IGA

(actinides gamma isotopy)

An automatic software for the determination of actinides isotopic abundances

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Presentation

- Introduction
  - ➔ History and philosophy
- IGA algorithm
  - ➔ Examples
- Validation and performances
  - ➔ Methods used for validation and testing
  - ➔ Typical results
  - ➔ Limitations

A little history

1998: request for a new code for the determination of the isotopic composition of Pu from a working group CEA-AREVA NC (formerly COGEMA)-IRSN

Specifications:

- Precise and robust at the same time
- Few input data
- **Varied and unknown experimental conditions (gain, energy range…)**
- Entirely automatic
- Clearness of results
- Traceability of the different versions

A little history

➔ 1999-2001: development of the **IGP** code (Plutonium Gamma Isotopy)

➔ 2002-2005: extension to uranium, and U-Pu mixtures in presence of other nuclides (other actinides, fission products…)

➔ **IGA code (Actinides Gamma Isotopy)**

➔ 2005-2008:

- Improvements (uncertainties calculation…)
- Performances evaluation
- Commercialization of IGA (version 7.0 rev. 281)
  - ITECH INSTRUMENTS (since January 2008) - info@itech-instruments.com
  - CANBERRA (from January 2009)

Adopted philosophy

Generic approach of the analysis

- No distinction between the isotopes in the analysis
- No specified list of peaks
- No preferential region of interest

➔ No constraint

- On the experimental setup (energy range, gain, channel number, resolution…)
- On the isotopes or on the peaks that have to be present in the spectrum

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- IGA algorithm
  - ➔ Examples
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  - ➔ Methods used for validation and testing
  - ➔ Typical results
  - ➔ Limitations

IGA algorithm

Input data

Identification of the isotopes and the spectrum type

Fine calibration

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
- Energy- and width-calibration

First estimate of efficiency and isotopic abundances

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
- Simultaneous efficiency and isotopic ratios fitting

Second estimate of efficiency and isotopic abundances

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
- Simultaneous efficiency and isotopic ratios fitting

Consistency tests

Main input data

- User data
  - Spectrum
  - Spectrum data: estimate of E_{min}, E_{max}, resolution at any given energy

    ➔ Approximate calibration

- Specific large atomic and nuclear database
  - 80 nuclides/elements (4000 lines)
  - Reduced to a working nuclides list (40 nuclides/elements) that can be modified according to user needs

→ Algorithm

IGA algorithm

Input data

→ **Identification of the isotopes and the spectrum type**

Fine calibration

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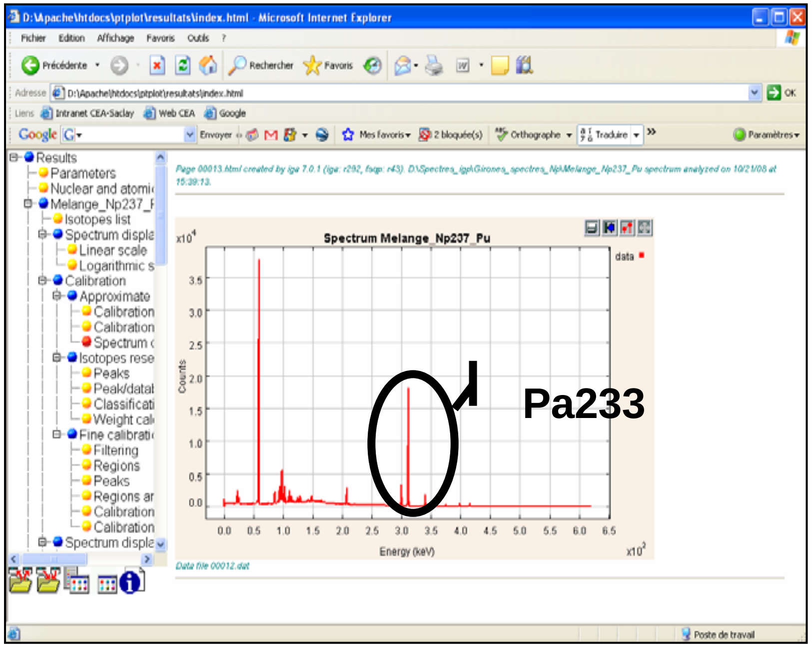
Consistency tests

Identification of the isotopes and the spectrum type

- Determination of present radionuclides
- Determination of the nature of the spectrum (Pu, U, UPu, « unknown »)

➔ List of radionuclides to be used

Example



➔ ^{233}Pa used in the processing sequence

→ Algorithm

IGA algorithm

Input data

Identification of the isotopes and the spectrum type

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Consistency tests

Database filtering

Automatic selection of the lines to consider and automatic construction of regions of interest

- According to the spectrum (energy range, resolution…)
- According to the isotopes found in the spectrum and the type of the spectrum (Pu, U…)
- According to the current analysis step (fine calibration…)

Why ?

- ➢ Different needs (isolated peaks…)
- ➢ Different available data (efficiency…)

➔ Theoretical list of lines grouped in regions of interest

→ Example

Database filtering – IGA example

number of regions	50
number of peaks	57
number of rays	68

Each following table corresponds to a region built from the atomic and nuclear lines filtered.

type	isotope	E (keV)	I (%)	T (s)	I/(AT)	(fI)/(AT)	daughter	width (keV)	type
gamma	Am241	55.560	0.0181	1.364e+010	5.5061e-017	9.2393e-019	.	.	.

type	isotope	E (keV)	I (%)	T (s)	I/(AT)	(fI)/(AT)	daughter	width (keV)	type
gamma	Pu239	56.828	0.00113	7.6084e+011	6.2142e-020	5.9675e-020	.	.	.

type	isotope	E (keV)	I (%)	T (s)	I/(AT)	(fI)/(AT)	daughter	width (keV)	type
gamma	Am241	57.850	0.0052	1.364e+010	1.5819e-017	2.6544e-019	.	.	.

type	isotope	E (keV)	I (%)	T (s)	I/(AT)	(fI)/(AT)	daughter	width (keV)	type
gamma	U237	59.541	34.5	5.832e+005	2.496e-009	4.8473e-019	.	.	.
gamma	Am241	59.541	35.94	1.364e+010	1.0933e-013	1.8346e-015	.	.	.

type	isotope	E (keV)	I (%)	T (s)	I/(AT)	(fI)/(AT)	daughter	width (keV)	type
gamma	U237	64.830	1.282	5.832e+005	9.2752e-011	1.8012e-020	.	.	.

type	isotope	E (keV)	I (%)	T (s)	I/(AT)	(fI)/(AT)	daughter	width (keV)	type
gamma	Am241	67.450	0.000421	1.364e+010	1.2807e-018	2.149e-020	.	.	.

type	isotope	E (keV)	I (%)	T (s)	I/(AT)	(fI)/(AT)	daughter	width (keV)	type
gamma	Pu239	68.699	0.0003	7.6084e+011	1.6498e-020	1.5843e-020	.	.	.

In fine calibration step, regions of more than 2 peaks are deleted.

→ Algorithm

IGA algorithm

Input data

Identification of the isotopes and the spectrum type

Fine calibration

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
- Energy- and width-calibration

First estimate of efficiency and isotopic abundances

- Filtering of the lines in the atomic and nuclear database
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Second estimate of efficiency and isotopic abundances

- Filtering of the lines in the atomic and nuclear database
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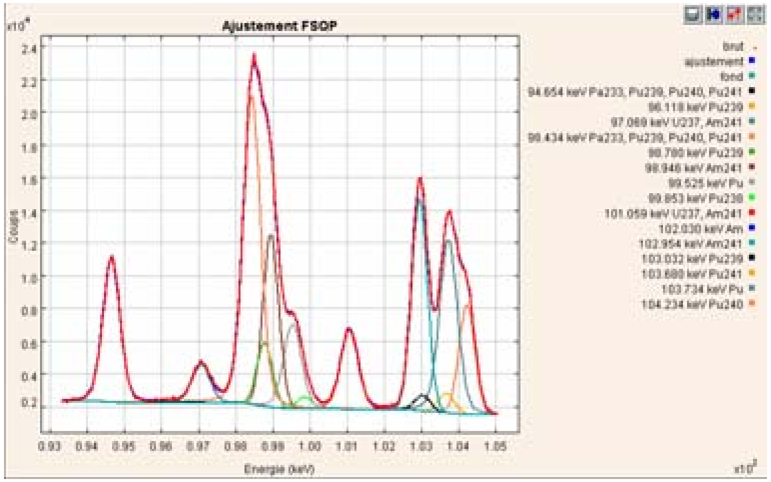
Consistency tests

Regions analysis

- Spectrum (fine calibrated or not)
- Filtered database (list of lines grouped in regions of interest)

Constrained fitting of each region

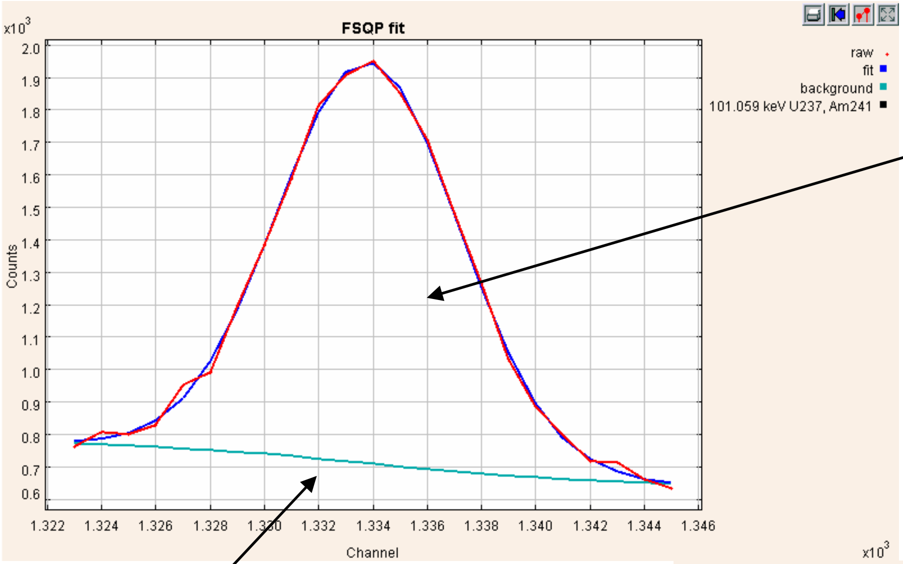
- Peak models
- Estimated efficiency
- Physical constraints (relative intensities, equilibria…)



- Peaks positions and σ for the energy- and width-calibration
- Heights (area) for the efficiency and isotopic ratios fitting
- « Quality criteria » of peaks fitting

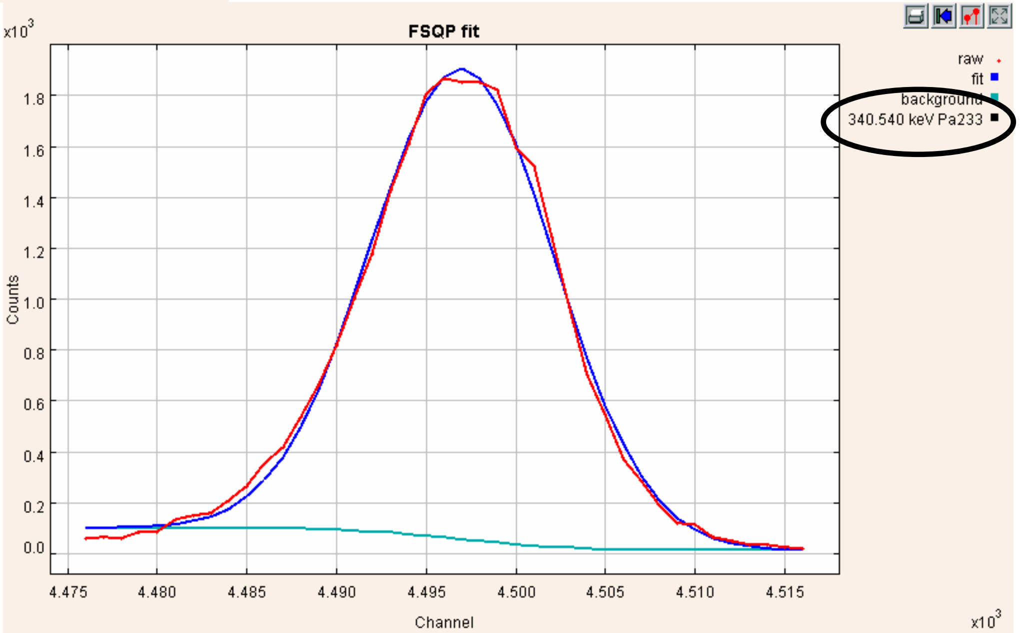
→ Example

Regions analysis – IGA example



Gaussian

Linear background + step



Peaks of ^{233}Pa are fitted and used just like the isotopes of Pu ➔

→ Algorithm

IGA algorithm

Input data

Identification of the isotopes and the spectrum type

Fine calibration

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
- Energy- and width-calibration

→ Example

First estimate of efficiency and isotopic abundances

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
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Second estimate of efficiency and isotopic abundances

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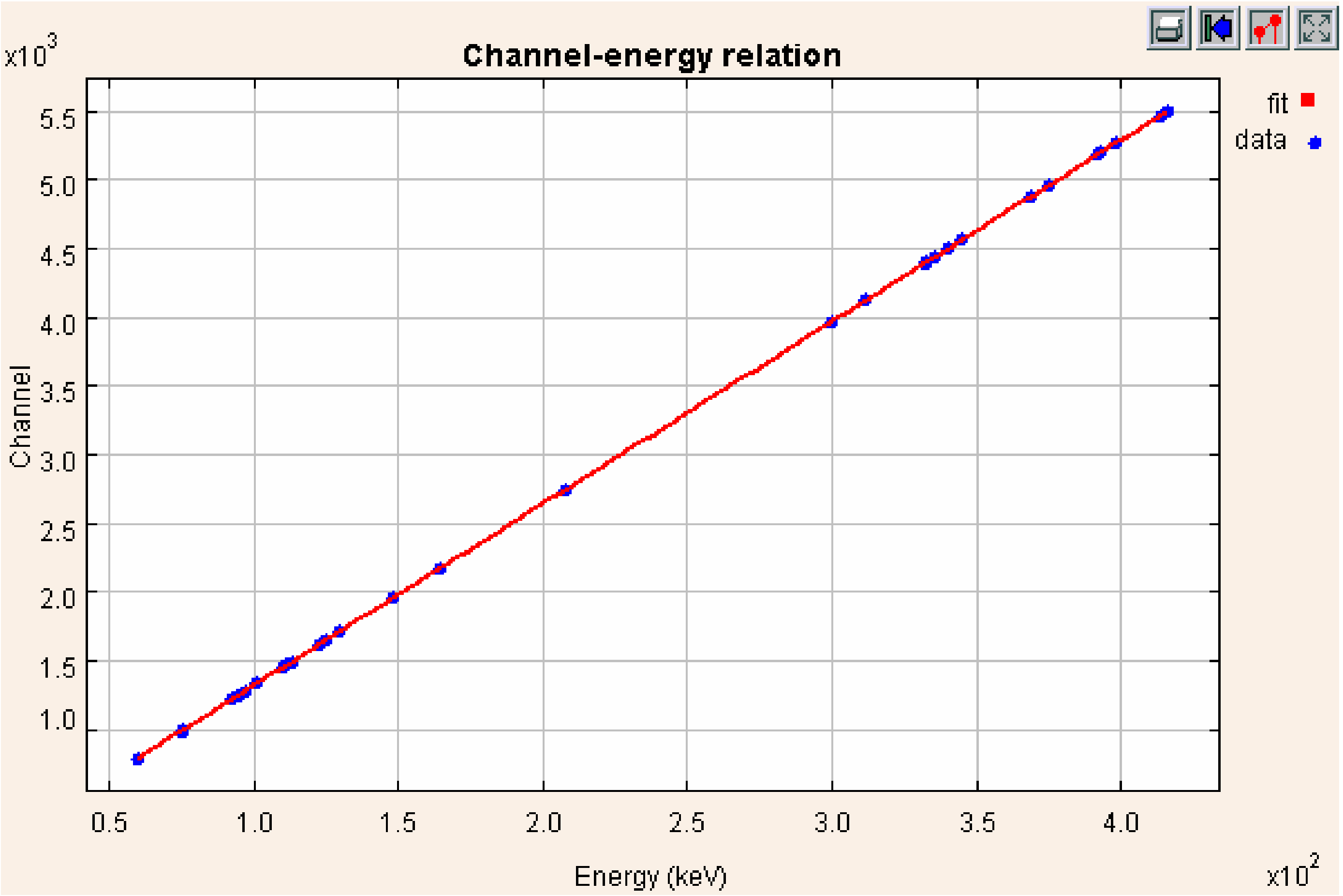
Consistency tests

Energy calibration – IGA example



- Channel-energy relation is described by a polynomial of degree 2.
- Variance-energy points are fitted by a linear relation.

→ Algorithm

IGA algorithm

Input data

Identification of the isotopes and the spectrum type

Fine calibration

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
- Energy- and width-calibration

First estimate of efficiency and isotopic abundances

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
- Simultaneous efficiency and isotopic ratios fitting

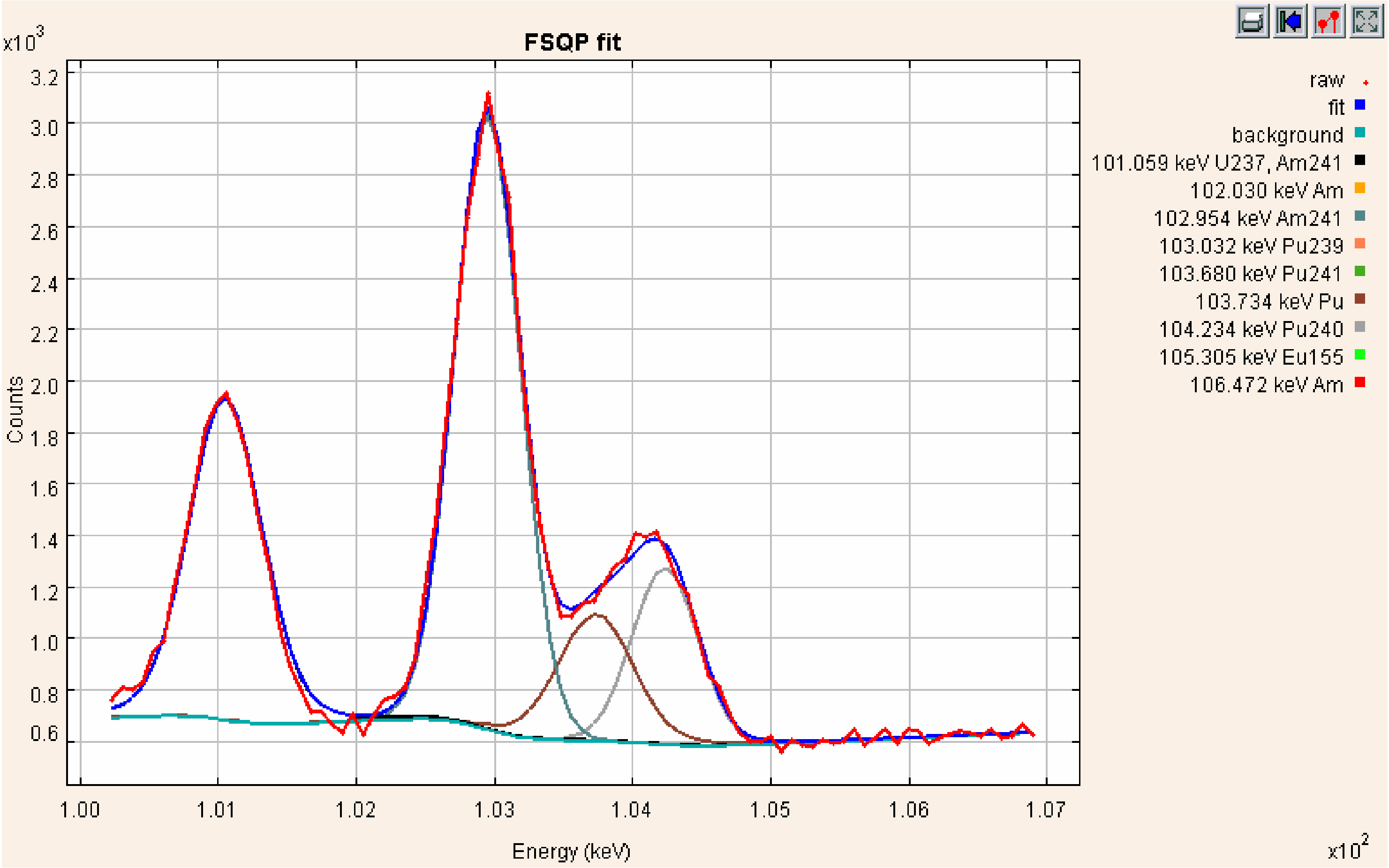
→ Example

Second estimate of efficiency and isotopic abundances

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
- Simultaneous efficiency and isotopic ratios fitting

Consistency tests

First estimate of efficiency and isotopic abundances – Region analysis – IGA example



- All the regions are fitted, even the more complex ones.
- Positions and resolutions are fixed during the fitting.

→ Algorithm

IGA algorithm

Input data

Identification of the isotopes and the spectrum type

Fine calibration

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
- Energy- and width-calibration

First estimate of efficiency and isotopic abundances

- Filtering of the lines in the atomic and nuclear database
- Analysis of the regions of interest
- → Simultaneous efficiency and isotopic ratios fitting

Second estimate of efficiency and isotopic abundances

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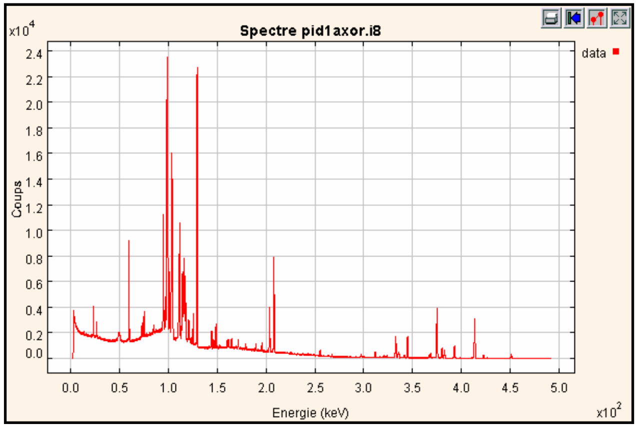
Consistency tests

General formula



Isotopic abundances ?

Measure → **Net peak area** = Data → **Atomic and nuclear data** X Unknown → **Efficiency** X Unknown → **Mass ratios**

Efficiency and mass ratios

$$\frac{\text{Net peak area}}{\text{Atomic and nuclear data} \times \text{Mass ratios}} - \text{Efficiency} = 0$$

Mass ratios: From the regions analysis

Efficiency: From an analytical model

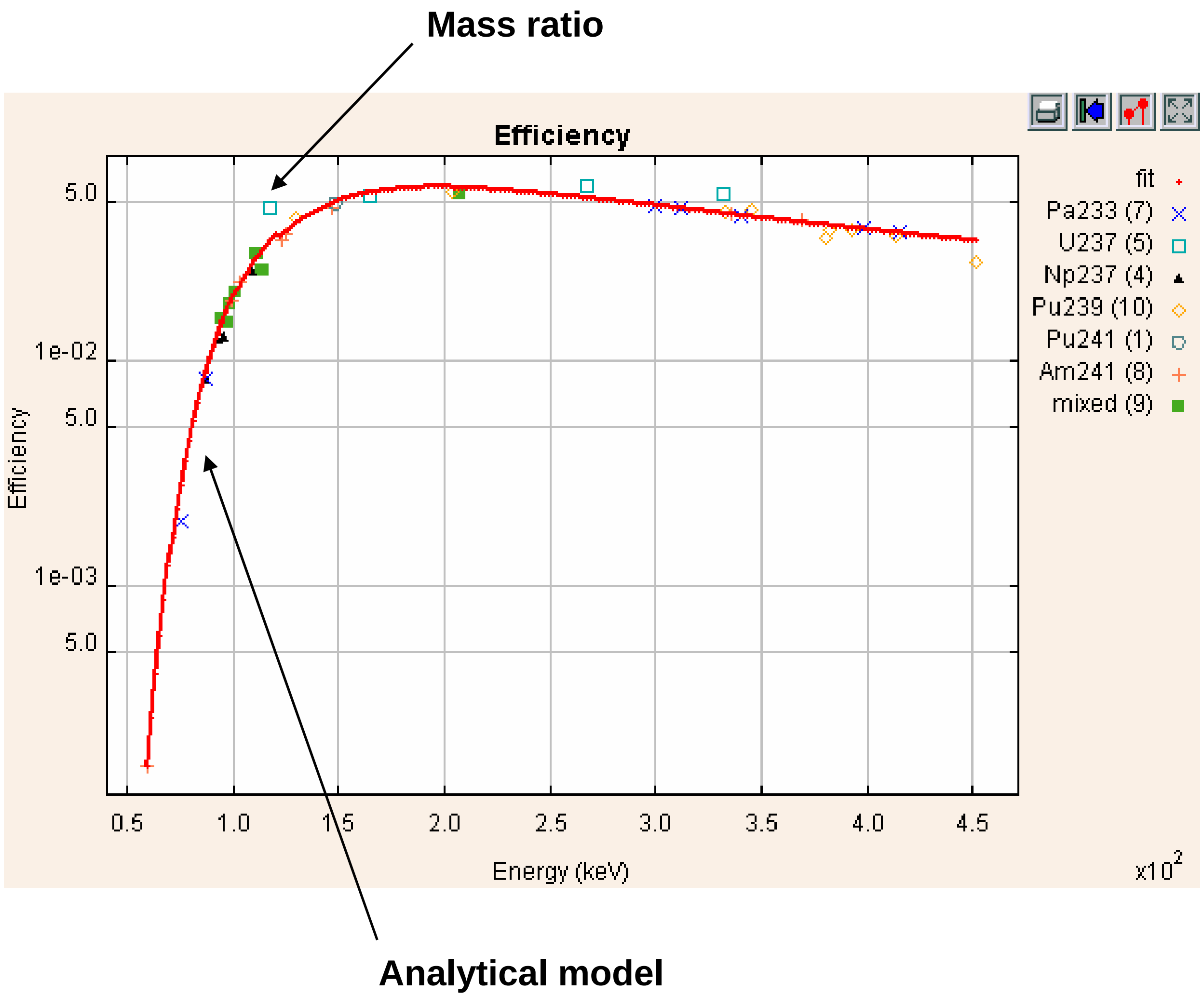
- Matrix and screen attenuation
- Self attenuation in the sample (Pu, U…)
- Detector efficiency

➔ **Minimization:**

- Mass ratios and parameters of the efficiency model are fitted **together**
- Globally carried out with **all the peaks** of the spectrum at the same time

→ Example

Efficiency and mass ratios (first estimate) – IGA example



→ Algorithm

IGA algorithm

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→ Second estimate of efficiency and isotopic abundances

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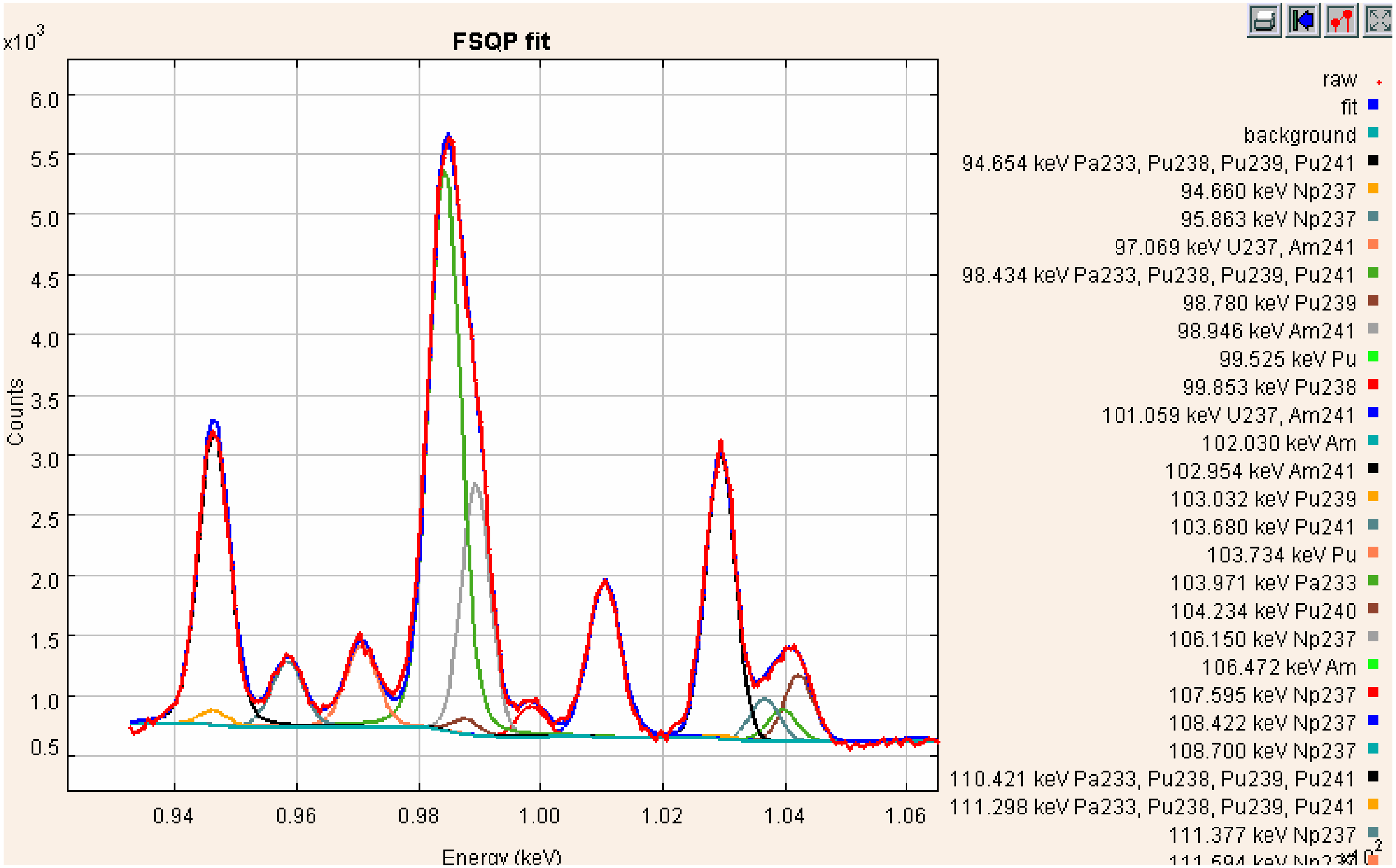
→ Example

Consistency tests

Second estimate of efficiency and isotopic abundances – Region analysis – IGA example

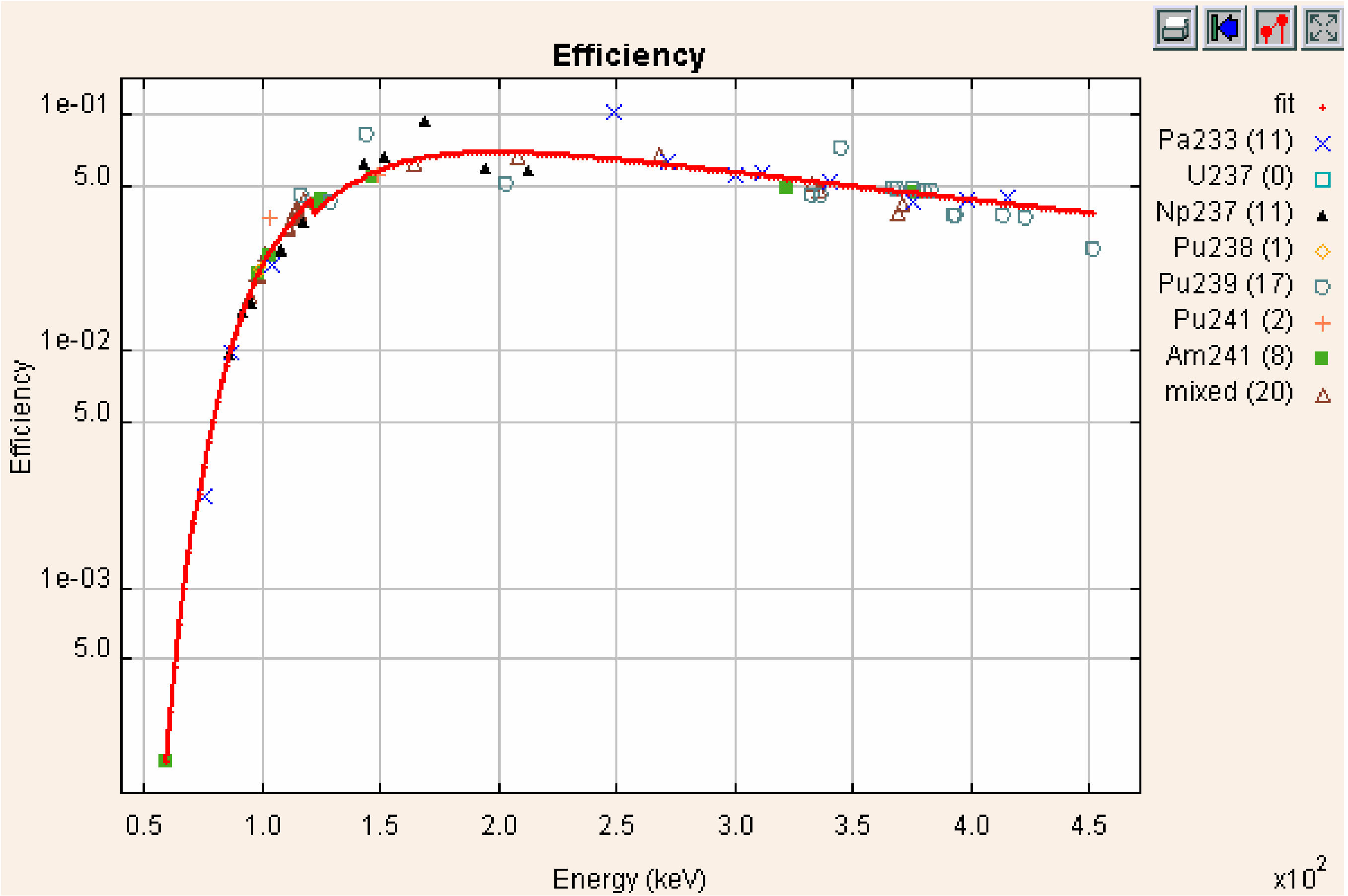


Better idea of efficiency ➔ Regions can be larger

➔ More constraints on the peaks

→ Example

Second estimate of efficiency and isotopic abundances – Efficiency and mass ratios – IGA example



→ Example

Isotopic composition - Final IGA results

Isotopes	Fractions (%)	
	LD	IGA
Pa233	8.8e-010	8e-007
U237	7.3e-010	3.2e-008
Np237	1.1	23.6 ± 2.5
Pu238	0.039	0.12
Pu239	5	82.3 ± 1.9
Pu240	2.3	16.5 ± 1.8
Pu241	0.095	1.1
Pu242	.	0.48
Pu	7.5e-018	**3.7e-018 ± 3.7e-018** *
Am241	0.0031	1.8
Am	6.8e-018	**3.4e-018 ± 3.4e-018** *
Σ (normalized isos)	.	100

→ Algorithm

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Consistency tests

Consistency tests

➔ Estimation of the results reliability

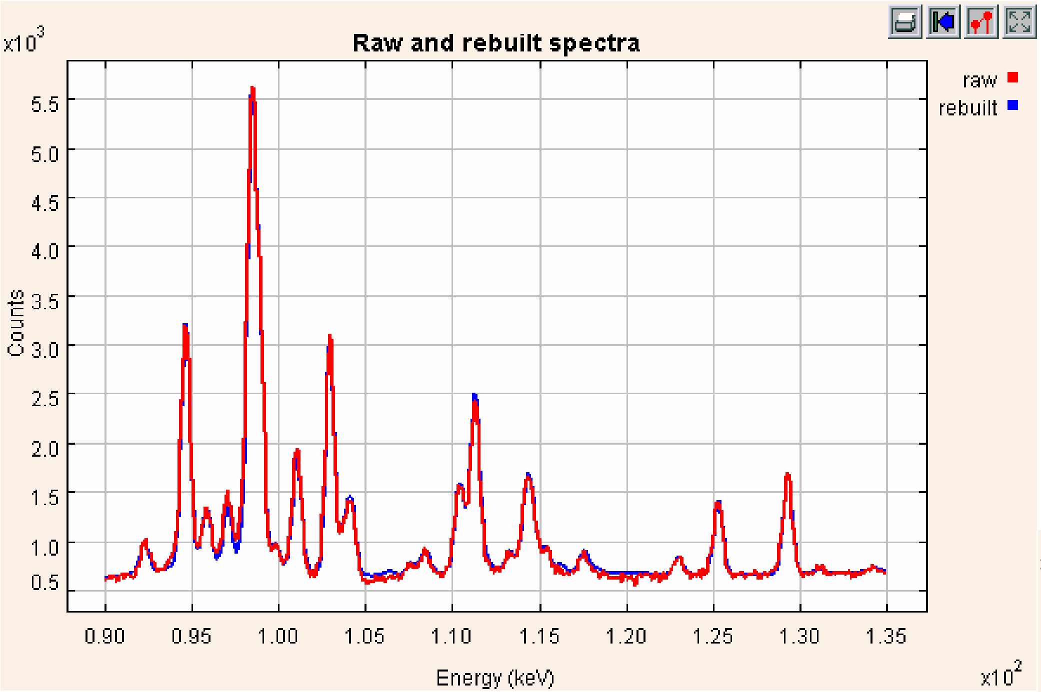
- Non analyzed peaks search
- Visual comparison between the raw spectrum and the spectrum rebuilt from IGA results →
- Comparison between the fitted areas and the areas calculated from mass ratios and efficiency results

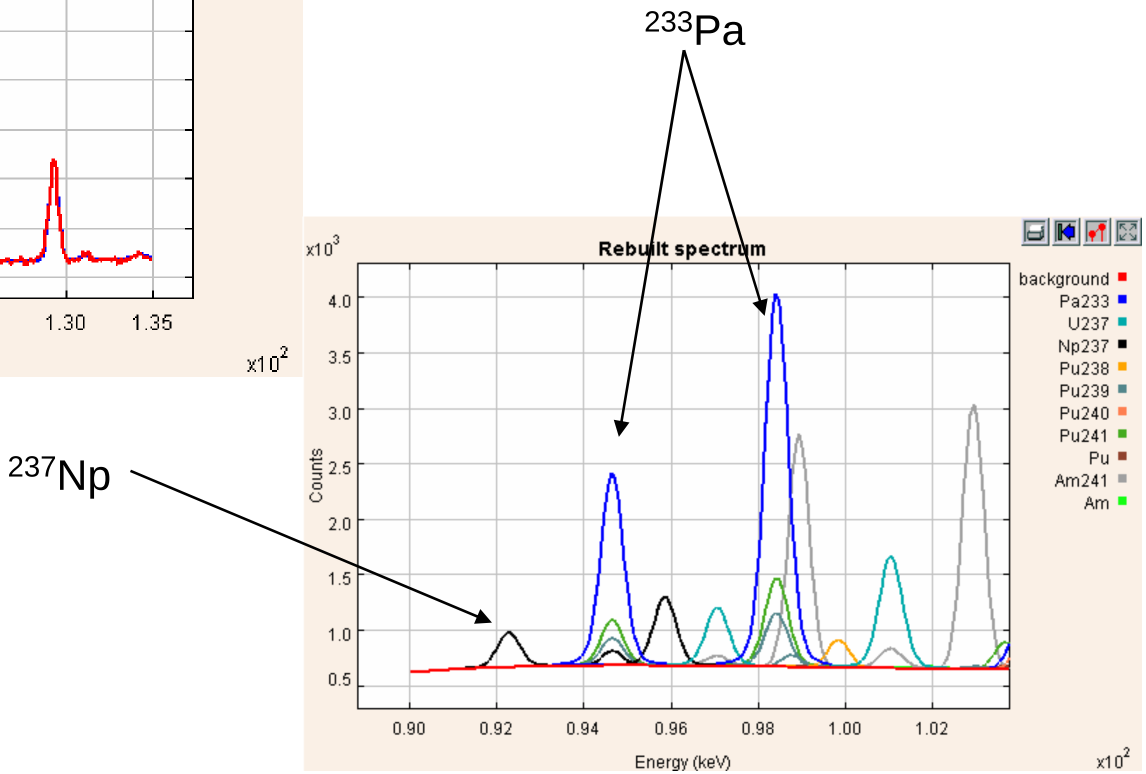
➔ Qualitative indications on the results consistency

➔ Uncertainties calculation for the major isotopes →

Consistency tests – IGA example

Visual comparison between raw and rebuilt spectra





Consistency tests – IGA example

Uncertainties calculation

Isotopes	Fractions (%)	
	LD	IGA
Pa233	8.8e-010	8e-007
U237	7.3e-010	3.2e-008
Np237	1.1	23.6 ± 2.5
Pu238	0.039	0.12
Pu239	5	82.3 ± 1.9
Pu240	2.3	16.5 ± 1.8
Pu241	0.095	1.1
Pu242	.	0.48
Pu	7.5e-018	3.7e-018 ± 3.7e-018 *
Am241	0.0031	1.8
Am	6.8e-018	3.4e-018 ± 3.4e-018 *
Σ (normalized isos)	.	100

Two tests of results coherence are carried out on each isotope:
- The first checks the presence of the main line of the isotope in the spectrum. The main line is the line with the largest area.
- The second compares the sum of fitted areas with the sum of theoretical areas (calculated from final fraction) of the isotope. The higher this deviation is, the more dubious the calculated fraction is.

LEGEND:

ITALIC	The main line of the isotope was not selected during filtering.
BLUE	The isotope fraction is lower than the fraction of detection limit.
UNDERLINED	The main line of the isotope is not kept or has a too large ratio between fitted and theoretical areas (10 %)
*	The area of the main ray of isotope was imposed on half of area of detection limit to this energy.
RED	They are the isotopes for which: - either fraction is underlined and difference between theoretical and fitted areas is higher than 50%, - or difference between theoretical and fitted areas is higher than 100%.

Warnings delivered by the code

Presentation

- Introduction
  - ➔ History and philosophy
- IGA algorithm
  - ➔ Examples
- Validation and performances
  - ➔ Methods used for validation and testing
  - ➔ Typical results
  - ➔ Limitations

Methods used for validation and testing

- Version control

  ➔ Each change in the code (including parameters) is managed by the *Subversion* tool.

    - Each version is identified by an identification number
    - Each change is documented
    - Come back to any older version is possible

- Two approaches to validate a new version

  ✓ To validate the coding

  ➔ Detailed inspection of the intermediate results for a few particular spectra

  ✓ To validate the improvement

  ➔ Calculation of the quantity $\sum_{i=238}^{241} \left| f_{ref_{Pui}} - f_{iga_{Pui}} \right|$ for each spectrum on a database of ≈ 350 spectra with various experimental conditions (≈ 160 Pu spectra, ≈ 130 U spectra, ≈ 60 UPu spectra)

Typical results - Examples

- Mean precision for the ESARDA "Pu-2000 exercise" (CEA/LNHB spectra)

^{238}Pu			^{239}Pu			^{240}Pu	^{241}Pu	^{241}Am
^{238}Pu > 0.15%	^{238}Pu < 0.15%	Total	^{239}Pu > 75%	^{239}Pu < 75%	Total			
≈ 3 %	≈ 7 %	≈ 5 %	≈ 0.2 %	≈ 1.3 %	≈ 0.7 %	≈ 3.5 %	≈ 2 %	≈ 3 %

Particular samples:

- Pure ^{239}Pu ➔ Ref: ^{239}Pu = 99.98 % **IGA result**: ^{239}Pu = 99.70 % - 100 %
- Pure ^{240}Pu ➔ Ref: ^{240}Pu = 99.96 % **IGA result**: ^{240}Pu = 99.90 %

- Mean precision for the "U exercise" – ESARDA 1997 (CEA/LNHB spectra)

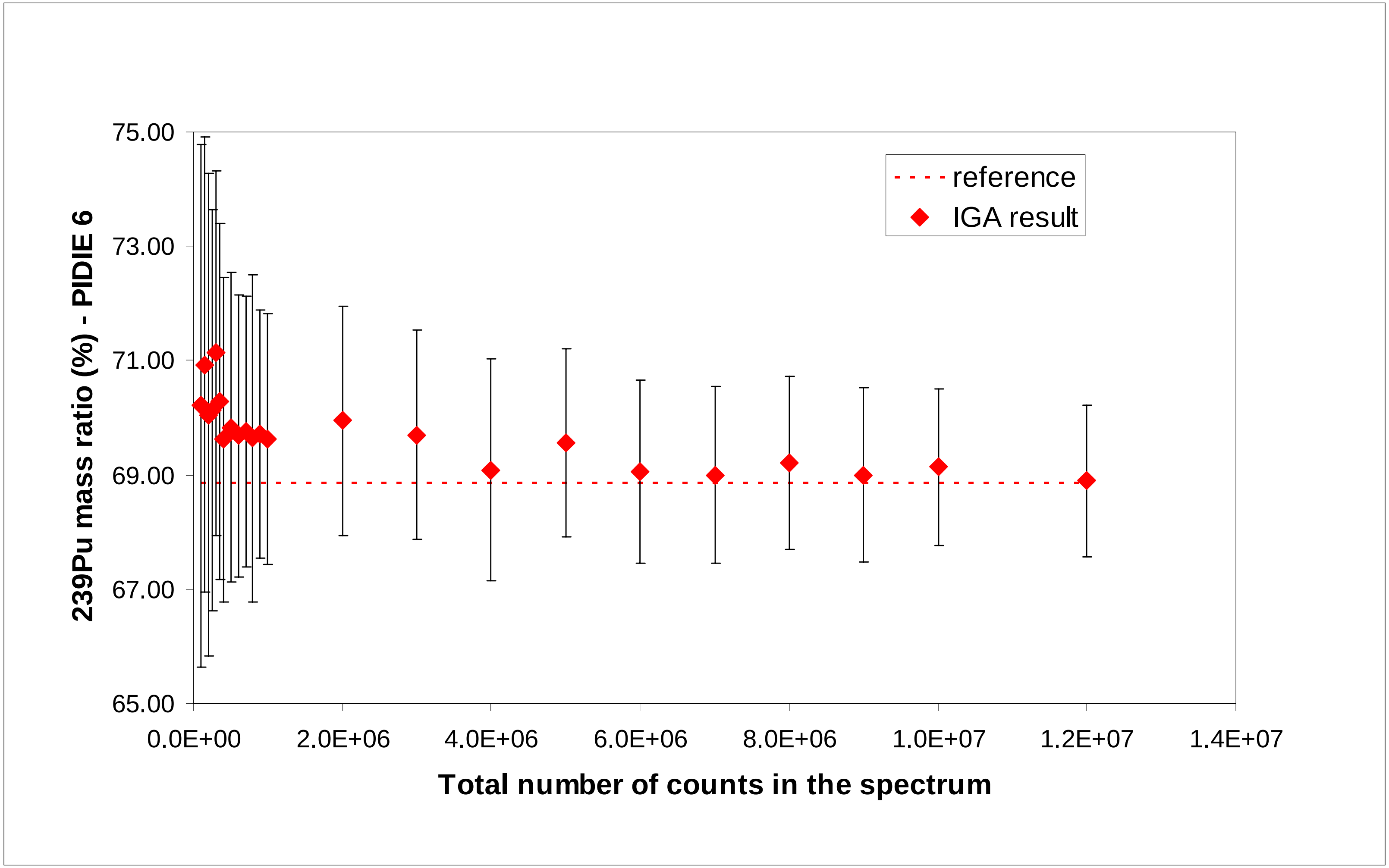
^{235}U ➔ ≈ 3 % (without freshly separated samples)

Limitations

- Precision on input data
  - **< 0.4 keV for E_{min} and E_{max}** (essential for the peak search)
  - < 0.1 keV for the resolution
- Recommended experimental conditions for the spectrum
  - **Total number of counts in the spectrum > 10^6 counts** →
  - Gain < 0.25 keV/channel
  - Energy range
    - Pu: not influential except for ^{238}Pu (< 700 keV)
    - U: not influential
  - **Resolution < 1.6 keV at 122 keV** (no knowledge for > 1.6 keV) →

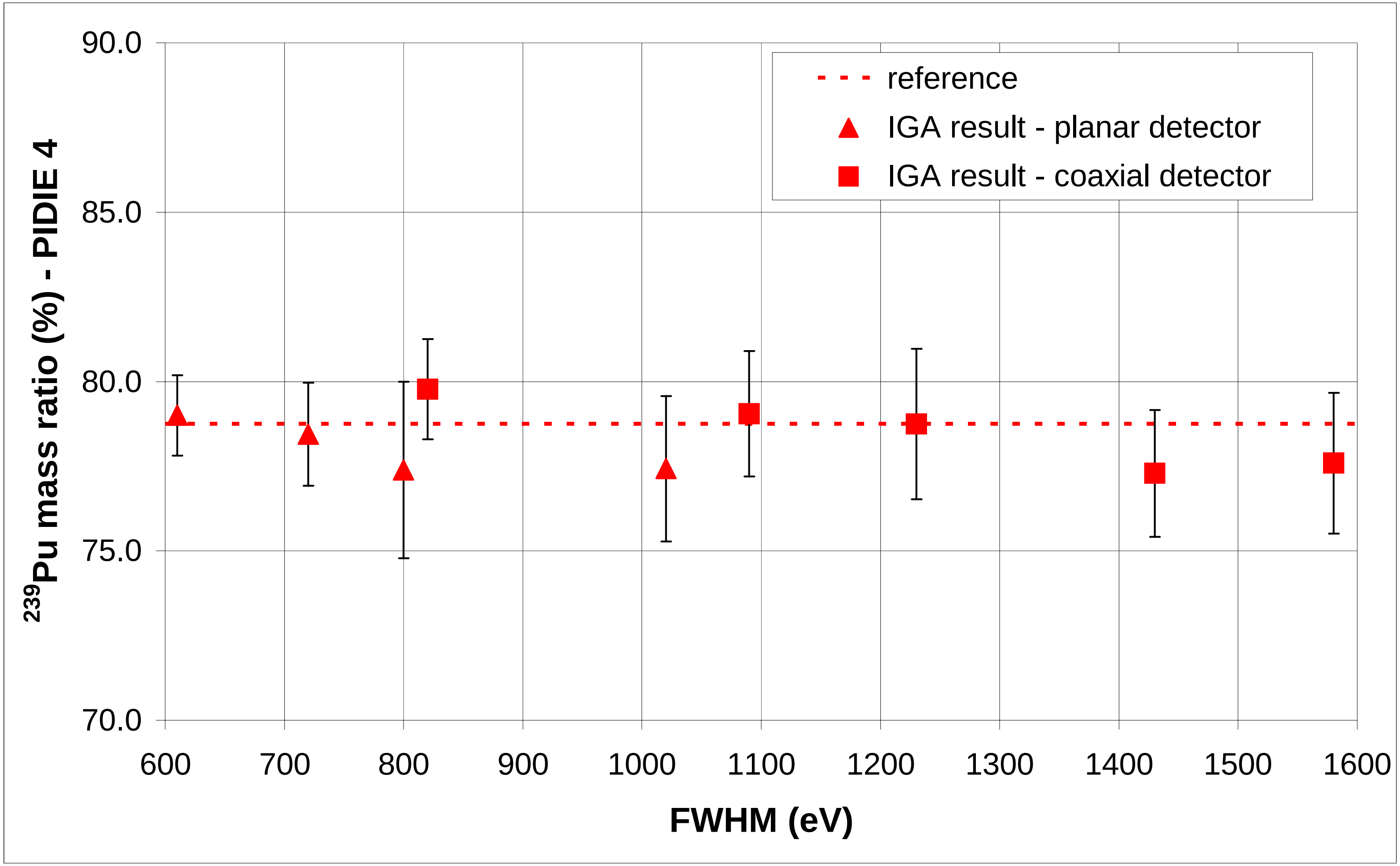
Performances – Example (1)

IGA result for ^{239}Pu versus total number of counts in the spectrum



Performances – Example (2)

IGA result for ^{239}Pu versus resolution



Summary of IGA capabilities

A **unique code** with a **user-friendly interface**

- easy to use
- fully configurable
  - from expert mode (access to each intermediate result)
  - to black box mode (final result only in a text format file)
  - batch mode possible
- high flexibility, able to adapt automatically to
  - any experimental configuration (energy range, gain, detector, screen…)
  - any isotope present in the spectrum (U, Pu, minor actinides, fission products,…)

➔ Open to various applications (fixed measurement station, in-situ measurements,…), including some hard cases (drums measurements, degraded resolution, …)

Thank you for your attention.

Thanks to the people who have participated in the IGA project:

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Frédérick Carrel, Mickaël Lemercier (performances evaluation)