Implementation of an experimental design to evaluate the codes used to determine the enrichment of uranium samples

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Abstract

Gamma spectroscopy is commonly used in nuclear safeguards to measure the enrichment of uranium applying either the enrichment meter method, or analysis codes developed to analyse several γ-rays or X- and γ-rays in defined regions of a uranium spectrum. An experimental design has been carried out for the measurement of uranium enrichment using this technique with different software used in safeguards applications: MGA, MGA++, PCFRAM and IGA. The main goal is to improve the estimates upon the uncertainties of the measurement taking into account realistic in situ acquisition condition.

To set up the experiment, uranium sources have been achieved. It is $U_3 0_8$ reference material in powder form, with a wide enrichment range in sealed container. Regarding the acquisition systems, several gamma spectroscopic germanium detector (planar, coaxial) and multichannel analysers have been used. Five entry parameters have been considered for the experimental design: ^{235}U enrichment, adjustment of the acquisition gain, source-detector distance, shielding, and matrix effect. With all these spectra, there is a direct comparison between the uncertainty announced by the different software and the difference between the computed enrichment value and the value of the reference material. The paper describes the methodology implemented to evaluate the results of such an experimental design, and exhibits the results obtained when applying the code MGAU, with the objective to confirm or to improve our current knowledge of uncertainties for these uranium enrichment measurements. The evaluation of the 3 other codes, which is currently in progress, will be subsequently published.

1. Introduction

Gamma spectroscopy is commonly used in nuclear safeguards to measure the enrichment of uranium samples applying either (1) the traditional enrichment meter method based on the counting of the 185.7 keV γ-ray of ^{235}U, or (2) analysis codes developed to analyse several γ-rays, or X- and γ-rays present in defined regions of the acquired uranium spectrum, without the need for any calibration. The isotopic composition analysis codes MGA [1], MGA++ [2], PCFRAM [3] or IGA [4], which analyze γ emission spectra of uranium or plutonium, produce results accompanied by an uncertainty whose dominating component has a statistical origin. The experience feedback at IRSN as well as the conclusions reported in [5] and [6] had emphasized situations of measurement where calculated uncertainties are not representative of the real bias made by the code. To address this concern, IRSN has carried out an experimental design for the measurement of uranium enrichment with the previous analysis codes in order to better estimate the measurement uncertainties taking into account realistic in situ acquisition conditions.

The paper first reports on the experimental setup, describing the characteristics of uranium reference samples especially achieved, the implemented acquisition systems, the parameters of the experimental design (^{235}U enrichment, adjustment of the acquisition gain, source-detector distance, shielding, and matrix effect), and the measurement procedure. With all these spectra, there is a direct comparison between the uncertainty announced by the different software and the difference between the computed enrichment value and the value of the reference material. The methodology developed in order to evaluate the results of the experimental design is presented in a second part. It is a calibration process which consists in a first step of determining a statistics-based criterion to stop each acquisition once the result has converged, evaluating the dominating parameters of the experimental design in a second step and finally implementing calibration tables with the objective to confirm or to improve our current knowledge of uncertainties for these uranium enrichment measurements. The last chapter exhibits the entire process of calibration of the code MGAU based on the current approach.

2. Experimental Setup

2.1. Material for the experimental design

To set up the experiment, standard trioxide uranium U_3O_8 sources have been achieved. The U_3O_8 powder is packaged in cylindrical containers made of resin. Enrichment range varies from depleted uranium (0.3%) up to highly enriched uranium (90%) and is certified by mass spectrometry (characteristics given in the table 1). The sources are also certified in uranium content and mass composition and sealed in conformity with the standards NFM61-02 (mechanics) and NFM61-003 (sealing). Three types of geometries, with the same resin bottom thickness of 2mm, have been designed in order to have a good compromise between the gamma emitting surface and the available mass for the manufacturing: the A type capsule (DxH: 38x5mm[1]) containing around 10g of uranium, the B type capsule (DxH: 50x5mm) containing between 12g and 33g of uranium and the C type capsule (DxH: 48x20mm => 50x33mm) containing between 86 and 120g of uranium.

Source	Capsule type	U mass (g)	^{234}U (%)	^{235}U (%)	^{236}U (%)	^{238}U (%)
8426-1	B	16.77 (0.32)	0.003 (0.001)	0.341 (0.01)	< 0.001	99.656 (0.012)
8426-2	B	15.3 (0.32)	0.004 (0.001)	0.539 (0.01)	< 0.001	99.457 (0.012)
8426-20	C	120.9 (0.36)	0.006 (0.001)	0.714 (0.01)	< 0.001	99.28 (0.012)
8426-4	B	14.68 (0.36)	0.011 (0.001)	1.532 (0.01)	0.002 (0.001)	98.455 (0.012)
8426-5	C	96.50 (0.30)	0.023 (0.001)	3.038 (0.01)	< 0.001	96.939 (0.012)
8426-6	B	12.46 (0.32)	0.035 (0.001)	5.49 (0.01)	< 0.001	94.475 (0.012)
8426-7	B	14.60 (0.32)	0.079 (0.001)	7.055 (0.01)	< 0.001	92.866 (0.012)
8426-8	B	32.62 (0.36)	0.076 (0.001)	11.304 (0.036)	0.021 (0.001)	88.599 (0.012)
8426-9	B	12.87 (0.36)	0.105 (0.001)	14.21 (0.036)	< 0.001	85.685 (0.012)
8426-10	A	8.73 (0.5)	0.147 (0.001)	21.902 (0.036)	< 0.001	77.951 (0.012)
8426-16[2]	B	13.82 (0.32)	0.0171 (0.001)	26.075 (0.036)	0.006 (0.001)	73.748 (0.012)
8426-11	C	87.45 (0.30)	0.299 (0.001)	29.187 (0.036)	0.004 (0.001)	70.51 (0.012)
8426-12	A	8.5 (0.32)	0.68 (0.001)	45.112 (0.036)	< 0.001	54.208 (0.012)
8426-13	A	9.88 (0.32)	0.514 (0.001)	57.042 (0.036)	0.001 (0.001)	42.443 (0.012)
8426-14	A	12.34 (0.32)	0.427 (0.001)	68.043 (0.036)	0.001 (0.001	31.529 (0.012)
8426-15	C	86.04 (0.30)	1.1 (0.001)	89.303 (0.036)	0.01 (0.001	9.588 (0.012)

Table 1: certified characteristics of the sources

Three sets of γ acquisition systems, containing one or more high purity germanium detectors and electronics but the same acquisition and γ rays treatment software were implemented to realize the experimental design taking into account the three configurations routinely used in the field for uranium or plutonium isotopic composition measurements.

- The first set “UPu Inspector”, dedicated to the direct measurement of the isotopic composition of uranium with the code **MGAU** (version 2.2 provided by Canberra), consisted of a planar detector (“GL1015R” or “BE1015R” from Canberra), a multi channel analyser “Inspector” or “Inspector2000” from Canberra and the software “Genie 2k 2.0”.
- The second set “MGA++”, dedicated to the direct measurement of the isotopic composition of uranium using the code **U235view** (version 1.06 provided by Ortec), consisted of a planar detector (“GL0515R” or “GL1015R” or “CPL5” from Canberra), a multi channel analyser “DSPEC” or “DSPP” provided by Ortec and the software “Gammavision 6.01”.
- The third set “FRAM”, dedicated to the direct measurement of the isotopic composition of uranium using the code **PCFRAM** (version 4.2 provided by LANL, with the parameter set “U121_1001Coax.pst”), consisted of a coaxial detector “GEM25175” from Ortec, a multi channel analyser “DSPEC” provided by Ortec and the software “Gammavision 6.01”.

The spectra acquired in the three configurations are available for the evaluation of the code IGA, which is able to analyse any kind of spectrum – as the user only needs to adjust the energy in the first and the last channel and the FWHM and channel of a peak of interest. In the same way, the spectra acquired with the planar detectors are available for the evaluation of the code PCFRAM using the parameter sets “U100keVLEU” and “U100keVHEU” for the uranium analysis of the planar detector spectra employing the 100 keV region.

The previous γ spectrometry systems are based on the acquisition of a γ emission spectrum for which it is useful to follow the evolution of measured enrichment versus elapsed time so far, as since the result of measurement and its uncertainty tend towards a limiting value.

[1] D x H = internal Diameter x U_3O_8 filling Height; [2] Source U_3O_8 + CaF_2

Thus, the analysis of spectra acquired during a measurement by the codes MGA, MGA++, PCFRAM or IGA is coupled with the self-incrementing software "AutoISO_PLUM" [7], developed at IRSN in order to drive a self-incremented γ spectrometry acquisition according to time, to visualize in real time on a graph the evolution of the measured quantity, which can be for instance the isotopic composition of uranium calculated by one of the 4 codes previously quoted, and to control indicators such as the counting statistics in the spectrum (cf. figure n°1). For this purpose, the user selects the number of spectra to be saved and analysed during a measurement, the measurement type, the code to be used, and the time interval between two successive acquisitions which can be cumulated or repeated. This tool helps the user to follow the convergence of a measurement, in order to stop it when there is no more significant improvement of the result and its precision with the counting time. All the relevant data accessible in the report issued by the code (live time, total counting in the spectrum, ^{234}U, ^{235}U and ^{238}U contents and associated uncertainties for MGAU) calculated for each time step are stored in an Excel file that can easily be exploited.

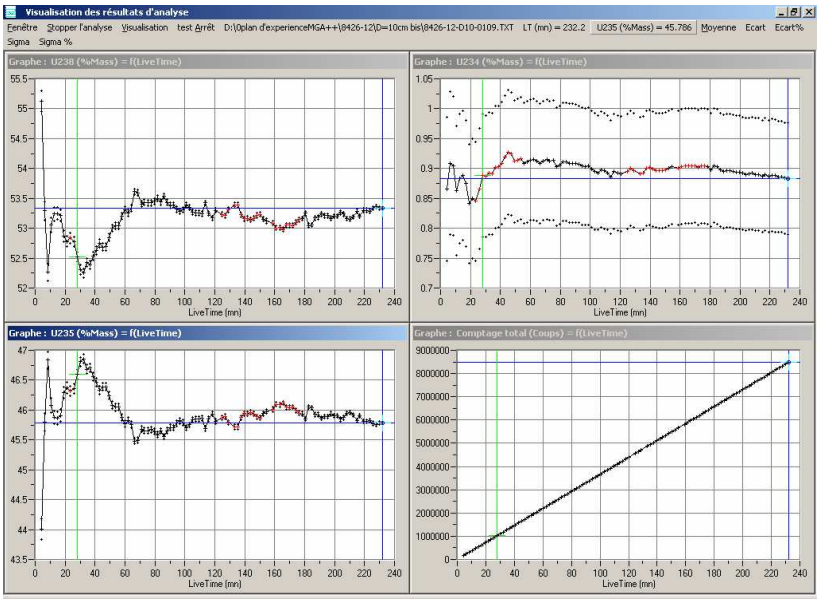

Figure n°1 : example of behaviour of the isotopic abundances of uranium (^{235}U, ^{235}U, ^{238}U) and the statistics in the spectrum according to the measurement time

The developer of the code provides a simple criterion to decide when to stop the acquisition, based on the total count in the spectrum with a recommended value greater than 10^6, which is just a general indication. The measurements performed in the scope of the experimental design have been voluntarily acquired on a long time period above this criterion in order to study the influence of statistics on the results and to determine a reliable criterion for the convergence of the results.

2.2. Parameters for the experimental design

The purpose of the experimental design is to highlight the factors which influence the result given by the software at the end of a measurement, and to determine their relative importance. The following parameters have been chosen in order to fulfil – as accurately as possible – realistic on-site measurement configurations:

- **^{235}U enrichment**: 16 ^{235}U enrichment values, ranging between 0,35% and 89% (cf. table 1) ;
- **Source-detector distance**: Two distances are considered: 10 cm (chosen in order to keep measurements away from the sum peaks phenomenon; this distance may vary depending on the dead-time of the acquisition system) and 45 cm (distance at which one generally stands when one deals with waste drums);
- **Shielding**: interposition of a screen between the source and the detector in order to evaluate the influence of the container. Measurements are taken with 2 thickness stainless steel screens: 2mm, and 6mm, which correspond to conditions really met in the field;
- **Effect of the operator**: The operator effect is simulated by slightly varying the adjustment of the acquisition gain of the measuring equipment, i.e. by modifying the gain so that the peaks of stronger energy are shifted of more or less 3 channels compared to their normal position. In the case of MGA and

MGA++, the peak with 185,7 keV is shifted of more or less 3 channels (configurations 1 and 2). In the case of FRAM, the peak with 185,7 keV is shifted of 3 channels (configuration 1) and the peak with 1001 keV is shifted of 3 channels (configuration 2).

- **Matrix effect**: verifications of ^{235}U enrichment for safeguards purposes are usually done on uranium-bearing products (UO_2, U_3O_8,…) as well as waste drums containing uranium in suspension in vinyl or metal matrices. Therefore the influence of this parameter is studied using 200 litres "mock-up" drums available in the laboratory, containing vinyl matrices of density 0.2g/cm3 (average case generally encountered) and 0.4g/cm3 (penalizing case, also met on site), in the centre of which a source of given enrichment is put down.

The output parameters, measured and studied are the **uncertainty** given by the software and the **discrepancy** between the certified enrichment of the source (reference material) and the value given by the software.

2.3. Measurement configurations

The experimental design represents an overall set of 224 measurements, 14 measurements being performed for each source (i.e. each enrichment) as stated in table 2 below. Depending on the measurement configuration, one measurement can be performed on a time period varying from 1 or several hours (for the basic configuration) up to several days (for the sources introduced in the mock-up drum), with a time step between each acquisition varying from 2 minutes up to 1 hour.

The configuration which involves a source alone placed at a distance of 10 cm from the front face of the detector is considered as the basic one.

Source	screen (mm)	Distance (cm)	Matrix (g/cm^3)	Gain
1	0	10	0	1
1	0	10	0	2
1	0	10	0	3
1	2	10	0	1
1	2	45	0	1
1	6	10	0	1
1	6	45	0	1
1	6	10	0	2
1	6	10	0	3
1	0	45	0	1
1	drum thickness	45	0,2	1
1	drum thickness	45	0,4	1
1	drum thickness	45	0,4	2
1	drum thickness	45	0,4	3

Source 1 : $^{235}U/U_{total}$ = 0,35%

Source/detector distance :
1. 10 cm
2. 45 cm

Thickness of the screen :
1. 2 mm
2. 6 mm

Gain of the acquisition system :
1. usual adjustment
2. 185 keV shifted of +3 channels
3. 1001 keV shifted of -3 channels
   (or 185 keV shifted of -3 channels)

Matrix density :
1. 0,2 g/cm^3
2. 0,4 g/cm^3

Table 2: acquisitions of the experimental design for each enrichment

Some repeatability measurements have also been performed in the basic configuration (source alone at a distance of usually 10 cm from the detector) in order to check the uncertainty given by the code according to its reproducibility.

3. Methodology of the evaluation

We propose hereafter to evaluate the performances of a code by performing a calibration which takes into account realistic in situ acquisition conditions as defined in the experimental design. The evaluation is executed in three successive steps. The first step consists in developing a statistics-based criterion to stop each acquisition at the beginning of the convergence according to the measurement time, in order to have an uncertainty which is representative of the result. The second step consists in analyzing the sensitivity of the measurement results (Comparison between the actual difference between the enrichment obtained with the code and the reference value and uncertainties also given by the code), compared to the variations of the 5 parameters of the experimental design. The third step consists in developing calibration tables which improve the enrichment values and the uncertainties given by the code.

3.1. Methodology used to develop the stopping criterion

The objective here is to detect, for each measurement, the region where the acquisition starts to converge according to the ^{235}U enrichment value. For this purpose, a criterion named "relative standard deviation" has been developed, which consists of calculating the variation of the results (^{235}U enrichment values) on N successive points of the data set which is analyzed. The relative standard deviation of this data set of N

points is, for example at the analysed point P_i (where $\sigma()$ is the standard deviation and $A()$ is the average):

$$\frac{\sigma(P_i, P_{i+1}, ..., P_{i+N-1})}{A(P_i, P_{i+1}, ..., P_{i+N-1})}$$

This relative standard deviation is then compared to a threshold value S. If this relative standard deviation is less than the threshold value, then the first point of this data set is taken as the stopping value. The corresponding ^{235}U enrichment value will be recorded as the result of the measurement.

The values of parameters N and S are defined following a dichotomy procedure for optimizing them, starting with a number of points N = 15 (one chooses a high number in order to obtain the most restrictive possible criterion) and trying to seek the threshold value S starting with boundary values of 0.001 and 0.05.

3.2. Methodology used to evaluate the dominating parameters

The knowledge of the parameters which mostly influence the measurements governs the implementation of the calibrations for the improvement of the results: one should act on the dominating parameters to correct the values provided by the codes. Two methods are applied, which should lead to the same conclusions: a statistical one and a probabilistic one.

The statistical method consists in assuming the existence of a linear relation between the input variables and the output variables. For each output variable (Y), we consider that:

$$Y = a_0 + \sum_i a_i X_i + \varepsilon$$

where a_i is the coefficient of the linear approximation for the input variable X_i and ε is the error between real results (Y) and those obtained by linear approximation. The coefficients a_i are found by using the least-square method to minimize the error represented by ε. The coefficients of the linear approximations thus contain information about the impact of each input data on the quantity studied: the higher this coefficient is, the stronger the influence of the parameter is. The coefficients are not compared just as they are: it is necessary to normalize them before, in order to take into account the range of the variations of the input parameter compared to those of the output parameter. The standardization corresponds to the following change (where $\sigma()$ is the standard deviation):

$$a_i^n = \frac{a_i \times \sigma(X_i)}{\sigma(Y)}$$

The Probabilistic method consists in calculating the variance of the results obtained in a specific configuration (for example all the measurements carried out without screen, then with a screen of 2 mm, then with a screen of 6 mm, if we consider the screen parameter). We obtain conditional variances which are divided by the variance calculated on the full result bank of the experimental design, which makes it possible to compare them. Thus, the higher the conditional variance, the more sensitive to the studied parameter the software is (it gives sometimes a weak variation, sometimes a strong variation).

The statistical method gives absolute information: one knows that that some parameter has an influence on the result, but one doesn't know if it is the case under all the conditions of measurement or some in particular. This method must be used at first in order to detect the dominating parameters. Then, the probabilistic method has the advantage of being more flexible than the statistical method: one can test all the configurations of interest (i.e. to implement classes according to the enrichment and the matrix, the enrichment and the screen, etc…). Moreover, we do not make any assumption here: we take account of the true values. That gives more precise results: one knows which parameters are influential and under which conditions. The two methods are complementary.

3.3. Methodology used to improve the results (enrichments and uncertainties)

The implementation of calibration tables gives an interesting overview of the performances of a code depending on the measurement configuration, by considering both ^{235}U enrichment value obtained and the uncertainty provided by the code.

The calibration tables of the ^{235}U enrichment measured values are developed by gathering measurements in homogeneous categories (i.e by distinguishing from the others the measurement conditions defined by the dominating parameters). The average of the difference between the measured ^{235}U enrichment value and the reference value for each measurement is calculated inside each category. This mean value represents

the correction to be applied to current measurements under the same conditions, which corresponds in more a general way to the observed bias.

The calibration tables of the uncertainties are developed by gathering the measurements in the same way. The average of the difference between the relative uncertainties given by the code and the relative errors actually made by the code on the true value determined for each measurement is calculated inside each category. This mean value represents the correction to be applied to the relative uncertainties currently provided by the code under the same conditions, which is a signature of the representativity of the announced uncertainties.

3.4. Methodology used to evaluate the influence of the counting statistics on the result

The stopping criterion defined above is set up on the basis of the evolution of the result according the measurement time. Its consistency is checked by reprocessing the same statistical analysis for the time T/2 and 2T with T being the stopping criterion time. However in practice, measurement times of more than 30 to 60 minutes are rarely allocated for safeguards verifications.

The analysis of the connections between the statistics in one or several peaks of interest and the measurement results (^{235}U enrichment and uncertainty) constitutes another possible track for the construction of an objective and physics-based stopping criterion. The point consists in determining if the convergence zone corresponds to similar counting statistics for the peaks studied by measurement category, then potentially developing a law which makes the evaluation of the expected precision possible in a given measurement configuration, for weaker statistics.

4. Results obtained with the code MGAU

The following paragraphs exhibit the results of the evaluation and calibration process of the code MGAU, applied to the 153 measurements of the experimental design performed with the "UPu Inspector" γ spectrometry system. Only 153 measurements were taken (whereas 224 where planned), as of the first tests it appeared that the parameter gain did not have impact on the results. Measurements to 45 cm with a screen of 2 mm were not carried out; the impact of the screen is appreciated through other measurements.

4.1. Stopping criterion

The parameters N (number of cumulated acquisitions) and S (threshold) implemented for the application of the stopping criterion to the "UPu Inspector" γ spectrometry system are given in table 3. It was not possible to develop a single criterion for all of the measurements: the behaviour of the measurement system is different in the presence of a screen or of a matrix, and according to the ^{235}U enrichment of the source.

no screen no matrix	with screen no matrix	with screen with matrix
$\%^{235}U < 1\ \%$ N = 14; S = 0.008		
$\%^{235}U > 1\ \%$ N = 14; S = 0.001	$\%^{235}U > 1\ \%$ N = 14; S = 0.0025	$\%^{235}U > 1\ \%$ N = 14; S = 0.0018

Table 3: parameters of applicability of the Relative Standard Deviation criterion

Once the criterion is satisfied, the acquisition converges towards a ^{235}U enrichment value. Under these experimental conditions, we will be able to qualify the uncertainties.

4.2. Repeatability measurements in the basic configuration

The acquisitions were performed for each of the 16 sources in the basic configuration with counting statistics taken in the convergence region of each basic measurement. The synthesis is presented in table 4.

The enrichment value given by MGA is stable from one measurement to another whatever the ^{235}U enrichment is (variance < 1%). It is the same for the announced uncertainties. The relative differences between measured and certified enrichment vary between -8% min and 15% max for the depleted or natural uranium and between -4% min and +5% max for enriched uranium. The uncertainty given by the code seems to cover the real error committed by the code in the majority of cases, considering a value of around 4-8% for depleted and natural uranium and a value of around 1% for enriched uranium (cf. table 4). As stated in ref. [5], our measurements shows a tendency to overestimate the enrichment for depleted uranium, but the uncertainties are quite high in our study (cf. "%Δ>0" column in table 4). Most of the measurements done with high enriched uranium (>20%) underestimate the true value, but it is not systematic, perhaps it is due to the measurement precision which is only around 1-2%.

4.3. Evaluation of the performances of the code (uncertainty components)

The dominating parameters for the relative differences between measured and certified enrichment are the enrichment and the matrix. According to the boundaries chosen for the screen thickness, its influence is less important. The figures 1 to 3 exhibit the difference between the measured enrichment and the certified value, for the optimal configuration with the best precision (figure 2), for a configuration with a stainless steel screen put between the sample and the detector (figure 3), and for a configuration with a mock-up drum (figure 4).

% ^{235}U	Nb of Acquisitions	Average value of abs(Δ)	Average value of Unc.	% (Unc > Δ)	% (Δ>0)
0.341	86	5.8	8.7	80%	88%
0.539	32	6.0	4.5	31%	100%
0.714	76	4.0	7.0	86%	63%
1.532	42	2.7	1.3	14%	100%
3.038	31	0.6	0.9	77%	48%
5.49	52	1.7	0.9	13%	98%
7.055	289	0.9	1.1	66%	68%
11.304	12	0.6	0.7	67%	8%
14.21	176	0.8	0.9	65%	25%
21.902	36	0.4	0.6	81%	67%
26.075	224	1.0	1.2	68%	34%
45.112	21	1.3	1.6	62%	43%
57.042	97	0.6	0.8	67%	43%
68.043	191	1.0	1.1	63%	28%
89.303	104	0.8	1.7	95%	81%

Δ = relative discrepancy between the measured ^{235}U enrichment and the reference value (%); Unc = code relative uncertainty (%)

Table 4: repeatability measurements synthesis

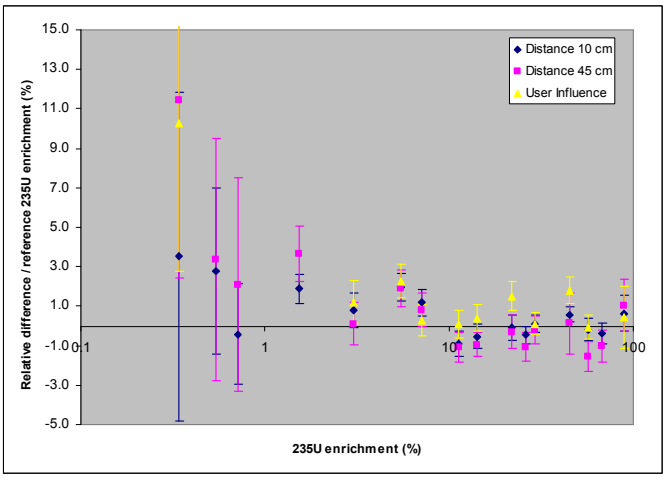

Figure 1: optimal configuration, distance and user effect

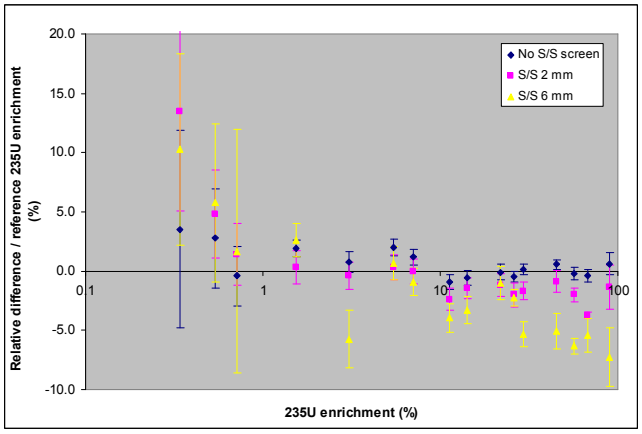

Figure 2: optimal configuration, stainless steel screen effect

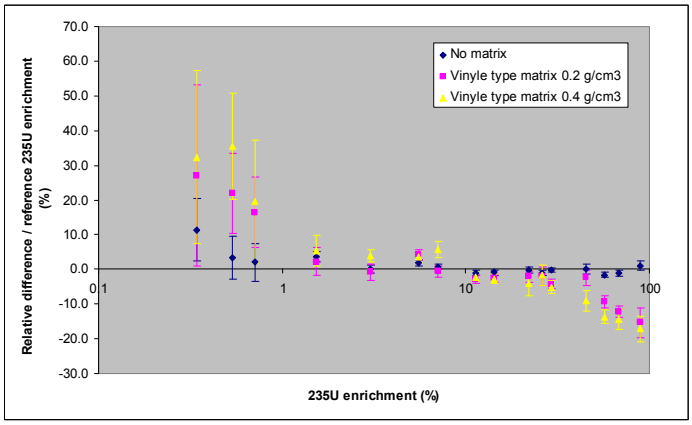

Figure 3: 45 cm configuration, waste drum effect

Regarding ^{235}U enrichment in the optimal configuration, as expected the best accuracy of less than 1% is obtained in the middle of the range around 10%, whereas depleted uranium is the most difficult to evaluate (around 3%). The measurement uncertainties are weaker at the extremes, even if it is much less pronounced for high enriched uranium. The presence of a screen between the source and the detector has a limited influence on the ^{235}U enrichment, which is in agreement with the measurements reported in ref. [5]. The screen effect is observed with a stainless steel absorber of thickness 6 mm, for high enriched uranium > 30%, which causes a discrepancy of around 3-4% compared to the optimal configuration. The matrix is the most influent parameter, which results in discrepancies of more than 15% at the extremes of the range.
The distance and the operator adjustment of the gain do not have any influence on the results, considering the boundary values chosen for our study (figure 2). The tendencies of overestimation of depleted and natural uranium and underestimation of high enriched uranium already pointed in reference [5] seem to be confirmed by the shapes of the figures especially when a screen or a matrix is interposed between the source and the detector.

The application of the calibration methodology described in the third chapter, implemented with the results issued from the application of the stopping criterion previously defined, leads to the values exhibited in the tables 4 (enrichment corrections) and 5 (uncertainty corrections), which distinguish 4 enrichment classes (depleted and natural, low enriched uranium up to 10%, enriched uranium between 10 and 50%, and high

enriched uranium from 50% up to 90%) and 3 separating measurement configurations: the optimal one without any absorber, the configuration with a stainless steel absorber and the configuration of a waste drum with different matrixes. The values presented in table 4 have been converted to relative values compared to the median value of the enrichment in each class considered, for representativity reasons.

measurement conditions	0 to 1%	1% to 10%	10% to 50%	50% to 90%
no screen no matrix	4,4%	**0,6%**	**0,2%**	**0,0%**
with screen no matrix	2,0%	**-1,1%**	**-2,6%**	**-3,8%**
with screen with matrix	25,4%	**1,9%**	**-3,1%**	**-14,4%**

Table 4 : calibration table for the measured ^{235}U enrichments

measurement conditions	0 to 1%	1% to 10%	10% to 50%	50% to 90%
no screen no matrix	3,84%	-0,26%	0,16%	0.92%
with screen no matrix	13.66%	-0,12%	-1.21%	-0.40%
with screen with matrix	-1.81%	-0,21%	-0.87%	-10.14%

Table 5 : calibration table for the uncertainties

The values given in table 4 should represent the correction to apply to measured enrichments, after convergence, in order to improve the result compared to the certified value. Only the bold values are reproducible with a measurement time multiplied by two with reference to the time given by the stopping criterion, which means that the criterion needs to be improved for depleted and natural uranium. The corrections to apply to the relative uncertainties given by the code at the convergence time, calculated in table 5 using the results issued from the stopping criterion give an idea of the representativity of those uncertainties at a given moment (underestimation for negative values), but they are probably not representative in all cases. The definition of a stopping criterion using statistics concepts only, based on the shape of the behaviour of the measured value according to time, does not seem to be the most appropriate approach to evaluate the expected uncertainties. The study is currently moving towards the evaluation of the evolution of a result according to the statistics and/or the shape of the peaks of interest used by the code to analyse a spectrum, that is to say the definition of a more physics-related stopping criterion.

5. Conclusions

The paper reports on an experimental design that has been performed on U_3O_8 standard for the determination of ^{235}U enrichment. The experimental design represents an overall set of 224 measurements, 14 mesurements being performed for each U_3O_8 standard taking into account the five following influencing factor: ^{235}U enrichment, source-detector distance, shielding, effect of the operator, matrix effect. Three sets of γ acquisition systems have been implemented.

The main goal of such an experimental design is the mastery of the uncertainties and a good knowledge of the fields of applicability of the isotopic composition determination codes for a better use on site. The originality of this experimental work lies in the approach used to evaluate the performances in several realistic in situ acquisition configurations, based on incremented acquisitions according to measurement time, and a calibration process to treat the experimental results and assess the performances that can be reached under several statistical conditions.

The first results of the statistical analysis, which is reported in this paper, have been applied to the experimental data obtained with the code MGAU with the objective (1) to develop and validate the methodology, and (2) to demonstrate it on this code. The results issued from the code MGAU confirm some of the tendencies already pointed in reference [5] and highlight the limitations of the code for waste drum assay. The evaluation of an adequate stopping criterion for an incremented measurement needs to be further studied. Once the analysis method is validated, it will be used for the other codes.

6. References

[1] R. Gunnink et al., *MGAU: a new analysis code for measuring U-235 enrichments in arbitrary samples*, URCL-JC-114713, IAEA Symposium on International Safeguards, Vienna, March 8-14, 1994

[2] T.Twomey et al., *Recent Developments of the MGA++ Codes*, ESARDA 21th annual meeting, Sevilla 1999

[3] TE. Sampson, TA. Kelley, *PC/FRAM user manual version 4.2*, LA-UR-99-998, 1999

[4] AC. Simon et al., *A new computer code for the determination of the isotope composition of actinides by X and γ ray spectrometry and its applications*, European Nuclear Conference, Versailles 2005

[5] AN. Berlizov et al., *Performance testing of the upgraded uranium isotopics multi-group analysis code MGAU*, NIM Section A, 575 (2007) 498-506

[6] *Proceedings of the international workshop on gamma evaluation codes for plutonium and uranium isotope abundance measurements by high resolution gamma spectroscopy: current status and future challenges*, ITU Karlsruhe, November 14-16, 2005

[7] JL. Dufour et al., *AutoISO : a new acquisition and data review software for the use of isotopic composition analysis codes (MGA++, PCFRAM and IGA*, ESARDA Bulletin n°35 (2006), 2-9