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Abstract

The IGA code (a French acronym standing for actinides gamma isotopy) is a tool developed by the CEA LIST to determine the isotopic composition of plutonium and uranium, based on the automatic analysis of the γ/X spectrum emitted by a nuclear sample. Its main feature is its generic approach of the problem. As a consequence, the IGA code is a very flexible tool, because no particular experimental setup is imposed to the user for the acquisitions, in terms of energy range, gain, channel number or detector resolution. However, these experimental conditions can have an impact on the quality of the results. So, a study has been carried out to evaluate the effect of different experimental parameters on the IGA performances for plutonium spectra and quantify these performances. The study has been led according to three different approaches: analysis of IGA results on a whole spectrum database coming from French laboratories, analysis on artificially modified spectra and finally analysis of results on new real spectra acquired in the laboratory on certified samples.

Keywords

Gamma spectrometry, IGA code, isotopic composition, spectral analysis, X-ray spectrometry

A. Introduction

The knowledge of the actinide isotopic composition is a major issue in the field of nuclear industry. Several codes have been developed in this aim for many years. MGA and PC/FRAM are the most used [1]–[3]. The IGA code (a French acronym standing for actinide gamma isotopy) is a more recent tool developed by the CEA LIST in collaboration with AREVA NC to determine the isotopic composition of plutonium (Pu) and uranium (U), possibly in presence of fission products, activation products or other actinides [4]–[6]. It is now commercialized by ITECH INSTRUMENTS and CANBERRA and it is used in France by several laboratories of CEA, AREVA NC and IRSN. The principle is based on the automatic analysis of the γ/X spectrum emitted by the different radionuclides in a nuclear material and measured by a germanium detector.

Each peak in the spectrum is linked to the different isotopes by its position (that gives qualitative information) and its net area (that gives quantitative information). More precisely, the peak areas are linked to the isotope mass ratios via their nuclear data and via the relative detection efficiency. This is described by:

$$S_j = \sum_i f_i \frac{I_{ij}}{T_i A_i} \varepsilon_j \qquad (1)$$

with:

- S_j is the measured net area of the peak at the energy E_j
- f_i is the unknown mass fraction of the isotope i relative to a reference isotope

- I_{ij}, T_i *and* A_i are the known atomic or nuclear data for isotope i at energy E_j ; I_{ij} is the probability of photon emission of isotope i at energy E_j, T_i being its half-life and A_i its atomic mass
- ε_j is the unknown global detection efficiency at energy E_j, normalized in relation to the reference isotope.

The difficulty is that no information on the efficiency (the detector efficiency, the self-attenuation in the sample, as well as the attenuation in screens) is available at the beginning of the analysis. To determine the mass ratios of the different isotopes, the spectrum analysis proceeds as follows:

- identification of radionuclides and spectrum type (Pu, U, U Pu, or no U nor Pu) ;
- fine energy and resolution calibration ;
- estimate of mass ratios and efficiency (first pass) ;
- estimate of mass ratios and efficiency (second pass) ;
- consistency tests.

The originality of the IGA process lies in the following points: first, the initial stage of identification enables the code to work with all the radionuclides visible in the spectrum, including the presence of other unexpected radionuclides. That means that all the detected radionuclides will be considered and treated in the same way.

Second, at each of the three following stages, the general nuclear database specific to the IGA code, which contains all the X- and γ-rays of the considered isotopes, is automatically filtered and the selected lines automatically grouped into regions of interest, according to the spectrum itself (its resolution, its energy range), the detected radionuclides and the current analysis stage. For instance, for the calibration stage, only the regions of interest that do not contain more than two peaks are kept, whereas for the following stages, all the regions, even the more complex ones, are kept. The selected regions of interest will be then deconvolved to extract the parameters of interest for the following process (position and width of the peaks for the calibration stage, heights of the peaks for the other stages). An exemple of deconvolved region is given in Fig. 1.

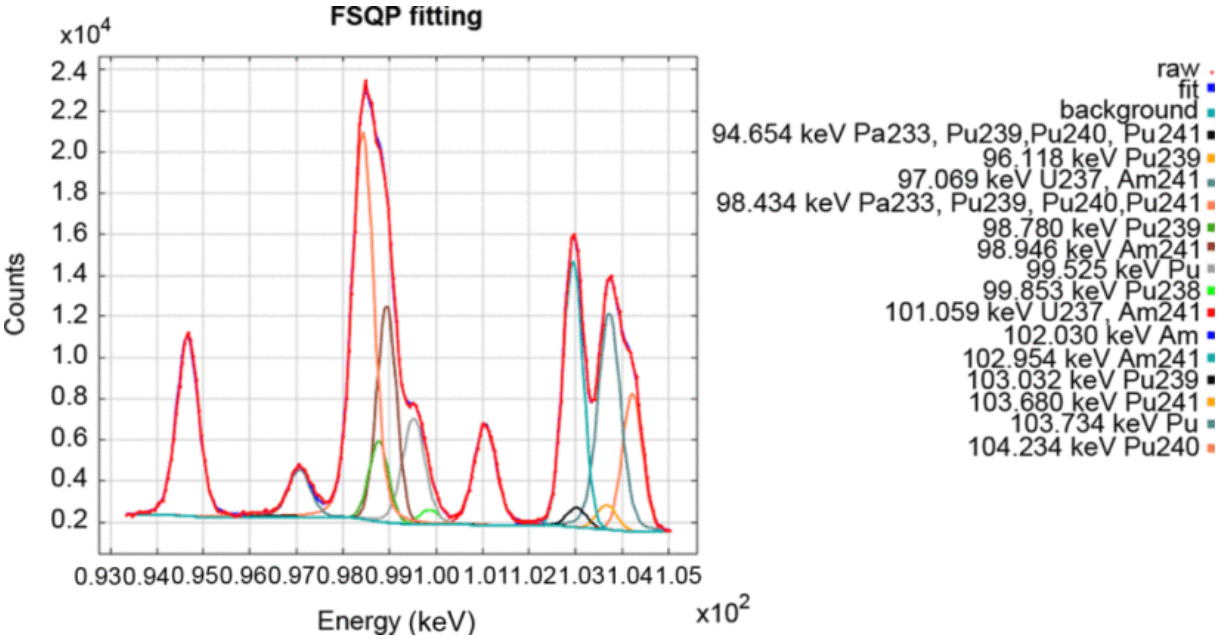

Fig. 1. Example of deconvolved region between 93 and 105 keV for a spectrum with 93.90% ^{239}Pu.

Third, mass ratios and efficiency are simultaneously determined thanks to a general optimization process that fits both the mass ratios and the parameters of an efficiency model in considering all the peaks of the spectrum at the same time.

More precisely, (1) is written under the new following form:

$$\frac{S_j}{\sum_i f_i \frac{I_{ij}}{T_i A_i}} - \varepsilon_j = 0 \qquad (2)$$

where the symbols have the same meaning as in (1). Efficiency will be described through a parametric model including self-absorption in Pu or U, absorption in a screen and a general model of detector efficiency. Equation (2) will be globally minimized on all the peaks of the spectrum at the same time. The unknown parameters to fit in this minimization process are both the mass ratios and the parameters of the efficiency model. Once a first estimate of the variables is determined, the whole step is carried out a second time to lead to the final isotopic composition.

Finally, the consistency tests enable the code to provide qualitative indications on the reliability of the results. They are also used to calculate uncertainties on the results, according to a very particular calculation mode, which is not based on statistical uncertainties, but try to consider the whole coherence of the results. Indeed, uncertainty for each isotope is obtained from the raw deviations between the measured area in the spectrum and area expected from the results. They are then propagated to the other isotopes, because final mass fraction of an isotope depends on the other ones.

For the user, the IGA software is very easy to use, because it needs few input data to run. It also benefits from a user-friendly web interface, configurable from an expert mode to a "black box" mode and includes a batch mode too. In particular, in the first mode, the user has access to all the intermediate results and graphs of the analysis at a very detailed level via the web interface.

But its major feature is its flexibility: as seen before, there is no distinction of treatment between the different isotopes or the different peaks in the spectrum. This enables the code to accept any type of spectrum, but also any experimental setup, without imposing particular gain, or energy range to the user. From this point of view, it can be said that the IGA code has an intermediate running mode between the other two codes MGA and PC/FRAM: MGA carries out an automatic spectrum analysis without any user specification by imposing experimental conditions. At the opposite, the PC/FRAM code runs with toolboxes, in which all the parameters (for example, the list of peaks to be considered by the code for each step) can be modified by a knowledgeable user. This gives the user the possibility to change the experimental setup. With the IGA code, the user has to enter a few data in order to give the code information about the energy range or the resolution. He can also modify a few parameters if really needed, like the ^{242}Pu calculation mode. But the code automatically determines itself which parameters are the most pertinent for the analysis (the lists of peaks to be used for instance).

But if no constraint is imposed for the experimental setup, the quality of the results can depend on these experimental conditions. The aim of the following study is to give indications on the performances of the code according to several experimental parameters, like energy range, gain, counting statistics or resolution.

For this study, three types of approach have been successively followed:

- global analysis of the results on a general Pu spectrum database
- evaluation of the results on reduced spectrum databases, for which parameters are modified artificially
- evaluation of the results on new real spectra acquired on certified samples, whose parameters are modified experimentally.

B. Global Study on a Large Spectrum Database

The first approach consisted in globally considering all the available Pu spectra (about 1150 spectra), coming from different laboratories of CEA, AREVA or IRSN, to evaluate the effect of some experimental parameters on the results. First, the studied parameters are the energy range, the gain and the counting statistics. For each isotope of interest (^{238}Pu to ^{241}Pu and ^{241}Am; ^{242}Pu is never considered because it is not determined from the spectrum), the criterion on the results is the relative difference between the mass ratio obtained by the IGA code and the mass ratio of reference. Figs. 2 and 3 illustrate the ^{239}Pu results versus gain and counting statistics. Results versus energy range have the same aspect as for gain, so they are not displayed here.

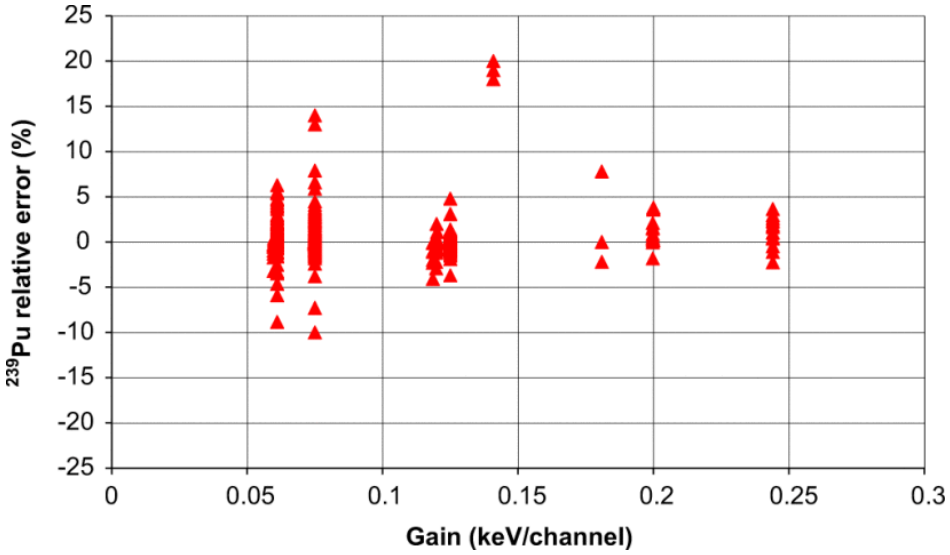

Fig. 2. Relative errors between the ^{239}Pu mass ratio obtained by IGA and the ^{239}Pu mass ratio of reference for the global Pu spectrum database according to gain.

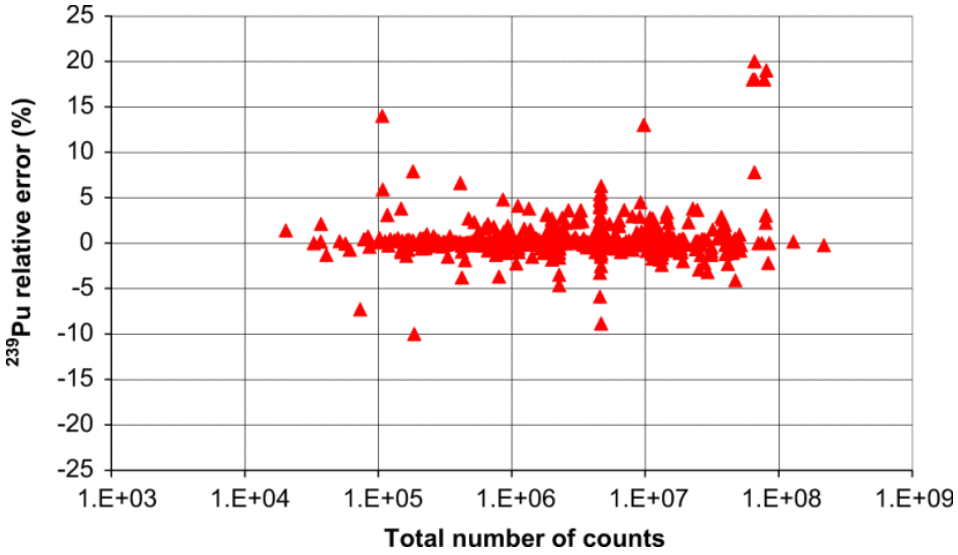

Fig. 3. Relative errors between the ^{239}Pu mass ratio obtained by IGA and the ^{239}Pu mass ratio of reference for the global Pu spectrum database according to total number of counts in the spectrum.

Even for the counting statistics, which could be a significant parameter for the accuracy of the results, no clear tendency appears for ^{239}Pu from these graphs. The conclusion is the same for the other isotopes, except for ^{238}Pu, for which a positive impact of the increasing of the counting statistics can be globally observed.

The first problem is the disparity of the database, which is not regular for the three parameters. Moreover, another more significant parameter has to be taken into account, the isotopic composition itself. The same type of graph according to the ^{239}Pu mass ratio of reference is displayed in Fig. 4. This graph shows the predominance of ^{239}Pu mass ratio on the accuracy of the results: they clearly improve when ^{239}Pu mass ratio increases. This phenomenon is well known in the domain of Pu isotopic composition measurement, due to the difficulty to correctly analyse ^{239}Pu peaks when its mass ratio decreases [7]. A similar behavior can be observed for ^{238}Pu, whose results improve when its mass ratio increases.

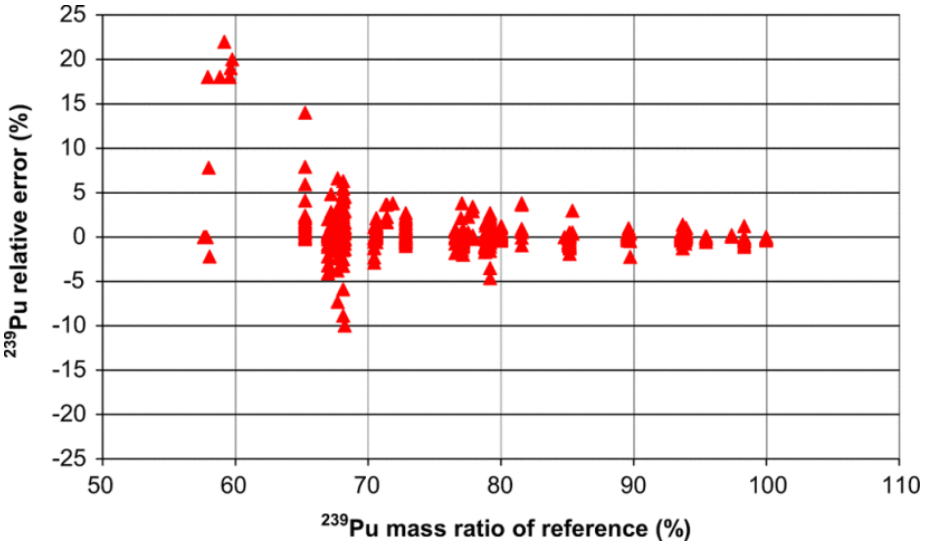

Fig. 4. Relative errors between the ^{239}Pu mass ratio obtained by IGA and the ^{239}Pu mass ratio of reference for the global Pu spectrum database according to ^{239}Pu mass ratio.

However, our purpose is to give to the user some quantitative indications on the influence of experimental parameters that he can control. So it has been decided to use another approach, described in part III.

C. Study on Homogeneous Spectrum Series-Artificially Degraded Spectra

1. Method

Instead of the global spectrum database, it was decided to work on sets of homogeneous spectra including various isotopic compositions, acquired in the same experimental conditions and to artificially modify only one parameter among energy range, gain and global counting statistics. Each set of spectra is chosen in the general spectrum database according to the studied parameter.

Thus, regarding the energy range, the set of original spectra will have a maximum energy as high as possible, like 1 MeV and each spectrum of the set will be then truncated to obtain new sets of artificial spectra with reduced energy ranges. For the gain parameter, the set of original spectra is acquired with a small gain like 0.060 keV/channel and channels are merged to create new spectra with a higher gain. Concerning the counting statistics, from a set of experimental spectra acquired with a high counting statistics, like about 10^7 total counts in the spectrum, other spectra with smaller counting statistics are generated using a Monte Carlo technique.

For each parameter, several sets of spectra are studied to confirm the observed tendency. Moreover, when the number of spectra in a collection is weak, the set can be completed with artificial spectra, also generated from the original spectra by Monte Carlo, but with the same counting statistics, in order to obtain a reliable tendency based on several dozens of spectra for each set.

2. Results

For each spectrum, the relative error between the mass ratio obtained by the IGA code and the mass ratio of reference is calculated for each isotope. In order to clearly show the impact of the parameters on the results, the cumulative distribution function of the relative error is displayed, i.e., the relative error according to the corresponding proportion of spectra. For example, Fig. 5 shows that for 75% of the spectra of each set, the resulting relative error is smaller than 2% for ^{239}Pu.

An example of results concerning the influence of energy range for ^{239}Pu is given in Fig. 5, for a set of spectra whose energy range is equal to 0–1 MeV (gain of 0.060 keV/channel, counting statistics between 2.10^6 and 8.10^7 counts, a 0.9 mm thick Cd filter). The influence of gain for ^{239}Pu is displayed in Fig. 6 for the same set. Fig. 7 gives the influence of counting statistics for a set of spectra whose number of

counts is initially about 10^7 counts in the spectrum for all the spectra (maximum energy of 600 keV, gain equal to 0.075 keV/channel).

According to Figs. 5 and 6, the assessment of the ^{239}Pu mass ratio is not strongly influenced by either the energy range or the gain: in Fig. 5, relative errors remain very close whatever the energy range is. However, it has to be noted that the test was not carried out on spectra acquired with very absorbing screens disturbing the detectionof low-energy photons (about 100 keV), like lead. The conclusion could be different in this case. Regarding the gain, results are also rather similar for values of gain up to 0.240 keV/channel. A difference of behavior can be noted for 0.360 keV/channel, for which relative errors are doubled.

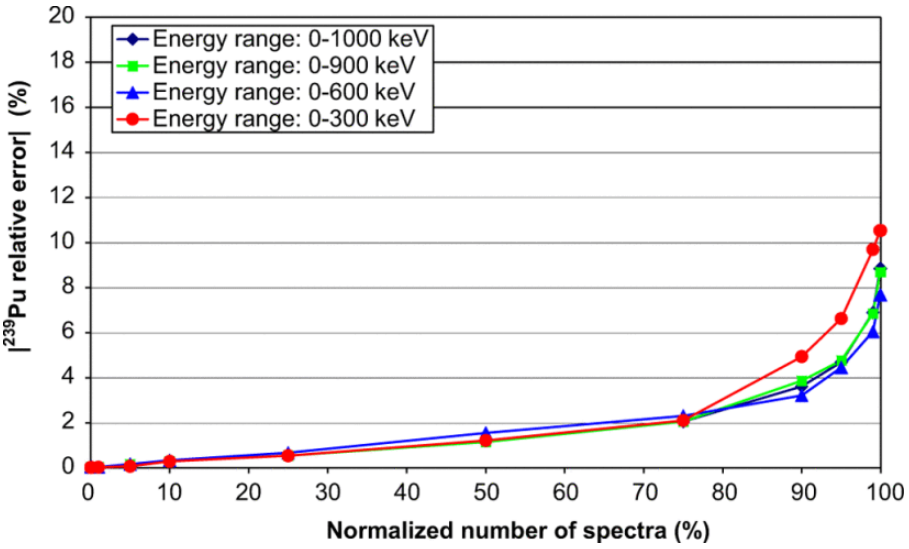

Fig. 5. Relative errors between the ^{239}Pu mass ratio obtained by IGA and the ^{239}Pu mass ratio of reference calculated for the original set of spectra with an energy range equal to 0–1 MeV and for the three sets of artificial spectra corresponding to 0–900 keV, 0–600 keV and 0–300 keV.

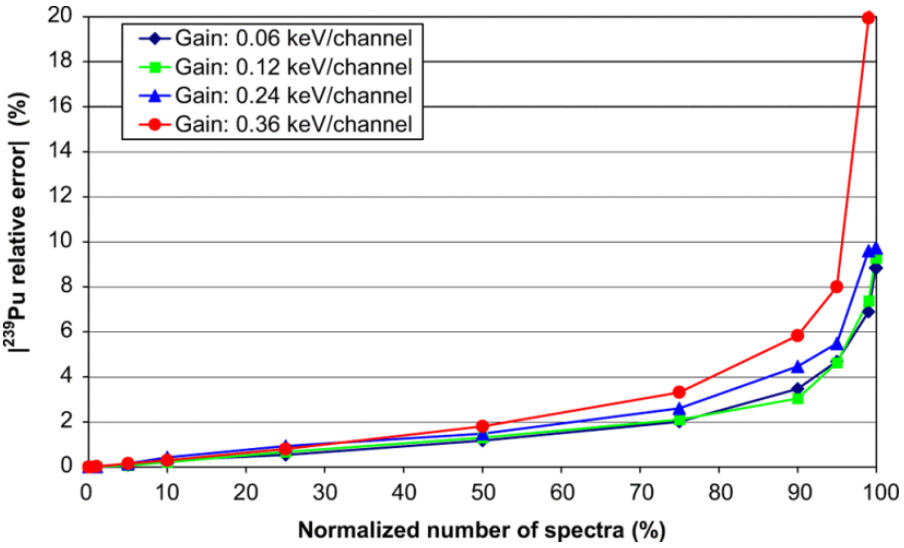

Fig. 6. Relative errors between the ^{239}Pu mass ratio obtained by IGA and the ^{239}Pu mass ratio of reference calculated for the original set of spectra with a gain equal to 0.060 keV/channel and for the three sets of artificial spectra corresponding to 0.120 keV/channel, 0.240 keV/channel and 0.360 keV/channel.

By contrast, the counting statistics has a strong impact on the results (see Fig. 7): from 10^7 counts to 10^6 counts, ^{239}Pu relative errors remain in the same order of magnitude, but they are tripled for 10^5 counts. Results for ^{240}Pu, ^{241}Pu and ^{241}Am follow the same profile for the three parameters, so they are not displayed here. The exception is ^{238}Pu, for the two parameters of gain and energy range. The corresponding results are shown in Fig. 8 for energy range.

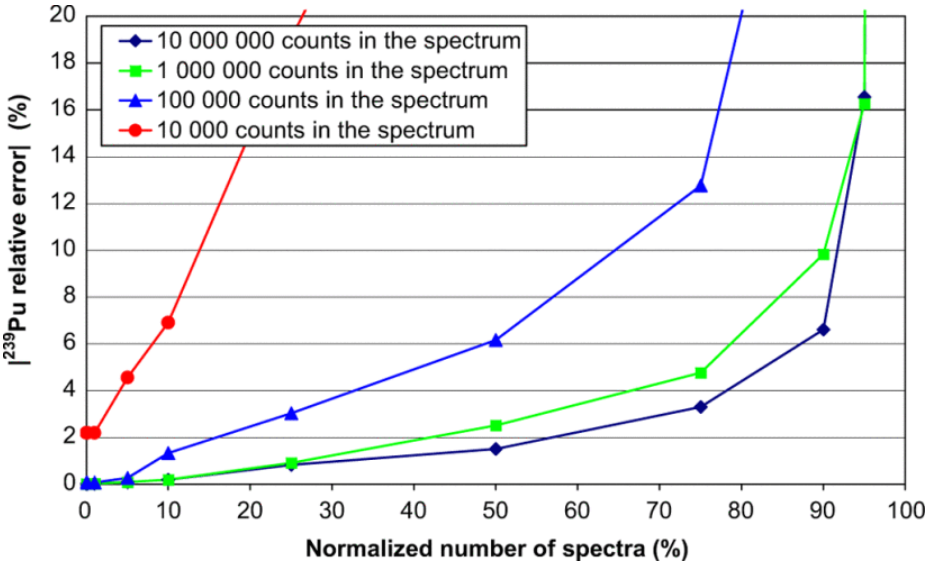

Fig. 7. Relative errors between the ^{239}Pu mass ratio obtained by IGA and the ^{239}Pu mass ratio of reference calculated for the original set of spectra with a counting statistics equal to about 10^7 counts and for the three sets of artificial spectra corresponding to 10^6 counts, 10^5 counts and 10^4 counts in the spectrum.

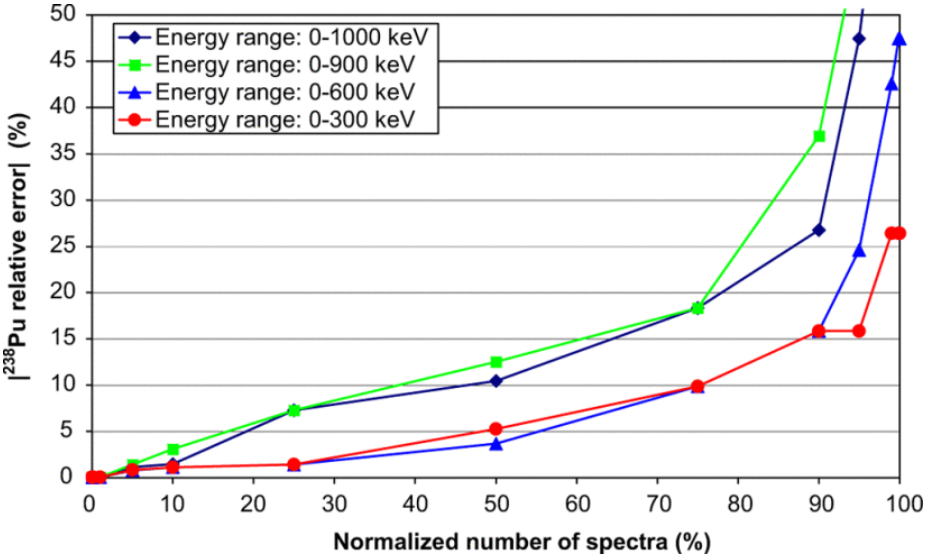

Fig. 8. Relative errors between the ^{238}Pu mass ratio obtained by IGA and the ^{238}Pu mass ratio of reference calculated for the original set of spectra with an energy range equal to 0–1 MeV and the three sets of artificial spectra corresponding to 0–900 keV, 0–600 keV and 0–300 keV.

Unlike the other Pu isotopes, ^{238}Pu results are clearly degraded for the greater energy ranges. Regarding the gain, a paradoxical behavior can be observed in some cases: if energy range is of 900 keV or 1 MeV, results are better for high values of gain. The reason lies in the ^{238}Pu peak at 766.4 keV: if energy range is higher than 700 keV, this peak is used in the spectrum analysis. But in some cases, there is confusion with the peak of ^{241}Am at 767.0 keV and the region is badly deconvolved. The ^{238}Pu results are then affected by the erroneous determination of 766.4 keV peak area. When gain is high, the code can consider the 766.4 keV peak to be of poor quality in some cases; it is then rejected by the analysis.

As said before, all these general results have been confirmed on several spectrum sets acquired with equivalent or other experimental conditions.

It can be noted that the only three studied parameters do not enable to define relative errors for each isotope. Indeed, although counting statistics for Fig. 5 or Fig. 6 is smaller than for original spectra of Fig. 7, it can be observed that relative errors are globally smaller. Counting statistics is certainly a major

parameter for the quality of results, but it has to be remembered that isotopic composition, for instance, is another one.

The advantage of this approach is to clearly show the effect of a parameter on the accuracy of the results. However, it gives no information on the dispersion of the results for a given configuration, which is linked partly to statistical uncertainty. Moreover, each spectrum set covers a large range of isotopic composition and in some cases samples of different nature (mass, conditioning…). The purpose of the last part is to more precisely quantify the IGA performances according to different parameters including isotopic composition and resolution from new experimental spectra acquired in the laboratory.

D. Study on New Experimental Spectra

1. Introduction

In order to confirm and complete the preceding results, spectra have been acquired in the laboratory with certified samples from PIDIE exercise (Plutonium Isotopic Determination Intercomparison Exercise) led by the ESARDA (European Safeguards Research and Development Association) at the end of the 80's. The isotopic compositions of the three used samples are in Table I. They are given in percentage of the total plutonium mass of each sample, which is about 0.45 g.

Sample	^{238}Pu (%)	^{239}Pu (%)	^{240}Pu (%)	^{241}Pu (%)	^{242}Pu (%)	^{241}Am (%)
PIDIE 2	0.019	89.65	10.08	0.164	0.094	0.530
PIDIE 4	0.094	78.74	19.91	0.681	0.568	2.68
PIDIE 6	0.824	68.87	24.66	2.00	3.65	7.00

Table I Isotopic Composition of the Certified PIDIE Samples

In addition to the isotopy, three other parameters will be considered here: the counting statistics, the gain and the resolution of the whole spectrometry system, which could not be studied by the preceding method. The resolution will be quantified by the FWHM (Full Width at Half Maximum) measured at 122 keV. For each studied parameter, several counting statistics from 10^5 counts to 10^6 counts in the spectrum at the minimum were carried out and for each counting statistics, at least 10 spectra were acquired.

The greatest part of the spectra has been acquired with a planar detector (Canberra GL2015 detector, 2000 mm^2, 15 mm thick, TRP preamplifier, associated with a digital gamma-ray spectrometer Ortec DSPEC). For the resolution parameter, a coaxial detector (Ortec GEM40P-PLUS, diameter equal to 68.83 mm, 44.45 mm thick, p-type, relative efficiency of 40%, TRP preamplifier, associated with a digital gamma-ray spectrometer Ortec DSPECPro) were also used to complete the resolution range. About 3000 spectra were acquired for this study. In every case, a Cd screen, 1 mm thick, was put in front of the detector in order to absorb a large part of the 59.54 keV photons of ^{241}Am. The distance between the sample and the detector is about 20 cm. No collimator or shield is used.

The following chapters give some of the most significant conclusions of this experiment.

2. Influence of Counting Statistics

For this series of experiments, the planar detector has been used with the following settings: resolution of 0.610 keV at 122 keV, gain equal to 0.075 keV/channel, 16 384 channels (maximum energy 1228 keV). For PIDIE 2 sample and PIDIE 4 sample, about ten counting statistics have been tested between 10^5 counts to 10^6 counts. For PIDIE 6 sample (the weakest ^{239}Pu mass ratio), the total number of counts in the spectrum was extended up to 1.2 10^7 counts. Each acquisition was repeated at least ten times, which corresponds to a total number of about 120 spectra by sample for PIDIE 2 and PIDIE 4 samples and 230 spectra for PIDIE 6 sample. It has to be noted that no failure of the IGA code was observed on these 470 spectra.

Figs. 9 and 10 present the mean of IGA results for the two major isotopes ^{239}Pu and ^{240}Pu. They are displayed for the three samples according to the counting statistics and compared to the reference values. The IGA code provides the uncertainty on the result for the isotopes whose mass ratio is higher than

10%. Consequently, the mean of the uncertainties given by the code for both isotopes was calculated for the ten spectra (or more) of each experimental configuration and set out on the graphs. Considering the particular calculation mode of the uncertainties, they can be considered as given at 2 standard deviations.

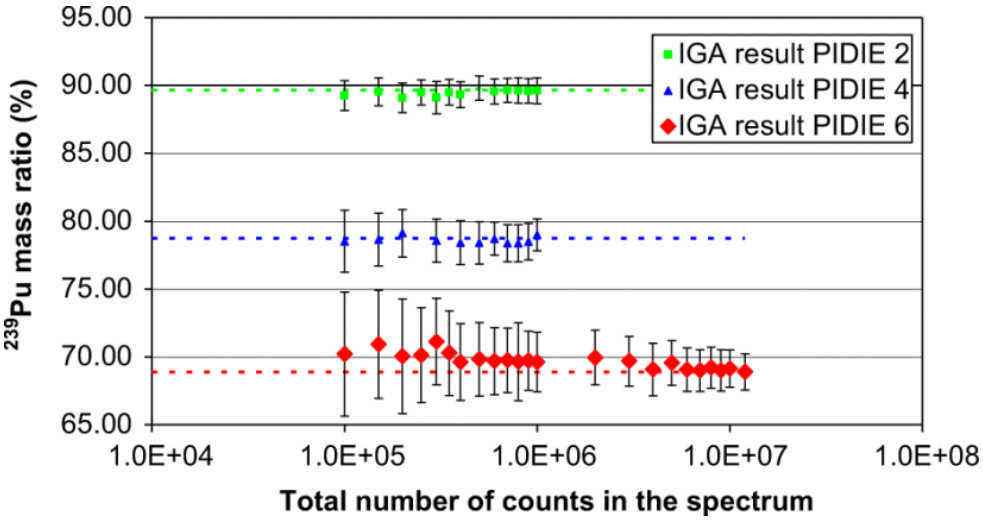

Fig. 9. IGA results for ^{239}Pu (mean of mass ratios obtained by IGA on ten or more spectra) for the three samples according to the total number of counts in the spectrum. Reference values are displayed with the dotted lines. Error bars correspond to the mean of the uncertainties provided by the code, calculated for the ten spectra of each experimental configuration.

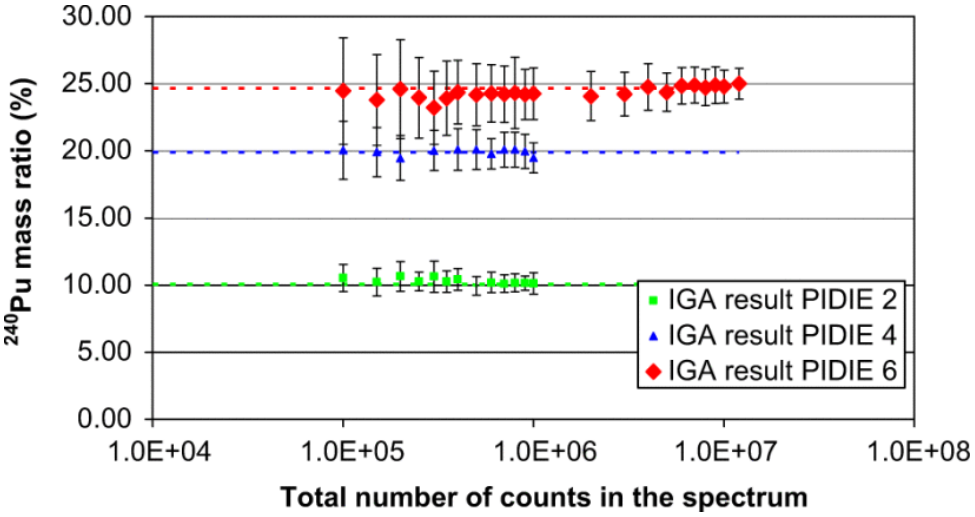

Fig. 10. IGA results for ^{240}Pu (mean of mass ratios obtained by IGA on ten or more spectra) for the three samples according to the total number of counts in the spectrum. Reference values are displayed with the dotted lines. Error bars correspond to the mean of the uncertainties provided by the code, calculated for the ten spectra of each experimental configuration.

First of all, it clearly appears that the accuracy of the results and the uncertainties depend on the sample itself, i.e., on the ^{239}Pu mass ratio: the results are better for PIDIE 2 sample than for PIDIE 6 sample. That confirms the tendency observed in part II for ^{239}Pu. Paradoxically, this is also true for ^{240}Pu uncertainties, although ^{240}Pu amount is the lowest one for PIDIE 2. Indeed, results for an isotope are linked to the other isotopes, in particular to the major isotope ^{239}Pu. So it can often be observed a dependence of the quality of all results according to ^{239}Pu. Second, the graphs show the effect of counting statistics: the results and their uncertainties globally improve when the counting statistics increases. This effect is more obvious on PIDIE 6 sample (the weakest mass ratio of ^{239}Pu); for PIDIE 2 and PIDIE 4 samples, results seem more stable on the counting statistics range because, on one hand they are already rather good from 10^5 counts and on the other hand the range is smaller.

In order to quantify the accuracy that can be expected according to isotopic composition and counting statistics, typical relative errors and the corresponding standard deviations for several counting statistics ranges, established from the ten spectra of each experimental configuration, are given for each sample in Tables II–IV. More precisely, these typical relative errors are the mean, on each counting statistics range, of the absolute values of the mean relative errors already calculated on the ten spectra of each configuration. In the same way, the given standard deviations are the mean, for each counting statistics range, of the standard deviations of the relative errors calculated on the ten spectra. It can be remarked that the statistics ranges are not exactly the same in the three tables. Indeed, the decrease of the relative error or of the relative standard deviation is not a regular phenomenon according to the total number of counts: threshold effects can be observed and limits are not the same for the three samples.

Counting statistics	10^5 – 5.10^5 counts	5.10^5 – 10^6 counts
^{238}Pu	15 *(40)*	15 *(20)*
^{239}Pu	<1 *(<1)*	
^{240}Pu	4 *(6.5)*	<1 *(4)*
^{241}Pu	2.5 *(7)*	<1 *(3.5)*
^{241}Am	2.5 *(5)*	<1 *(3)*

Table II Typical Relative Errors (%) and Typical Standard Deviations (%, Between Brackets and in Italics) Obtained for PIDIE 2 Sample (^{239}Pu=89.65%) According to the Counting Statistics Range

Counting statistics	10^5 – 7.10^5 counts	7.10^5 – 10^6 counts
^{238}Pu	60 *(15)*	15 *(20)*
^{239}Pu	<1 *(1.3)*	<1 *(0.8)*
^{240}Pu	1 *(5)*	1 *(3)*
^{241}Pu	1.5 *(3.5)*	1.5 *(3)*
^{241}Am	1 *(3)*	1 *(2)*

Table III Typical Relative Errors (%) and Typical Standard Deviations (%, Between Brackets and in Italics) Obtained for PIDIE 4 Sample (^{239}Pu=78.74%) According to the Counting Statistics Range

Counting statistics	10^5 – 4.10^5 counts	4.10^5 – 4.10^6 counts	4.106 – 1.2 107 counts
^{238}Pu	50 *(40)*	5 *(12)*	3.5 *(3)*
^{239}Pu	2 *(4)*	1 *(2)*	<1 *(1)*
^{240}Pu	3 *(5)*	2 *(3)*	1 *(3)*
^{241}Pu	4 *(6.5)*	3 *(4)*	1 *(1.5)*
^{241}Am	4.5 *(5)*	3 *(2.5)*	1 *(1.5)*

Table IV Typical Relative Errors (%) and Typical Standard Deviations (%, Between Brackets and in Italics) Obtained for PIDIE 6 Sample (^{239}Pu=68.87%) According to the Counting Statistics Range

For the three samples, as soon as the total number of counts in the spectrum is enough (more than about 5.10^5 counts for the samples of higher ^{239}Pu mass ratio and about 5.10^6 counts for the sample of smaller ^{239}Pu mass ratio), accuracy for each Pu isotope is about 1% (less than 1% for ^{239}Pu), for a dispersion of a few percent (1% or less for ^{239}Pu). Even when counting statistics is smaller, accuracy can be considered as acceptable according to the application, since relative errors remain of a few percent or less than 1%.

The exception is ^{238}Pu, for which results are globally more degraded than for the other Pu isotopes, except for high counting statistics on PIDIE 6 sample, which contains the highest quantity of ^{238}Pu. In part III, it has been shown that the IGA analysis can be disturbed for ^{238}Pu when the energy range is higher than 700 keV. So, all the spectra have been truncated to 4096 and 8192 channels to form spectra of maximum energy equal to 307 keV and 614 keV. As expected, no significant difference has been obtained for ^{238}Pu between the 307 keV configuration and the 614 keV configuration. The new results for spectra of 614 keV energy range are presented in Table V. They confirm the part III conclusion, since they are quite better in some cases (in bold in Table V). The analysis of detailed results for a few spectra shows that the 766 keV peak of ^{238}Pu is implicated in the bad results again. It can be used even when statistics in the region is too weak and with a too high weight compared to the other ^{238}Pu peaks (99.9 keV and 152.7 keV). So, weight calculation mode has to be slightly modified to remedy the problem.

PIDIE 2	3 *(25)*		10 *(20)*
PIDIE 4	10 *(15)*		5 *(5)*
PIDIE 6	3.5 *(6)*	2.5 *(3)*	2.5 *(1.5)*

Table V Typical Relative Errors (%) and Typical Standard Deviations (%, Between Brackets and in Italics) Obtained for ^{238}Pu With the Spectra Truncated to 614 Kev. Limits of Counting Statistics Ranges are the Same as in Tables II–IV for the Corresponding Sample

3. Influence of Gain

For this series of experiments, the planar detector was used with the following settings: resolution of 0.610 keV at 122 keV, 16 384 channels (maximum energy 1228 keV), counting statistics equal to 10^6 counts in each spectrum. Two new values of gain were employed, 0.125 keV/channel and 0.250 keV/channel. Ten spectra at the minimum were acquired for each experimental setup.

The mean of the relative errors and the corresponding standard deviations established from the ten spectra of each experimental configuration are given for each sample in Tables VI–VIII. 90 spectra were analyzed for that; only five failures of the IGA code have occurred, all for the configuration 0.250 keV/channel with PIDIE 6 sample. The reason is difficulties for IGA to calibrate the spectra.

Gain	0.075 keV/channel	0.125 keV/channel	0.250 keV/channel
^{238}Pu	10 *(20)*	10 *(15)*	2 *(25)*
^{239}Pu	<1 *(<1)*	<1 *(<1)*	<1 *(<1)*
^{240}Pu	<1 *(6)*	<1 *(3)*	<1 *(4.5)*
^{241}Pu	<1 *(3)*	3 *(2)*	<1 *(4.5)*
^{241}Am	<1 *(2.5)*	1 *(<1)*	1 *(2.5)*

Table VI Mean Relative Errors (%) and Corresponding Standard Deviations (%, Between Brackets and in Italics) Obtained for PIDIE 2 Sample (^{239}Pu=89.65%) for the Three Values of Gain (Counting Statistics 10^6 Counts)

Gain	0.075 keV/channel	0.125 keV/channel	0.250 keV/channel
^{238}Pu	15 *(15)*	2.5 *(8)*	15 *(25)*
^{239}Pu	<1 *(<1)*	<1 *(1)*	<1 *(1)*
^{240}Pu	2 *(2.5)*	<1 *(4)*	1.5 *(4)*
^{241}Pu	3 *(3)*	<1 *(4)*	<1 *(3)*
^{241}Am	2 *(2.5)*	<1 *(2.5)*	<1 *(2)*

Table VII Mean Relative Errors (%) and Corresponding Standard Deviations (%, Between Brackets and in Italics) Obtained for PIDIE 4 Sample (^{239}Pu=78.74%) for the Three Values of Gain (Counting Statistics 10^6 Counts)

Gain	0.075 keV/channel	0.125 keV/channel	0.250 keV/channel
^{238}Pu	7 *(6)*	5 *(5)*	4 *(10)*
^{239}Pu	1 *(2)*	1 *(1.5)*	4.5 *(5)*
^{240}Pu	1.5 *(4.5)*	4 *(3)*	10 *(10)*
^{241}Pu	4 *(3)*	2 *(2.5)*	3 *(4.5)*
^{241}Am	3.5 *(2)*	3 *(1.5)*	1 *(5)*

Table VIII Mean Relative Errors (%) and Corresponding Standard Deviations (%, Between Brackets and in Italics) Obtained for PIDIE 6 Sample (^{239}Pu=68.87%) for the Three Values of Gain (Counting Statistics 10^6 Counts)

Besides ^{238}Pu, no deep deterioration is observed when gain is 0.125 keV/channel or 0.250 keV/channel compared to 0.075 keV/channel, except for ^{239}Pu and ^{240}Pu on PIDIE 6 sample and gain of 0.250 keV/channel (in bold in Table VIII). In the other cases, results of relative error and standard deviation globally remain smaller than 5% for the three gains and the three samples.

For ^{238}Pu, results appear very odd again since they sometimes seem better for high gains than for the initial configuration with 0.075 keV/channel, which has already been seen in part III. Actually, when relative error is smaller but relative standard deviation is high, each individual relative error is high but on both sides from zero. The 766 keV peak of ^{238}Pu did not seem to be a criterion here. On the contrary, for PIDIE 4 sample, the weight of the 766 keV peak is involved again.

4. Influence of Resolution

For this series of experiments, the planar detector was used with the following settings: gain of 0.075 keV/channel, 16 384 channels (maximum energy 1228 keV), counting statistics equal to 10^6 counts in the spectrum. Resolution has been increased from 0.610 keV to 1.020 keV at 122 keV. To extend the resolution range, spectra were also carried out with a coaxial detector, with the same gain, channel number and counting statistics and with resolution values between 0.820 keV and 1.580 keV. For both detectors, resolution was degraded by decreasing the rise time from 4 μs to 0.8 μs. In order to avoid any distortion of peak shape, the flattop was also modified in consequence. In the two thirds of the cases, ten spectra have been acquired for each experimental configuration again. In a few cases, about 30, 100 or even 150 spectra have been registered for a configuration.

Figs. 11 and 12 show the mean of ^{239}Pu IGA results and ^{240}Pu IGA results on the ten (or more) spectra acquired in the same experimental setup. They are displayed for the three samples according to the FWHM, compared to the reference values. The error bars correspond to the mean of the uncertainties given by the code for ^{239}Pu and for ^{240}Pu on the ten (or more) repeated spectra. It has to be remarked that between 240 and 300 spectra were acquired per sample for this experiment. That corresponds to a total number of 791 spectra analyzed by the IGA code and it should be noticed that it failed on five spectra only.

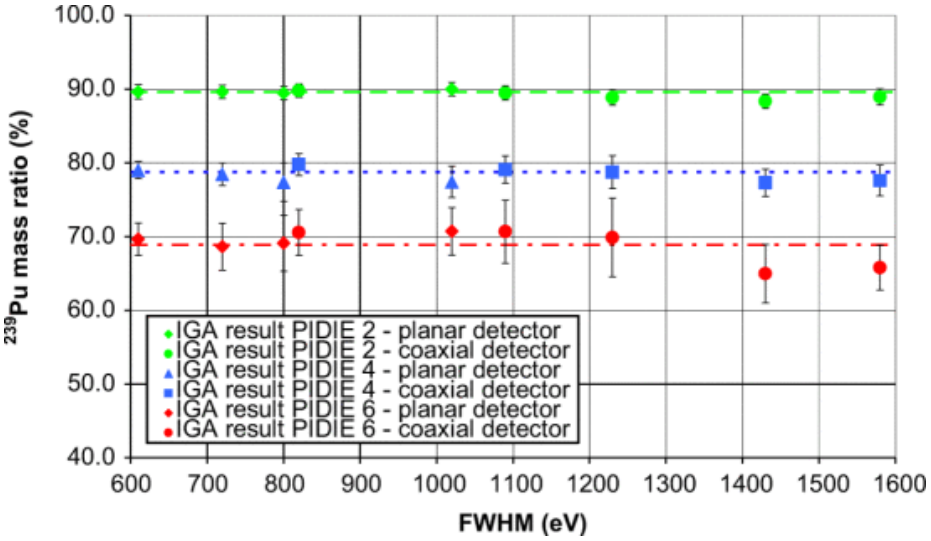

Fig. 11. IGA results for ^{239}Pu (mean of mass ratios obtained by IGA on ten or more spectra) for the three samples according to FWHM measured at 122 keV (counting statistics: 10^6 counts, gain 0.075 keV/channel). Reference values are displayed with the dotted lines. Error bars correspond to the mean of the uncertainties provided by the code, calculated for the ten spectra of each experimental configuration.

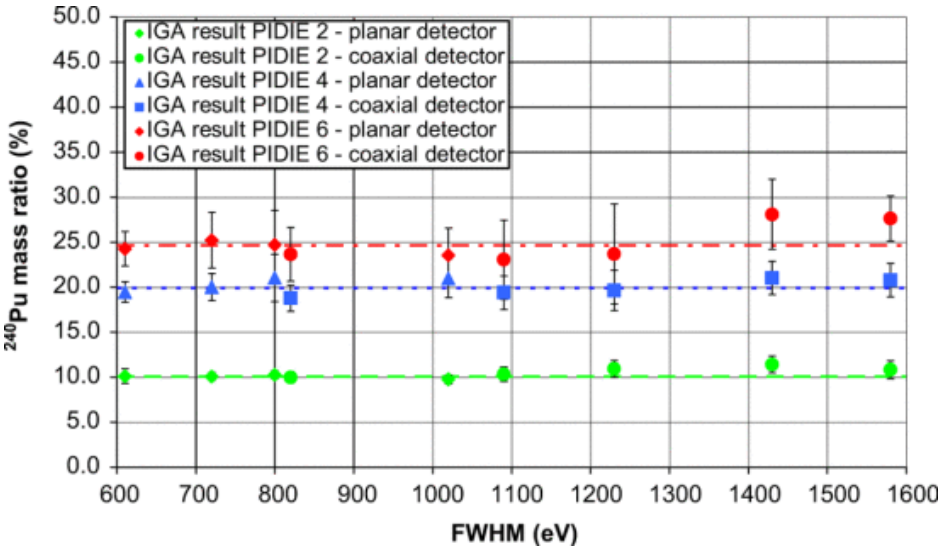

Fig. 12. IGA results for ^{240}Pu (mean of mass ratios obtained by IGA on ten or more spectra) for the three samples according to FWHM measured at 122 keV (counting statistics: 10^6 counts, gain 0.075 keV/channel). Reference values are displayed with the dotted lines. Error bars correspond to the mean of the uncertainties provided by the code, calculated for the ten spectra of each experimental configuration.

Figs. 11 and 12 illustrate a second very interesting result: relative errors and uncertainties for ^{239}Pu and ^{240}Pu are relatively insensitive to the resolution, in particular for PIDIE 2 and PIDIE 4 samples. PIDIE 6 sample, as for the other tested parameters, is more affected by changes in the experimental configuration, due to a more general difficulty to extract information.

To complete the graphs, Tables IX–XI provide the mean relative errors and the corresponding standard deviations for four values of FWHM (minimum and maximum values for both detectors) and all Pu isotopes.

FWHM at 122 keV	0.61 keV (planar detector)	0.82 keV (coaxial detector)	1.02 keV (planar detector)	1.58 keV (coaxial detector)
^{238}Pu	10 *(20)*	45 *(40)*	40 *(>100)*	>100 *(35)*
^{239}Pu	<1 *(<1)*	<1 *(<1)*	<1 *(<1)*	<1 *(1)*
^{240}Pu	<1 *(6)*	<1 *(4)*	3 *(3.5)*	7 *(9)*
^{241}Pu	<1 *(3)*	4.5 *(4.5)*	1.5 *(4.5)*	<1 *(4)*
^{241}Am	<1 *(2.5)*	4.5 *(4.5)*	<1 *(3.5)*	4.5 *(4.5)*

Table IX Mean Relative Errors (%) and Corresponding Standard Deviations (%, Between Brackets and in Italics) Obtained for PIDIE 2 Sample (^{239}Pu=89.65%) for Four Values of Resolution (Counting Statistics 10^6 Counts, Gain 0.075 KeV/Channel)

FWHM at 122 keV	0.61 keV (planar detector)	0.82 keV (coaxial detector)	1.02 keV (planar detector)	1.58 keV (coaxial detector)
^{238}Pu	15 *(15)*	10 *(20)*	10 *(20)*	50 *(20)*
^{239}Pu	<1 *(<1)*	1.5 *(1.5)*	1.5 *(2.5)*	1.5 *(2.5)*
^{240}Pu	2 *(2.5)*	5.5 *(5.5)*	5.5 *(8.5)*	4.5 *(10)*
^{241}Pu	3 *(3)*	<1 *(3)*	1.5 *(3)*	1.5 *(2)*
^{241}Am	2 *(2.5)*	<1 *(2.5)*	<1 *(3)*	3.5 *(3.5)*

Table X Mean Relative Errors (%) and Corresponding Standard Deviations (%, Between Brackets and in Italics) Obtained for PIDIE 4 Sample (^{239}Pu=78.74%) for Four Values of Resolution (Counting Statistics 10^6 Counts, Gain 0.075 KeV/Channel)

FWHM at 122 keV	0.61 keV (planar detector)	0.82 keV (coaxial detector)	1.02 keV (planar detector)	1.58 keV (coaxial detector)
^{238}Pu	7 *(6)*	3.5 *(9)*	2 *(10)*	6.5 *(15)*
^{239}Pu	1 *(2)*	2.5 *(4)*	2 *(6)*	4.5 *(5)*
^{240}Pu	1.5 *(4.5)*	4 *(10)*	3 *(15)*	12 *(10)*
^{241}Pu	4 *(3)*	<1 *(5)*	<1 *(5)*	<1 *(10)*
^{241}Am	3.5 *(2)*	1.5 *(4.5)*	1.5 *(6)*	5.5 *(5.5)*

Table XI Mean Relative Errors (%) and Corresponding Standard Deviations (%, Between Brackets and in Italics) Obtained for PIDIE 6 Sample (^{239}Pu=68.87%) for Four Values of Resolution (Counting Statistics 10^6 Counts, Gain 0.075 Kev/Channel)

The tables confirm the preceding tendency: IGA results remain acceptable when resolution degrades, not only on ^{239}Pu, but also on the other Pu isotopes (except ^{238}Pu in particular for the first two samples again). Values higher than 1% for ^{239}Pu and higher than 5% for the other isotopes (besides ^{238}Pu) are in bold in Tables IX–XI.

E. Conclusion

The purpose of the described studies was to assess the IGA performances that can be expected on Pu spectra according to some experimental parameters, like energy range, gain, total number of counts, or resolution. Indeed, the IGA code is a very flexible code that accepts any kind of spectrum without imposing constraints on the experimental setup. However, the question was to have an idea of what are the most significant parameters to obtain the best results.

The three approaches have shown that the predominant parameters are the isotopic composition itself, in particular for ^{239}Pu and ^{238}Pu and then, the counting statistics. In comparison, gain and energy range do not influence the results strongly. Thus, typical accuracy for the different Pu isotopes has been given for different isotopic compositions and counting statistics ranges.

It is important to emphasize that the high flexibility of IGA has been confirmed during this study. Indeed, even in cases where experimental setup is degraded (counting statistics down to 10^5 counts, gain up to 0.250 keV/channel, or FWHM up to 1.580 keV at 122 keV), the analysis is still possible and results can be considered as acceptable in most cases, with precision values remaining better than 5%. In particular, the good results obtained with a degraded resolution allow us to think that the analysis could be possible on CdTe spectra [8].

To complete this important work, other experiments on Pu could be carried out, first to achieve the panel of the IGA performances according to gain and counting statistics with a coaxial detector, also largely used in the field of applications, second to characterize the IGA behavior in presence of very absorbing screens (for instance copper, steel, or lead) between the sample and the detector, which could modify the conclusion relative to energy range. Another point would be to correlate the relationship between the accuracy of the results and the number of counts in key peaks for each isotope instead of the total number of counts in the spectrum.

Finally, the same type of study is in progress on uranium spectra and has to be led on uranium-plutonium mixture spectra.

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