 7 – IGA/Sandra general algorithm

This algorithm makes use of a number of modules that are common to the IGA application, such as:

- rough calibration,
- isotope identification,
- filtering of the nuclear and atomic database,
- analysis of the regions of interest,
- simultaneous determination of efficiency and isotopic ratios.

The common modules are however somewhat modified to adapt them to the role:

- a nuclear and atomic database specific to the requirements of the installation,
- database filtering in which efficiency can be taken into account from the start, using the absolute efficiency supplied by the user,
- filter thresholds directly adapted to the low statistics,
- fine calibration carried out on the basis of all the isotopes listed, and hence isotope identification in a single cycle,
- a simplified parametric model for the relative efficiency used in the phase of simultaneous fitting of the efficiency and the isotopic ratios,
- special rules for management of the equilibria making it possible to determine the radionuclides more particularly sought for.

What is special about the tool developed is that it not only calculates the activity in a new way from the overall isotopic abundances determined by IGA, but it can calculate it directly in a conventional manner from the areas of the fitted peaks and from the absolute efficiency supplied by the user:

$$\mathcal{A}_{ij} = \frac{S_{ij}}{t.I_{ij}.\varepsilon_j} \tag{6}$$

where

- $\mathcal{A}_{ij}$ is the activity in Bq of isotope i calculated at energy E_j,
- S_{ij} is the area of the line at energy E_j of isotope i,
- t is the spectrum acquisition time in seconds,
- $I_{i,j}$ is the intensity of the line at energy E_j of isotope i,
- ε_j is the absolute efficiency at energy E_j.
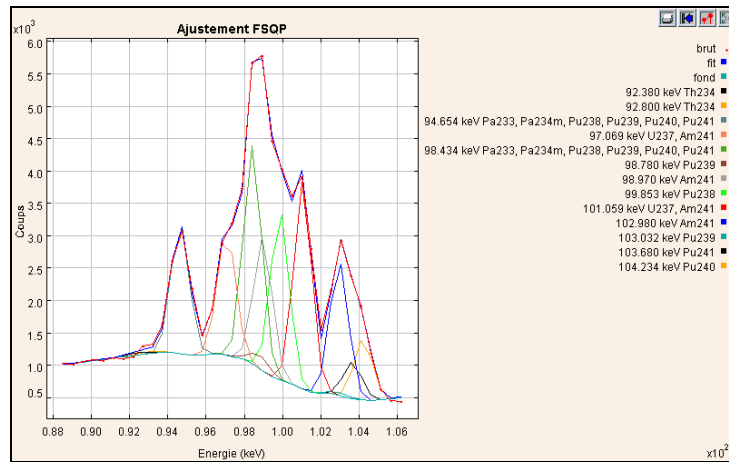
Figure 8 shows an example of a fitted region.

Figure 8 – Example of a fitted region in a waste package spectrum

The activity of each radionuclide is then calculated as the weighted mean of the different activities found for each of the lines of the radionuclide in question.

The user then has two sets of activity results of which he can evaluate the consistency using a browser-like interface that can show all the intermediate results. The so-called conventional method has the advantage of determining the activities in a similar way to that employed by the user while benefiting from far better region fitting. The IGA method, on the other hand, uses far more information than in the case of conventional analysis and, above all, imposes consistency of the results over the entire spectrum.

The IGA/Sandra application has been tested with some forty CEA Fontenay aux Roses waste packages spectra by comparing the results with those obtained by the operator's conventional expert appraisal method. The difficulty is that in this case no results from another method of measurement (with a calibration waste package or a waste package measured by destructive analysis) were available.

4. CONCLUSIONS AND OUTLOOK

The IGA application, with its innovative features, can analyse spectra corresponding to any actinides. Spectra for different configurations, such as plutonium in the presence of large amounts of [37]Np/[233]Pa or [243]Am/[239]Np, or even mixtures of different isotopes of plutonium, uranium, curium and neptunium (simulated spectra) could also be processed with the IGA application due to its generic nature. However, no in-depth study has yet been made of these new actinides; as in the case of plutonium and uranium, it may be necessary to determine special categories in order to make proper allowance for their particular features (high self-attenuation, self-fluorescence lines etc.).

The IGA/Sandra application developed for measuring the activity of waste packages has thus benefited from the general approach and the IGA calculation kernel, originally mainly intended for uranium and plutonium.

Conversely, the development work carried for this utilisation was re-used to supplement and improve IGA for other actinides and contingent contaminants. For instance, the development of consistency tests and the peak identification module have made the code more adaptable.

A new software tool intended for the measurement of the apparent activity of radionuclides by spectrometry based on the use of CdZnTe detectors is now being studied, and is already using the existing IGA code identification module.

Bibliographic references

[1]   J. Morel, M. Etcheverry, M. Vallée, "Détermination de la composition isotopique du plutonium par spectrométrie gamma, logiciel COMPOS", Note Technique CEA/LMRI/90/269 (1190).

[2]   R. Gunnink, "MGA : a gamma-ray spectrum analysis code for determining plutonium isotopic abundances", UCRL-LR-103220 Vol. 1 & 2, ISPO n° 317 (1990).

[3]   A. Pluquet, I. Espagnon, R. Junca, J. Morel, A.C. Simon. Détermination de la composition isotopique des actinides, Bulletin du BNM n° 123, Volume 2003-2, Journées de spectrométrie Gamma et X, Octobre 2002.

[4]   C. Morillon, M-M. Bé, Jacques Lamé…[et al.], "Bibliothèque des émissions gamma et alpha Nucléide-LARA", Commissariat à l'Energie Atomique, 91191 Gif-sur-Yvette, France, impr. CEA, Ed. 2000.

[5]   R. B. Firestone, "Table of Isotopes, eighth edition", Lawrence Berkeley National Laboratory, University of California, John Wiley & Sons, 1986.

[6]   ENSDF, http://ie.lbl.gov/ensdf.

[7]   E. Storm and H. I. Israël, "Photon cross sections from 1 keV to 100 MeV for elements Z = 1 to Z = 100", Nuclear data tables section A, vol. 7, pp. 565-681, June 1970.

[8]   S. Steenstrup. A simple procedure for fitting a background to a certain class of measured spectra. J. Appl. Cryst., 14 : 226, 1981

[9]   P. J.M. Van Espen and K. H.A. Janssens. Spectrum Evaluation, Handbook of X-ray Spectrometry. Marcel Dekker, New York, 1993.