ABACC's Experience on using Medium Resolution Gamma-ray Spectrometry Systems based on Lanthanum Bromide Detectors for Uranium Enrichment Measurements

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ABSTRACT

Verification of the declared ^{235}U enrichment, or simply enrichment, is routinely performed by nuclear safeguards inspectorates during on-site inspections at bulk handling facilities, in particular commercial fuel fabrication, conversion and enrichment plants. Non-destructive assay (NDA) methods have been used for several years for prompt obtention of results whenever their performances are able to fulfill pre-established acceptance criteria. For decades the most frequently used methods were based on high- and low-resolution gamma-ray spectroscopy (HRGS or LRGS) using high-purity germanium (HPGe) semiconductor and sodium iodide (NaI) scintillation detectors, respectively. Since 2016, Cerium-doped Lanthanum Bromide ($LaBr_3(Ce)$) scintillation detectors have been tested and approved for regular use during safeguards inspections conducted by the Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC) and the International Atomic Energy Agency (IAEA) in Argentinean and Brazilian nuclear facilities. This medium-resolution gamma-ray spectroscopy (MRGS) option offers similar usability than low-resolution systems based on NaI detectors, i.e., typical detector size and operation at room temperature, and detection efficiency. On the other hand, it has better stability and energy resolution. For spectra evaluation and calculation of enrichment, the latest version of the NaI Gamma Enrichment Measurement (NaIGEM) code has been used. The goal of this paper is to present the measurement performance observed by ABACC from actual field applications and compare it with typical performance values achievable with low resolution systems. The results indicate that some applications may benefit from the new method, whereas some others have not yet experienced any significant improvement.

INTRODUCTION

Measurements of ^{235}U enrichment by non-destructive assay (NDA) methods based on gamma-ray spectrometry are routinely performed by nuclear safeguards inspectorates for verification of the declared value in "infinitely" thick uranium samples during on-site inspections at fuel fabrication, conversion and enrichment plants. Among several advantages, the methods allow for prompt obtention of the results. The most frequently used options are based on high- and low-resolution systems using high-purity germanium (HPGe) semiconductor and sodium iodide (NaI) scintillation detectors, respectively. More recently, Cerium-doped Lanthanum Bromide ($LaBr_3(Ce)$) scintillation detectors have been tested and approved for regular use by the Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC) and the International Atomic Energy Agency (IAEA) as a medium-resolution option of similar usability, i.e., detector size and operation at room temperature, and efficiency, but with better stability and energy resolution than NaI. For

spectra evaluation and calculation of the enrichment results, the NaIGEM code [1, 2] has been used. It employs fitting procedures to determine the intensity of gamma-rays emitted by ^{235}U in the region 120 – 300 keV, as well as interferences caused by high-energy gamma-rays from ^{238}U decay and low-angle Compton scattering in the same energy region. If a significant amount of ^{238}Th is also present in the sample (commonly observed in reprocessed uranium or natural Th), the intensity of the corresponding 239 keV decay peak may be relevant and then the code is capable of computing. At the end, the intensity of the most prominent 186 keV peak from ^{235}U decay is determined and used to derive the enrichment of the measured sample based on previous calibration of the measurement system with well-known reference samples. The code was originally developed for analyzing only NaI spectra, but the latest versions are capable of interpreting spectra collected with $LaBr_3$ detectors as well. Some studies [3] have already been conducted in order to assess the performance of such detector for enrichment determinations in a laboratory environment. In this paper we discuss performance results obtained under conditions found in actual field safeguards inspections where additional limitations and difficulties are commonly faced.

MEASUREMENT SYSTEMS

The standard $LaBr_3$ detector currently used by ABACC is a Brillance 380 model, which has a 38 mm diameter by 38 mm length cylindric crystal produced by Saint Gobain [4]. In order to minimize background interference and establish a well-defined measurement geometry, 10 mm lead lateral shielding and frontal collimation (25 mm dia.) are used. The selected digital multichannel analyzer (MCA) is the model Base-527 by GBS Elektronik GmbH [5], a compact unit designed to compose a single cylindrical piece when it is connected to preamplifier pins on the back of the detector piece. The MCA may be controlled by means of a dedicated cost-free software named WinSPEC, also provided by GBS. For all measurements, the MCA is adjusted for collecting gamma spectra with 512 channels and energy calibrated with slope of 0.62 keV/Ch. Enrichment calibration requires a single measurement. A set of certified reference materials composed by five aluminum cans containing about 200 grams of U_3O_8 in each with enrichment ranging from depleted to 4.46 ^{235}U wt% is used [6]. Depending on the samples to be measured in the field, additional stainless steel or monel alloy attenuators may be placed between the bottom of the reference material and the collimator opening in order to better simulate actual measurement geometry. This is of particular importance when dense and thick-walled containers like large UF_6 cylinders are measured, otherwise the results may be affected by significant biases that arise from improper correction of the gamma-ray attenuation in the container wall in comparison with the reference material. In case of fuel rods, working standards of similar geometry are used for calibration. Typical calibration measurements, in a laboratory, are collected with 30- or 60-min counting times. The NaIGEM code version 2.1.4 [2] is used for data analysis. A digital ultrasonic thickness gauge capable of providing readings with resolution of 0.1 mm or better (ABACC currently uses the model DM4 DL by Krautkramer) is used to determine the wall thickness of the measured item, which is essential for the code to properly correct gamma-ray intensities for wall attenuation. The material type must also be known.

The IAEA uses a slightly different MRGS system, but also based on the NaIGEM code for data analysis. The field experience has demonstrated that both ABACC and IAEA systems provide similar performances.

FIELD RESULTS

Drums containing about 220 kg of natural UO_2 are routinely handled at some conversion and the fuel fabrication plants. During safeguards inspections, the drums are verified by weighing using an electronic balance and enrichment measurements using a portable MRGS system as described in session 2. Measurement live time is set at 300 seconds. The uncertainty associated with the declared value is assumed to be negligible.

Table 1 presents the summary results obtained during five consecutive years, including some statistical evaluation. The analysis of variance (ANOVA) method [7] was used to estimate two uncertainty components: between different years and within a single year. The total uncertainty is given by the square root of the sum of squares of these two components. For comparison purposes, the typical uncertainty value for LRGS systems (NaI) is 5.8% according to international target values obtained from historical evaluation of actual measurement data collected during inspections and published by the IAEA [8]. The observed total uncertainty was 2.9%, indicating that MRGS provided improved performance (50% better) in comparison with LRGS for this application.

Table 1: Summary Results - Declared to Measured (O-I) % Deviation for Enrichment Measurements of Natural UO_2 Drums by MRGS.

Year	O-I Difference (% rel.) (average per year)	O-I Difference (% rel.) (Std deviation per year)
#1	-0.37	2.81
#2	1.64	3.71
#3	0.89	2.18
#4	-0.38	2.85
#5	0.51	1.42
Statistical Parameters	Total Number of Measurements = 117 Overall O-I Difference (% rel.) = 0.35 Overall Standard Deviation = 2.9%	
Uncertainty Estimates (ANOVA)	$u_{between}$ = 0.7% u_{within} = 2.8% u_{total} = 2.9%	

Enrichment of drums containing LEU clean scrap are also measured using MRGS. However, the total number of measurements available is smaller than in the previous case. In addition, the uncertainty associated with the declared value cannot be considered negligible due to important sources of errors such as the use of nominal values and heterogeneity in the physical form of the material in the drum.

Table 2 presents the summary results obtained during three consecutive years, including some statistical evaluation. For comparison purposes, the typical uncertainty value for LRGS systems (NaI) is 3.6 [8]. The observed total uncertainty for enrichment measurements with MRGS was 4.4%. However, it includes contributions from both the operator and the inspector measurements. Supposing these contributions are of similar magnitude, one can estimate the MRGS uncertainty for a single LEU scrap measurement as the combined value (4.4%) divided by the square root of 2, which gives 3.1%.

Table 2: Summary Results - Declared to Measured (O-I) % Deviation for Enrichment Measurements of Low Enriched Scrap in Drums by MRGS.

Year	O-I Difference (% rel.) (average per year)	O-I Difference (% rel.) (Std deviation per year)
#1	-0.31	2.78
#2	-1.31	5.97
#3	0.87	2.14
Statistical Parameters	Total Number of Measurements = 14 Overall O-I Difference (% rel.) = 0.34 Overall Standard Deviation = 4.0%	
Uncertainty Estimates (ANOVA)	$u_{between}$ = 0.6% u_{within} = 4.4% u_{total} = 4.4%	

LEU fuel rods enriched from 0.85 to 4.25 ^{235}U wt% are another type of item commonly measured in fuel fabrication plants. Table 3 presents the summary results obtained during three consecutive years, including some statistical evaluation. For comparison purposes, the typical uncertainty value for LRGS systems (NaI) is 3.2 [8]. The observed total uncertainty for enrichment measurements with MRGS was also 3.2%.

Table 3: Summary Results - Declared to Measured (O-I) % Deviation for Enrichment Measurements of Low Enriched Fuel Rods by MRGS.

Year	O-I Difference (% rel.) (average per year)	O-I Difference (% rel.) (Std deviation per year)
#1	-2.04	2.87
#2	-0.20	3.59
#3	-2.23	1.80
Statistical Parameters	Total Number of Measurements = 14 Overall O-I Difference (% rel.) = -1.31 Overall Standard Deviation = 2.9%	
Uncertainty Estimates (ANOVA)	$u_{between}$ = 1.1% u_{within} = 3.0% u_{total} = 3.2%	

CONCLUSIONS

The performance of non-destructive enrichment measurements based on gamma-ray spectrometry using medium resolution $LaBr_3$ detectors and spectra analysis by a code that uses peak fitting algorithm (NaIGEM) during actual safeguards inspections in conversion and fuel fabrication plants has been evaluated by ABACC. Typical items are drums containing uranium oxide powders, scraps, as well fuel rods. The MRGS method has demonstrated to offer better performance in comparison the obsolete LRGS based on NaI detectors for measurements under good measurement conditions. Measurements of drums containing pure natural UO_2 were able to provide results with 50% better uncertainties than LRGS. On the other hand, for items subject to additional uncertainties in the declared values, i.e., scrap, the observed performances were similar. Future investigations aiming at quantifying item-specific sources of uncertainties for separation from the contributions associated exclusively with the measurement method are planned. This is a common practice is the area of

destructive analysis and should be more explored also in NDA applications.

ABACC has successful completed the migration from the low to the medium resolution gamma spectrometry method for enrichment measurements using $LaBr_3$ detectors and the NaIGEM code. The establishment of typical measurement uncertainties for the MRGS method in different applications is currently being discussed by experts and users world-wide, including ABACC, under the coordination of the IAEA.

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