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# Isotopic composition determination codes: current state-of-the-art, recent developments and future challenges

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## Abstract

Determination of actinide isotopic composition is one of the most important tasks accomplished under IAEA safeguards treaties for verification and inspection activities. Spectrometers based on HPGe detectors have traditionally been the detectors of choice for such systems with a large array of various data analysis codes developed specifically to suit the performance of these detectors. Over the past decade in this domain there has been interest in using medium resolution room temperature detectors based on CdZnTe and LaBr<sub>3</sub>(Ce) crystals. Moreover, progress in analytical tools that could be applied to analyze worsened statistical quality spectral data has opened a wide range of possible applications where such compact detectors and absence of cryogenics are advantageous. In this paper we aim to provide a review on the recent advances in the domain of isotopic composition determination systems used as with traditional HPGe detectors as well as with medium resolution ones. We limit ourselves to the achievements in the European Union and divide our review in three parts. First, we provide an update on the current status of the French code IGA aimed at applications with traditional HPGe systems. Second, we present the recent results in the domain of isotopic composition determination codes development for medium resolution detectors, namely the codes MCSIGMA and CAMILA developed in the framework of a PhD project in Belgium. Third, we give an outlook on the current research activities conducted at CEA List Saclay in the domain of isotopic composition determination algorithms applied to medium resolution detectors.

## 1. Introduction

Determination of actinides isotopic composition is an important activity accomplished under IAEA safeguards agreements and regulations. These activities include traditional verification and confirmatory measurements of Special Nuclear Materials (SNM) as well as Material Protection, Control and Accountability (MPCA) applications for SNM identification and characterization at the nuclear facilities performed by their operators. Passive non-destructive assay methods featuring X- and gamma-ray spectrometry are the preferred methods and High-Purity Germanium (HPGe) detectors have traditionally been the detectors of choice for such applications. They benefit from the high-energy resolution yielding the possibility to distinguish closely positioned peaks in complex spectra such as those of plutonium or mixed materials. They are available in various different sizes and types, their response has been very well studied and an array of various different codes for determination of actinides isotopic composition was developed over the years. Among such codes, the most well-known codes that are commercially available nowadays are the MGA code [1], MGAU code [2-7], the MGA++ package suite [8-10], FRAM code [11-14], NaIGEM code [15], and the IGA code [16]. Among the less well-known codes are the TRIFID code [17], the PISA code [18], LEPA and PUANAL [19], GAMANAL [20], GRPANL [21] and GRPNL2 [22], GRPAUT [23, 24].

However, HPGe detectors require cooling either via the means of liquid nitrogen or thermoelectric cooling, what makes the equipment rather heavy, bulky and thus inconvenient to use for hand held devices for nuclear inspectors, limited space locations or unattended nuclear installations. Nowadays, there is an increasing interest in using gamma-ray measuring devices based on the room temperature operated medium resolution detectors. Among such devices with a promising performance are semiconductor detectors based CdZnTe (CZT) and CdTe crystals and scintillation devices based on LaBr<sub>3</sub>(Ce) crystals. CZT detectors benefit from compact size, high stopping power and their energy resolution can be as good as 2 % at 661.7 keV [25]. LaBr<sub>3</sub>(Ce) detectors offer a high light output (~60000 photons/MeV), fast response (decay constant <30 ns) and show an energy resolution of ≤ 2.9% at 661.7 keV [26-28]. Nuclear safeguards is one of the foreseen application fields for these detectors

where they represent a promising alternative to HPGe detectors used for qualitative and quantitative X- and gamma-ray spectrometry of various actinide materials as well as spent fuel assemblies [29-32].

However, due to a significantly different spectrometric performance and worse energy resolution, application of room temperature medium resolution detectors to isotopic composition determination tasks is a new challenge to take up. Thus, for example, CZT detectors exhibit asymmetrical photopeaks in their spectra and for small crystals, the counting efficiency limits the usable spectral range for analysis. LaBr<sub>3</sub>(Ce) detectors exhibit internal activity of <sup>138</sup>La radioisotope, which limits its application in low count rate measurements. Besides, significant peak overlapping in complex spectra is common challenge for medium resolution detectors due to their worse energy resolution compared to HPGe detectors. The traditional codes used for determination of uranium (U) and plutonium (Pu) isotopic composition were optimized for the spectrometric performance of HPGe detectors and cannot be directly used with room temperature devices, such as CZT and LaBr<sub>3</sub>(Ce) since they require specific parameters and high resolution spectra quality [33]. As a result, to answer the question of the feasibility of medium resolution detectors for actinides isotopic composition determination tasks in safeguards applications, algorithms that would take into account their specific spectrometric performance are necessary. That requires adaptation of the mathematical formalism in their unfolding routines, models and statistical tools used to analyze such data.

Nowadays, application of passive X- and gamma-ray assay methods for isotopic composition determination of U and Pu materials in safeguards tasks still widely relies on the performance of HPGe detectors. As a result, sustainability of isotopic codes optimized for HPGe detectors is an important framework of support programs to the IAEA. As for application of room temperature medium resolution detectors for determination of actinides isotopic composition, the progress in this domain depends on technical criteria, which reflect performance assessment and data analysis tools development process for these detectors in order to consider the three main elements relevant to the problem: (a) applicability of medium resolution detectors to isotopic composition determination tasks using various different data analysis methods, (b) related accuracy and uncertainty budgets, (c) the safeguards requirements with respect to the performance limits in specified applications.

Thus, in this paper we aim to fulfill a three-fold goal. First, we will outline the current state-of-the-art with an existing French code IGA (Actinide Isotopic Composition) for HPGe detectors as these systems remain the benchmark detectors that are used for isotopic composition determination tasks. Second, we will also provide an overview on the most recent developments in the domain of isotopic codes for room temperature medium resolution detectors, limiting ourselves to the developments in the EU. Finally, we will present experimental results obtained with different types of medium resolution detectors (CZT, CdTe and LaBr<sub>3</sub>(Ce+Sr)) and review the current research activities conducted at CEA LIST Saclay in this domain.

## **2. Sustainability and progress in the domain of isotopic composition codes**

### ***2.1. State-of-the-art with IGA code***

The IGA code was developed by CEA List in partnership with COGEMA company, which in early 2000's became AREVA NC and, after restructuring of the latter in 2017, nowadays is known as ORANO. The IGA code is dedicated to the determination of the actinide isotopic composition by analysis of X- and gamma-ray spectrum obtained using HPGe detectors. The code can be used for U, Pu and mixed U+Pu spectra analysis.

Compared to other isotopic codes, the IGA code has a number of attractive performance features. First of all, it has flexibility regarding measurement settings. Thus, the energy range and the number of channels are free for the end-user (whereas for example, the reference MGA code requires setting of a pre-defined energy range with a standard energy gain equal to 75 eV.channel<sup>-1</sup>). Secondly, the IGA code requires only three parameters to use it (such as minimal and maximal energy and a resolution value at any energy selected by the user). Finally, a crucial advantage of the code is the ability to have access to all detailed analysis results performed by the code (net peak areas, efficiency curve...).

The core processing algorithm of the code is based on the principle of simultaneous determination of the relative counting efficiency and mass fractions, which is accomplished via an iterative process in

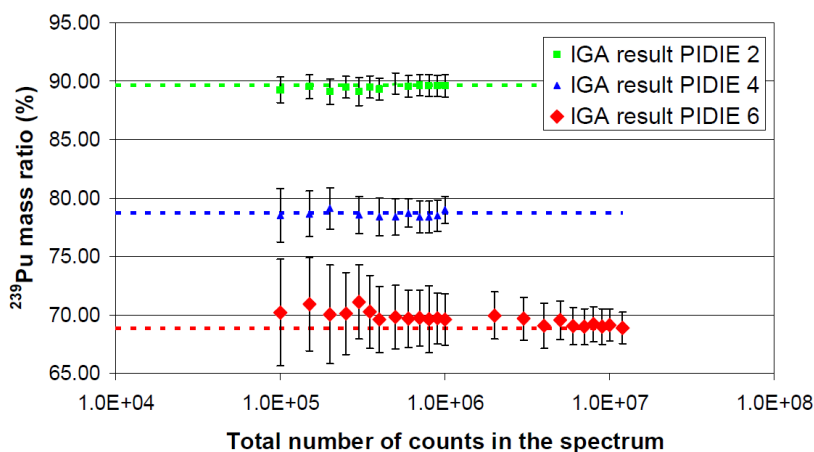
several steps. The counting efficiency model includes three-terms, self-attenuation in the U and/or Pu matrix, attenuation in a screen, and an analytical expression approximating the efficiency of the detector. Gamma-ray peaks are described using a Gaussian model and X-ray peaks using a Voigt model. The code includes an integrated readable atomic and nuclear databases (~ 90 elements/radionuclides, ~ 4000 X- and  $\gamma$ -rays) and a working database (~ 40 elements/radionuclides). Automatic filtering of the database is applied according to the spectrum and the analysis step, depending on present isotopes, energy resolution and range, peak intensities, or aim of the current step. The most recent version of the code is 7.2. A detailed description of the code working principles, data unfolding and analyzing routines is described in the reference [34]. The code is currently commercialized in an industrial version by Mirion Technologies (CANBERRA).

The performance characterization of the code was carried-out during several extensive exercises with U and Pu standards. The performance characteristics in terms of determination accuracy (the mean relative deviation between the IGA code result and the reference isotope fraction) and corresponding standard deviation for different plutonium radioisotopes are summarized in Table 1. The measurements were conducted using a 124 cm<sup>3</sup> coaxial HPGe detector (energy resolution 0.9 keV@208 keV). Details on the experimental configuration and parameters are presented in the reference [35].

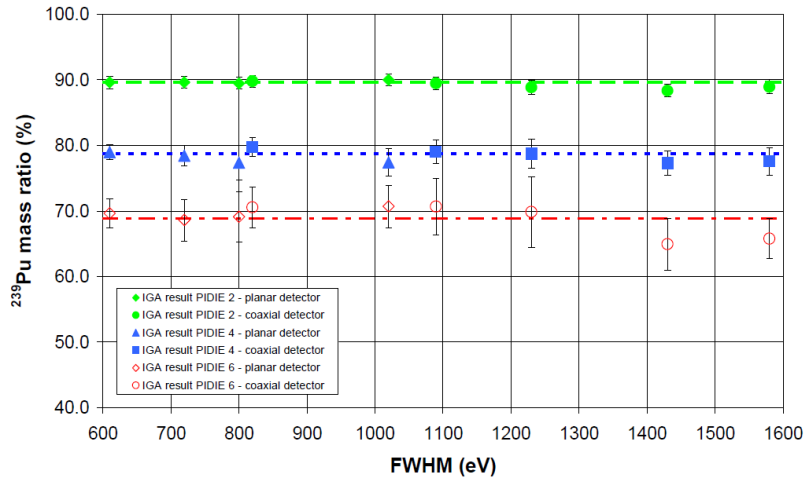
**Table 1:** IGA code accuracy for different Pu isotopes [35].

<sup>238</sup> Pu			<sup>239</sup> Pu			<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>241</sup> Am
<sup>238</sup> Pu > 0.15%	<sup>238</sup> Pu < 0.15%	Total	<sup>239</sup> Pu > 75%	<sup>239</sup> Pu < 75%	Total			
3.4% (1.6)	7.2% (7.5)	5.7% (6.1)	0.2% (0.2)	1.3% (1.1)	0.7% (0.9)	3.5% (6.3)	2.0% (2.0)	2.9% (3.7)

The impact of such parameters as energy resolution, counting statistics and Pu isotopic composition has been investigated on the code performance using a set of PIDIE (Plutonium Isotopic Determination Inter-comparison Exercise) samples [36]. Corresponding results are summarized in figures 1 and 2 [37].



**Figure 1:** Impact of the counting statistics on the IGA code performance with Pu samples of different isotopic composition [37].



**Figure 2:** Impact of the detector energy resolution on the IGA code performance with Pu samples of different isotopic composition [37].

The code has demonstrated its ability to analyze spectra having a degraded energy resolution (resolution at 122 keV close to 1.6 keV) and with different types of HPGGe detectors (planar and coaxial). Moreover, thanks to the code's flexibility, analysis is always possible, even if some key peaks are missing in the spectrum (for instance, the intense 208 keV peak, due to gamma-rays emitted by  $^{237}\text{U}/^{241}\text{Am}$ ). With improved counting statistics ( $> 10^6$  counts) the dispersion of the determined  $^{239}\text{Pu}$  mass values decreases as well as the corresponding uncertainties. Error bars correspond to the mean of the uncertainties provided by the code, calculated for the ten spectra of each experimental configuration. A detailed assessment campaign of the IGA code performance with plutonium spectra analysis is presented in the reference [37].

The assessment of the code performance with U spectra analysis was conducted using a set of reference U samples with different degrees of enrichment to investigate the impact of various parameters. The impact of the detector energy resolution on the mean relative deviation between the IGA code result and the reference enrichment in U spectra of 0.7% enrichment is displayed in Table 2. For these experiments, the data set contained 126 original spectra with a unique global counting statistics setting ( $2 \times 10^6$  counts in the whole spectrum). The impact of the counting statistics and detector energy resolution on the code performance with U spectra of 93% enrichment is summarized in Table 3 [38].

**Table 2:** Impact of detector energy resolution, 0.7% enrichment [38].

FWHM@185 keV	Mean relative deviation
830 eV	3.9 %
900 eV	5.7 %
1160 eV	8.9 %
1450 eV	6.2 %
1640 eV	17.7 %

**Table 3:** Impact of counting statistics, 93% enrichment [38].

Counting statistics, MCps	Net peak area@185 keV, counts	Mean relative deviation	
		FWHM@185 keV =780 eV	FWHM@185 keV =1210 eV
1	$2.3 \times 10^5$ (0.2 %)	23.3 %	9.3 %
3	$7.0 \times 10^5$ (0.1 %)	6.5 %	4.5 %
5	$11.6 \times 10^5$ (0.1 %)	2.0 %	1.5 %
7	$16.3 \times 10^5$ (0.1 %)	2.1 %	2.0 %
9	$21.0 \times 10^5$ (0.1 %)	1.0 %	1.8 %
10	$23.3 \times 10^5$ (0.1 %)	1.6 %	-
12	$28.0 \times 10^5$ (0.1 %)	1.3 %	-

A detailed assessment campaign of the IGA code performance with U spectra analysis is presented in the reference [38].

## 2.2. Recent developments in the domain of isotopic codes for MRGS systems

Over the past decade, there has been a vast progress and development in the domain of room temperature medium resolution detector technologies for various fields of ionizing radiation detection and measurement. Room temperature medium resolution detectors represent a trade-off between their room temperature operation and significantly better spectrometric performance compared to NaI detectors. Due to their attractive features, they become promising candidates for various fields of applied radiation detection and measurement. Among such fields are safeguards applications where room temperature operation and compact design makes CZT and LaBr<sub>3</sub>(Ce) advantageous candidates for a wide range of tasks. Determination of actinides isotopic composition is one of the tasks where room temperature medium resolution detectors are proposed as a feasible alternative to traditional HPGe systems.

In quantitative gamma-ray spectrometry if the photopeaks are sufficiently resolved a connection between the observed count rate in the detector with the decay rate of radioisotopes of interest can be made. The essential equations relating the measured net count rate in the photopeak to the activity of the isotope are given by Eq. (1)-(2):

$$\text{Decay rate}^j = -\lambda^j * N^j \quad (1)$$

$$NCR_i^j = \lambda^j * N^j * I_i^j * \varepsilon_i \quad (2)$$

Where  $NCR_i^j$  is the measured net count rate in the photopeak at energy  $i$  of radioisotope  $j$ ,  $\lambda^j$  is the decay constant for radioisotope  $j$ ,  $N^j$  is the number of atoms for the radioisotope  $j$ ,  $I_i^j$  is the gamma-ray emission probability at energy  $i$  from radioisotope  $j$  and  $\varepsilon_i$  is the detection efficiency at this energy. The quantity  $\varepsilon_i$  in Eq. (2) is the most difficult to determine as it depends on many different factors and parameters. Among them are the properties of the detector (its type, shape and size), gamma-ray energy, shape and size of the sample, degree of gamma-ray attenuation in the sample and on the way to the detector, presence of spectrum filters, measurement geometry and conditions (e.g. constant/variable background, acquisition time) and others.

As can be seen from Eq. (1) and (2), conceptually the mathematical formalism embedded in traditional codes optimized for HPGe detectors implies a rigorous coupling between a model used to describe the physical processes/interactions and a statistical tool used to analyze and interpret the corresponding experimental data (such as peak shape models, counting efficiency model etc.). The mentioned physical premise is fundamental for the algorithms widely used in safeguards applications for the determination of actinides isotopic composition. One of such algorithms is known as the *isotopic ratios* method with an intrinsically calibrated counting efficiency curve. Since it is based on a physical premise given by the proportionality of the observed signature photopeak net count rates to the decay rates of the corresponding radioisotopes, the key role is played by the physical models used to extract the information from the measured spectrum.

Although Pu spectra analysis using medium resolution quality spectra is currently challenging due to the significant peak overlapping when using the isotopic ratios method, analysis of U spectra is possible.

Recently, a code MCSIGMA was developed by [39]. It works with LaBr<sub>3</sub>(Ce) detectors and uses the gamma-ray peaks above 100 keV. The core processing algorithm of the code is based on fitting of the modeled net count rates along with the isotopic ratios and the relative counting efficiency model coefficients in a system of simultaneous equations. The minimization criterion is set to minimize the sum of the weighted difference between the modeled net count rates and experimental net count rates, which are determined from the spectrum. The code features an empirical model used to describe the relative counting efficiency in the 143-1001 keV energy range. Details on the code data processing and analysis routines are described in detail in reference [39]. Testing of the algorithm was carried-out using CBNM certified U standards with enrichment degrees from 0.31% to 4.46%. Results of this work demonstrated that using a 2x2 inch LaBr<sub>3</sub>(Ce) detector and 7200 s spectra acquisition time the enrichment determination accuracy is around 3% and less than 30% of relative uncertainty range for standards with <sup>235</sup>U content from 1.95% to 4.46%. For natural (0.71%) and depleted (0.31%) U samples the accuracy and uncertainty budgets indicated a strong influence of the statistical quality of <sup>235</sup>U gamma-ray signatures.

Among the crucial factors that impact the accuracy and uncertainty in medium resolution spectra of actinides when performing quantitative analysis of radionuclides is attributed not only to the statistical quality of the photopeaks, but also the selection of the region-of-interest (ROI) boundaries. The latter is of particular concern for asymmetrical photopeaks of CZT detectors. A comprehensive assessment of this question in connection to the counting time and corresponding critical limits for the net counts in photopeaks of interest was conducted in reference [40].

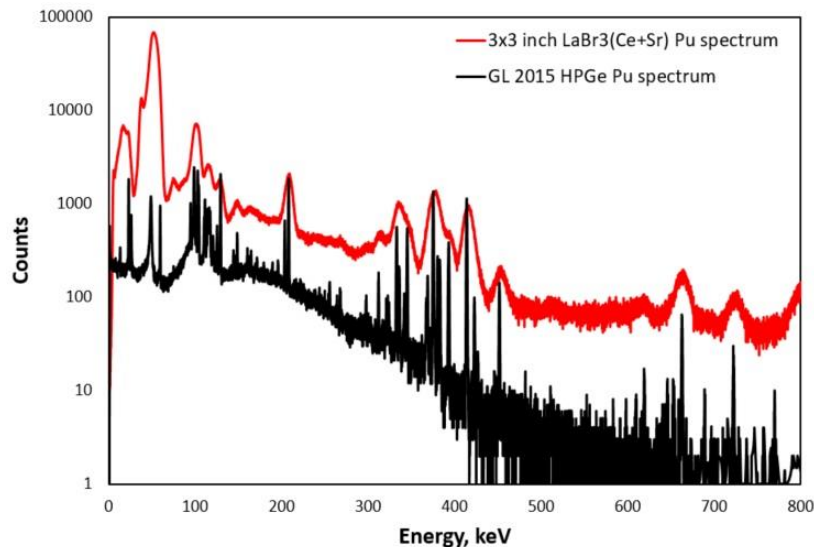
To overcome the challenges of the traditional net peak area-based algorithms application to medium resolution quality spectra analysis, intelligence-based methods for qualitative and quantitative information extraction can be used. Such methods imply a different conceptual design of the coupling physical model-statistical tool when extracting the information on the X- and gamma-ray signatures and treat spectrum using higher dimensionalities with respect to its features. The conceptual basis for such methods is that a spectrum is viewed as a layout of patterns corresponding to particular attribute characteristics of the sample assayed. In that respect, the traditional metrics of qualitative and quantitative information extraction can be avoided and replaced by analysis of the feature space and its functional behavior with respect to the attribute characteristics of the sample (e.g. U enrichment). Such methods can be based on the principal component analysis (PCA) and machine learning tools featuring neural networks (NN) to prepare the datasets, process, cluster and extract information from them.

Recently, such intelligence-based approaches were applied to medium resolution U spectra analysis, featuring the CAMILA code [41]. In this work, an architecture of a pattern recognition algorithm using NN was proposed as a tool to connect the observed spectral features of U spectra to the sample attribute characteristics (U enrichment). The code uses <sup>235</sup>U gamma-ray signatures in the 143-205 keV energy region and is based on built-in Matlab optimization and artificial intelligence toolboxes. Details on the code architecture and data analysis routines are described in detail in reference [41]. For performance assessment of the code, Monte Carlo simulated uranium spectra with enrichment degrees ranging from 0.5% up to 95%, different sample-to-detector distance and degree of attenuation through lead absorbers of variable thickness were used. The results of this work indicated that the performance of such an algorithm depends on the NN architecture and its parameters (such as the number of nodes in the hidden layer), number of training data sets used as well as on the feature vector space dimensions and spectra acquisition geometry. The achieved prediction accuracy for sample-to-detector distance and degree of attenuation recognition for data sets of nine U categories is of the order of 90%. Another approach realized in the CAMILA code is based on a PCA-based tool to cluster U spectra of different enrichment using same 143-205 keV energy region. Results of this work demonstrated that a PCA-based tool was capable to cluster U spectra of different enrichment and statistical quality and distinguish them from background radiation spectra. Tests were conducted using U spectra of CBNM standards with enrichment degrees from 0.31% up to 4.46% measured on a 500 mm<sup>3</sup> CZT detector [41]. Such an algorithm also allows to cluster spectra of different degrees of attenuation. The algorithms can be used for analysis if U spectra measured as on Cd-based as well as LaBr<sub>3</sub>-based detectors.

### 2.3. Research activities in the domain of detection technologies at CEA List

The domain of applied passive gamma-ray spectrometry remains important for a broad range of assay tasks. As a result, further progress and development activities represent an important subject of research. CEA List research activities in this domain deal with alternative ways for processing of plutonium spectra acquired with medium resolution detectors of different types. These types include LaBr<sub>3</sub> co-doped scintillators of different size, non-pixelated CZT detectors and pixelated Cd-based sensors.

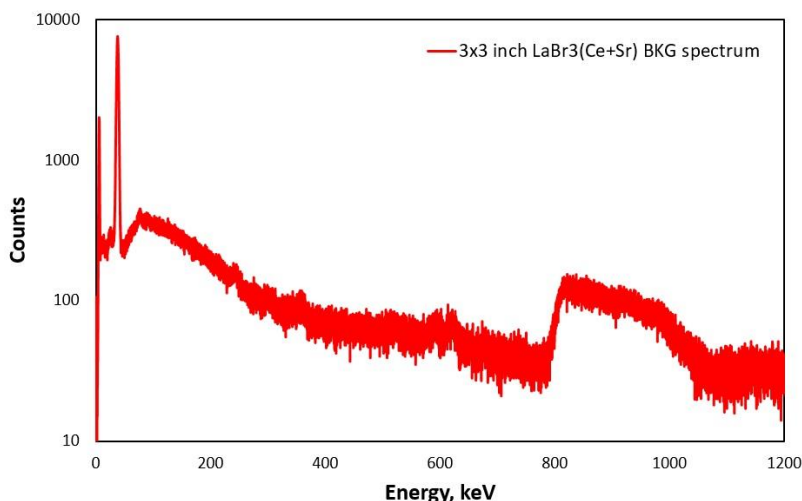
A comparison of Pu spectra (PIDIE 2 sample with 89% of <sup>239</sup>Pu) measured on a 3x3 inch LaBr<sub>3</sub>(Ce+Sr) and a GL 2015 HPGe detectors is shown in figure 3. As can be observed from this figure, the difference in energy resolution yields a dramatic impact on the X- and gamma-ray signatures overlapping.



**Figure 3:** Spectrometric performance comparison of 3x3 inch LaBr<sub>3</sub>(Ce+Sr) and GL 2015 HPGe detectors.

Moreover, in spectra measured on LaBr<sub>3</sub> detectors another spectrometric particularity arises. In La-halide based scintillators the self-activity of <sup>138</sup>La is observed, representing a major issue that reduces the detector sensitivity in low activity measurements and interferes with the gamma-ray signatures of interest in nuclear spectrometry applications, complicating the data analysis [42]. Natural Lanthanum is composed of two major radioisotopes: a stable <sup>139</sup>La with 99.91% abundance and radioactive <sup>138</sup>La with 0.09% abundance and a half-life of 1.05×10<sup>11</sup> years. <sup>138</sup>La radioisotope decays by an electron capture (e<sup>-</sup>) into <sup>138</sup>Ba with 66.4% probability and the remaining 33.6% decay by β<sup>-</sup> emission into <sup>138</sup>Ce. In both cases, <sup>138</sup>La decays into an excited state of the corresponding <sup>138</sup>Ce daughter nucleus which results in the emission of subsequent gamma-rays. For example, for the electron capture process, the 1436 keV gamma-ray along with 32-38 keV characteristic X-rays of Ba are emitted. In the case of β<sup>-</sup> emission, the 789 keV gamma-ray in coincidence with the continuum up to 255 keV can be observed in the measured spectra. Figures 4 indicate the mentioned particularities using a background spectrum measured on a 3x3 inch LaBr<sub>3</sub>(Ce+Sr) detector. Figure 4 indicates that these additional radiations impact the low-energy and the high-energy regions. The acquisition time for this spectrum was 1 hour. These particularities need to be taken into account when determining of the actinides isotopic composition using LaBr<sub>3</sub> detectors.

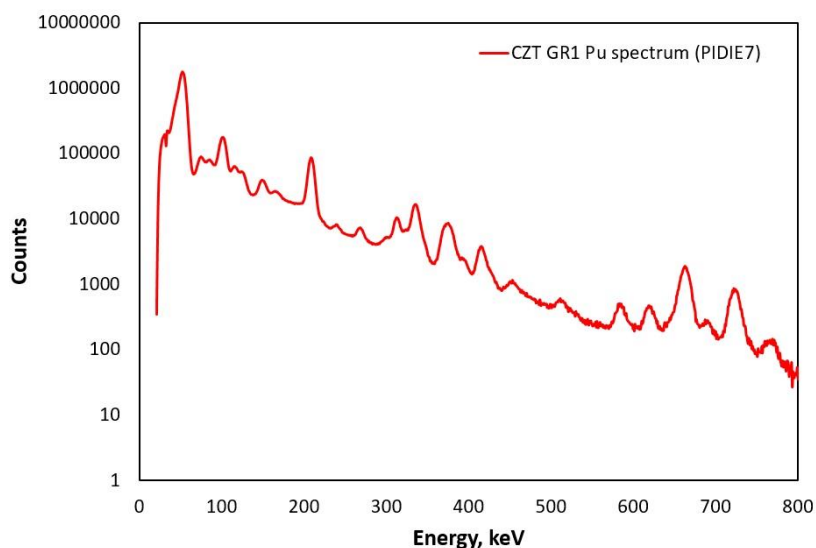




**Figure 4:** Background radiation spectrum measured on a 3x3 inch LaBr<sub>3</sub>(Ce+Sr) detector.

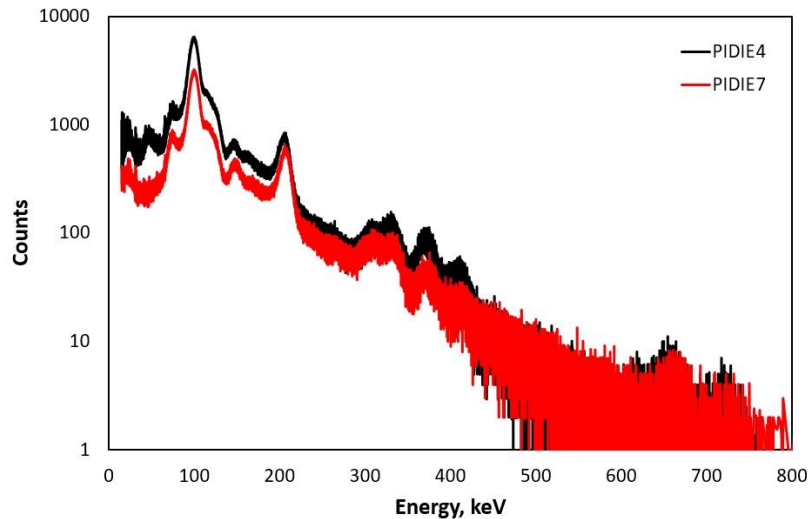
We would like to point out, that the degree of the observed self-activity impact on the spectra quality of La-based scintillators may vary as a function of the crystal size. Thus, the self-activity effects rate increases for crystals of large volume. For smaller crystals, a significant fraction of the X-rays can escape without detection.

Pu spectrum (PIDIE 7 sample with 65% of <sup>239</sup>Pu) measured on a CZT-based Kromek GR1 device is shown in figure 5. The energy resolution of this detector is stated at better than 2.5%@661 keV by the manufacturer [43]. The energy resolution of this detector is quite close to that of the presented above LaBr<sub>3</sub>(Ce+Sr) scintillator.



**Figure 5:** Plutonium spectrum measured on a CZT-based GR1 detector by Kromek.

However, thanks to the ongoing progress and development in the domain of Cd-based detector technologies other types of such detectors exist. One of such promising technologies is attributed to the pixelated CdTe-based sensor technology. At CEA List the CdTe-based Timepix3 pixelated detector is largely used for gamma imaging applications. The energy resolution of a 14x14 mm Timepix3 pixelated detector is 4.5%@661 keV [44]. Plutonium spectra of PIDIE 4 (with 78% of <sup>239</sup>Pu) and PIDIE 7 (with 65% of <sup>239</sup>Pu) samples measured on such a device are shown in figure 6.



**Figure 6:** Plutonium spectra measured on a TimePix3 CdTe detector.

As can be observed from figure 8, although with a pixelated CdTe detector technology the peak asymmetry is not pronounced, the energy resolution of a 14x14 mm Timepix3 pixelated detector unfortunately does not allow to resolve complex peak groupings in Pu spectra. Besides, figure 6 illustrates that with pixelated detectors the counting efficiency for an energy region exceeding 400 keV can be quite poor. Further research is required to investigate this matter.

Finally, a research direction devoted to development of spectra analysis tools based on likelihood maximization algorithms targets at processing of medium resolution spectra. Such an analysis tool represents a promising technology for various applied actinide analysis tasks using passive gamma-ray spectrometry [45]. It requires a combination of simulated and experimental spectra to solve for the relative isotopic composition of actinides. The research with likelihood maximization algorithms applied to medium resolution spectra analysis aims to complement the previous studies in the domain of isotopic composition determination algorithms by looking into the possibilities of determining of the Pu-isotopic composition using medium resolution quality spectra.

### 3. Summary

Over the years, intensive efforts have been made and are still going on to analyze spectra of uranium and plutonium materials, especially with respect to the determination of the isotopic composition. Currently, HPGe systems still represent the benchmark detectors which are widely used for such tasks. As a result, sustainability in the corresponding methodological supply of codes used for determination of actinides isotopic composition is important. One of such codes is the French IGA code intended to work with various different designs of HPGe detectors as for uranium as well as for plutonium spectra analysis. The code offers flexibility in terms of its parameters and is able to perform analysis of degraded quality spectra of uranium and plutonium materials.

However, rapid progress and development in the domain of room temperature detectors and the corresponding algorithms capable to analyze actinides spectra measured with such a medium resolution quality makes these detectors promising candidates for various spectrometric tasks. LaBr<sub>3</sub>(Ce) and CZT detectors are proposed as a feasible alternative to traditional HPGe and NaI detectors, allowing to address such disadvantages as the necessity of cooling, heavy, bulky instrumentation or low-energy resolution of NaI. LaBr<sub>3</sub>(Ce) and CZT detectors have demonstrated a promising potential for uranium enrichment determination tasks. Code MCSIGMA developed for LaBr<sub>3</sub>(Ce) detectors allows to use the isotopic ratios method with a fully automatic calibration of the relative counting efficiency curve. However, given the current state-of-the-art with LaBr<sub>3</sub> detectors, analysis of plutonium spectra measured on LaBr<sub>3</sub> detectors using net peak area based methods is currently challenging.

Another promising direction of research is devoted to development of algorithms capable to analyze spectra of medium resolution detectors that are based on intelligent non-net peak area based methods. Among such methods are principle component analysis, neural networks and likelihood-based

maximization algorithms. Such algorithms treat spectra using higher dimensionalities and can be fully disengaged from the counting physics.

#### **4. Acknowledgements**

The authors would like to thank Hamid Makil for preparation of plutonium samples used in the measurement campaign, Guillaume Amoyal for plutonium spectra measured on the Timepix3 sensor, Victor Spielmann for measured plutonium spectrum on a CZT GR1 detector and Romuald Woo for fabrication of Cu attenuators used in measurements with plutonium samples.

#### **5. Keywords**

Isotopic composition; IAEA safeguards; medium resolution; IGA; MCSIGMA; CAMILA

#### **6. Author biography**

Iaroslav Meleshenkovskii presently works as a research engineer at CEA List Saclay Center. Iaroslav Meleshenkovskii holds a master's degree (2015) in nuclear power engineering and thermal physics from Moscow Engineering Physics Institute and a doctoral degree (2019) in engineering sciences and technology from Université libre de Bruxelles. His work in the domain of nuclear metrology started in 2013 and is focused on measurement methods and instrumentation for active/passive non-destructive assay of nuclear materials. He developed methods for processing of medium resolution spectra. He is the author of the MCSIGMA and CAMILA codes.

Frédéric CARREL graduated from the ENSICAEN engineer school in 2004. His PhD (2007) was dedicated to the characterization of nuclear waste packages using photon or neutron active interrogation and delayed gamma-ray spectrometry. He worked for AREVA CANBERRA in 2008 and has been hired by CEA List the same year. His research topic is related to nuclear measurements (passive and active neutron measurements, gamma-ray spectrometry using various detectors, use of plastic scintillators, gamma imaging) and associated Monte Carlo simulation (MCNP code). He is part of the development team of the GAMPIX gamma camera. He is CEA senior expert in nuclear measurements from 2017. He is the Head of Sensors and Electronic Architectures Laboratory from the beginning of 2019.

Anne-Catherine Simon is a research engineer at CEA List Saclay. After an experience in gauge design for nuclear instrumentation, she specialized in the development of gamma and X-ray spectrometry computer tools for radionuclide identification and isotopic composition determination. She also developed and implemented methods for processing spectrometry data sets and images. She has been working in X-ray medical imaging and dosimetry for radiation therapy since 2016.

Isabelle Espagnon is a researcher at CEA List Saclay. She has a doctoral degree in nuclear physics. She has worked on methods for processing spectrometry data sets and images for many domains including biology. She has developed several computer tools for radionuclide identification and isotopic composition determination based on gamma and X-ray spectra.

Anne-Catherine Simon and Isabelle Espagnon are the authors of the IGA code.

#### **7. References**

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