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Electrically Cooled Germanium System for Measurements of Uranium Enrichment in UF6 Cylinders

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Abstract

Measurements of Uranium enrichment in UF6 cylinders is a significant part of the IAEA Safeguards verification activities at enrichment and conversion plants. Nowadays, one of the main tools for verification of Uranium enrichment in UF6 cylinders used by Safeguards inspectors is the gamma spectroscopy system with HPGe detector cooled with liquid nitrogen.
Electrically Cooled Germanium System (ECGS) is a new compact and portable high resolution gamma spectrometric system free from liquid nitrogen cooling, which can be used for the same safeguards applications. It incorporates the ORTEC Micro-trans-SPEC HPGe Portable Spectrometer [1], a special tungsten collimator and UF6 enrichment measurement software.
The enrichment of uranium is determined by quantifying the counting rate in the 185.7 keV peak in the conditions of infinite thickness of the measured material. Prior starting the verification of uranium enrichment at the facility, the ECGS has to be calibrated with a sample of known uranium enrichment, material matrix, container wall thickness and container material.
Evaluation of the ECGS capabilities was performed by carrying out a field test on actual enrichment verification of uranium in UF6 cylinder or other forms of uranium under infinite thickness conditions. The results of these evaluations allow to conclude that the use of ECGS will enhance practicality of the enrichment measurements and support unannounced inspection activities at enrichment and conversion plants.

1. Introduction

The primary non-destructive assay (NDA) approach in Safeguards for U-235 mass measurements in UF6 cylinders is to use high-resolution gamma spectroscopy (HRGS) or low-resolution gamma spectroscopy (LRGS) for uranium enrichment verification combined with weight measurements. The gamma spectroscopic systems presently used in safeguards inspections are the following:
- MMCG and IMCG – HRGS systems consisted of Mini Multichannel Analyzer MCA-166 (GBS Elektroniks) [2] or InSpector 2000 MultiChannel Analyzer (Canberra) [3] coupled with collimated High Purity Germanium detector (HPGe). The U-235 enrichment is determined from gamma spectra taken by these systems and normally analysed by the IMCA/MMCA software, which employs standard three-regions method [4], and requires calibration of the system with standards of known uranium enrichment, material matrix composition, container wall thickness and container material.

- MMCN and IMCN – LRGS systems consisted of Mini Multichannel Analyzer MCA-166 (GBS Elektroniks) or InSpector 2000 MultiChannel Analyzer (Canberra) coupled to collimated NaI detector (2" in diameter and 1/2" thick). The U-235 enrichment is determined from gamma spectra taken by these systems and normally analysed by IMCA/MMCA software, which requires calibration of the system with at least two standards of known uranium enrichment, material matrix composition, container wall thickness and container material. Alternatively U-235 enrichment can be analysed by NaIGEM code software [5].
- Hand-held Monitor System V. 5 (HM-5) [6] – hand-held LRGS systems with collimated NaI detector (1" in diameter and 1" thick) inside. The NaIGEM code has been included in the HM-5 firmware since 2005.

There are some difficulties/problems with these instruments used for U-235 enrichment measurements in UF6 cylinders, especially for the cylinder types 30B for $LEUF_6$ and 48Y for NUF_6 and DUF_6, which have wall thicknesses of 12-14 mm and 14-18 mm stainless steel correspondingly.

Because of high attenuation of U-235 peaks in container wall the MMCN and IMCN used with two-region method do not show satisfactory performance for NUF_6 and DUF_6 in 48Y cylinders, especially in high background conditions.

The use of HM-5 with NaIGEM code requires very long measurement time due to a small size of NaI detector.

The best measurement results are shown by the IMCG. However, there are another factors which introduce some limitations on the system usage in the field: first of all, HPGe detectors require cooling with liquid nitrogen, which is not everywhere easily available; secondly, the IMCG systems are bulky and heavy instruments and therefore they require extra fixtures/appliances to carry and/or hold them in proper measurement position; finally HPGe detectors also require significant time for detector cooling and warm-up before air shipment.

The ECGS system which was currently introduced into the Agency's Safeguards allowed to avoid most of listed problems and limitations.

2. Electrically Cooled Germanium System

First electrically cooled HPGe detectors appeared on the market about 20 years ago, but were rather big and heavy and could not be used as portable NDA instrument that time. However, the technology has being improved and several years ago a new generation of portable electrically cooled HPGe detectors were issued to the market by the main manufacturers of HPGe detectors.

The SGTS evaluated available models of electrically cooled HPGe detectors and found them applicable for Safeguards purposes for attended and unattended NDA measurements. A new compact and portable high resolution gamma spectrometric system called Electrically Cooled Germanium System (ECGS) was created on the basis of one electrically cooled HPGe detectors available on the market. It incorporates the ORTEC Micro-trans-SPEC HPGe Portable Spectrometer, a special tungsten collimator and UF6 enrichment measurement software. The tungsten collimator and UF6 enrichment measurement software were developed in accordance with SGTS technical requirements by ORTEC. The ECGS and the interface of UF6 enrichment measurement software are shown at the Figures 1 and 2 correspondingly.

Fig. 1. Electrically Cooled Germanium System

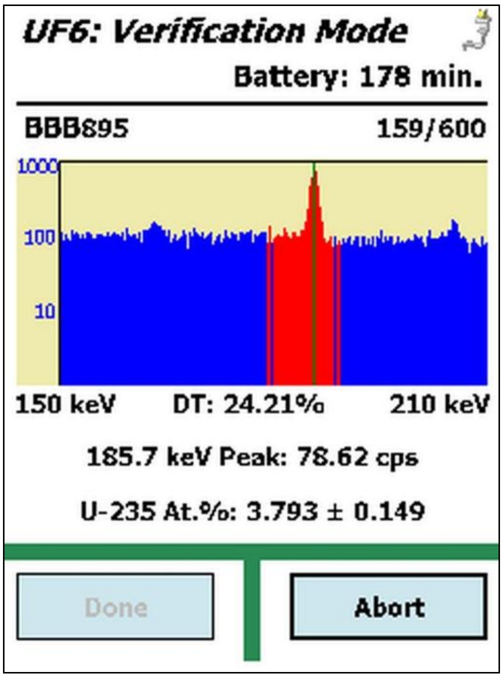

Fig. 2. The interface of UF6 enrichment measurement software

3. U-235 enrichment measurements with ECGS

The U-235 enrichment measurement with the ECGS were performed on several facilities. The conditions and the results of these measurements are described below.

3.1. The U-235 enrichment measurements UF_6 cylinders with feed (NUF_6) and tails ($DEUF_6$) material at an enrichment facility.

The ECGS with lead collimator (10 mm thick, 1 mm tin liner and 50 mm opening) has been calibrated with reference standards containing U_3O_8 of known enrichment at Safeguards Instrumentation Laboratory in Seibersdorf.
The measurements were performed in a storage area with 30B cylinders containing $LEUF_6$ and 48Y cylinders with NUF_6 and DUF_6, which were stored in large closely packed arrays (around 1-1.5 m from each other), stacked two high.

Fig. 3. ECGS hung in measurement position on 30B cylinder

The U-235 enrichment measurements were performed for NUF_6 and $DEUF_6$ material in 30B cylinders. The results of the measurements are shown in Table 1.

Cylinders			Measurements (300 s)				
Cylinder	Wall Thickness (mm)	Declared enrichment (%)	Gross Area (c)	Net Area (c)	Relative Net Error	Measured Enrichment (%)	**Relative Difference** (M-D)/D
001	16.8	0.714	19105	4812	6%	0.76	6%
002	16.3	0.209	19878	1338	24%	0.20	-5%
003	16.8	0.714	18409	4602	6%	0.72	1%
004	18.3	0.215	18056	903	35%	0.17	-21%
005	18	0.285	21849	1462	23%	0.27	-7%
006	10	0.296	29224	4397	9%	0.30	3%
007	9.5	0.291	25997	4457	8%	0.29	0%
008	9.8	0.291	26202	3945	9%	0.27	-9%

Table 1. Results obtained with the ECGS on tails and feed cylinders.

The measurement results obtained with the instrument for NUF_6 and $DEUF_6$ showed good agreement with the declared values (within 10%) for reasonable counting time of 300s. The only exception was for $DEUF_6$ in a 18.3 mm thick cylinder.

3.2 The U-235 enrichment measurements in UF_6 cylinders at the enrichment facility.

The ECGS with tungsten collimator (8 mm thick, 50 mm opening) has been calibrated at the facility against 30B cylinders containing $LEUF_6$ of known enrichment.
The measurements were performed in storage areas with 48Y cylinders containing DUF_6, which were stored in large closely packed arrays (around 70 cm from each other), stacked two high. There were also dozens of 30B and 48Y cylinders, containing NUF_6 and $LEUF_6$, these are positioned in separated arrays.
The U-235 enrichment measurements were performed for $LEUF_6$ in 30B cylinders, NUF_6 and $DEUF_6$ in 48Y cylinders. The results of the measurements are shown in Table 2.

	Cylinders			Measurements		
Run	Cylinder (B: 30B, Y: 48Y)	Wall thickness (mm)	Declared Enrichment (w/o)	Measured Enrichment (w/o)	Relative Difference (%)	Notes
1.	B001	12.6		-	-	Calibration – 600s
2.	B002	13.0	4.401	4.58 ± 0.10	4.1	Enriched – 300s
3.	B002	13.0	4.401	4.51 ± 0.09	2.5	Repeat measurement in same position
4.	B002	12.6	4.401	4.30 ± 0.096	-2.3	Re-measurement after wall thickness re-measured and revised.
5.	B003	12.1	2.38	2.36 ± 0.06	-8.4	Enriched – 300s
6.	Y001	16.8	0.252	0.346 ± 0.115	37.3	Depleted – 300s
7.	Y002	17.0	0.711	0.694 ± 0.68	-2.4	Natural – 300s
8.	Y004	16.9	0.223	0.144 ± 0.111	-35.4	Cylinder half full, detector at bottom of tank – 300s.
9.	Y005	16.2	0.250	0.202 ± 0.106	-19.2	Repeatability test # 1 - 300s.
10.	Y005	16.2	0.250	0.348 ± 0.105	39.2	Repeatability test # 2 - 300s.
11.	Y005	16.2	0.250	0.284 ± 0.105	13.6	Repeatability test # 3 - 300s.
12.	Y005	16.2	0.250	0.159 ± 0.106	-36.4	Repeatability test # 4 - 300s.
13.	Y005	16.2	0.250	0.381 ± 0.105	52.4	Repeatability test # 5 - 300s.
14.	Y005	16.2	0.250	0.293 ± 0.075	17.2	600s run
15.	Y006	16.4	0.191	0.168 ± 0.093	-12.0	Cylinder positioned on the edge of the array of 48Y cylinders - 300s.

Table 2. Results obtained with the ECGS on 30B and 48Y cylinders.

As it can be seen from the measurement results the ECGS gave excellent results for $LEUF_6$ and NUF_6 within 5% with a 300-600 s counting time.
However, the instrument showed poorer performance in the case of the measurements of $DEUF_6$ in 48Y cylinders giving unreasonably high random uncertainty.
This could be explained with measurement conditions for the close-packed 48Y cylinders with only a distance of only 70 cm between the cylinder faces. The measurement #15 of a cylinder on the periphery of the array showed relatively good result within 12%.

3.3 U-235 enrichment measurements of recycled UO_3

Problem of U-235 enrichment measurements of recycled uranium are well-known. Irradiation of uranium in reactor leads to accumulation of U-232 isotope in this uranium with content of a fraction of ppm. Despite of low quantity, activity of U-232 in recycled uranium is very high due to its short decay time (69 y) in comparison to other U isotopes.
The daughter product of U-232 is Th-228 (1.9 y) which decays to stable Pb-208 through the chain of short lived isotopes including Pb-212 and Tl-208. Typical spectrum of recycled uranium containing 238 keV peak from Pb-212. Due to its energy close to U-235 main group of peaks (143, 163, 185 and 205 keV), it influence on low-resolution gamma spectra analysis. In addition highly penetrating 2614 keV peak from Tl-208 increases Compton background in the spectrum, what leads to poor peak-to-background ratio, higher dead-time, bringing systematic bias which always takes place even after dead-time correction. There is also need for longer counting time to achieve certain statistical accuracy in the 185 keV peak area.

The measurements of bottles with recycled uranium were performed with different collimator setups.
The main purpose was to investigate signal (at 185 keV peak) to Compton background ratio and dead time under two collimator opening configurations:

- wide collimator opening for better peak-to-Compton background ratio, however, in this case the dead-time is higher due to less detector shielding;
- narrow collimator opening with better detector shielding and lower dead-time, however, it results to poor peak-to-Compton background ratio.

The ECGS was calibrated on cylinder with known U-235 enrichment at the facility. Two setups of a tungsten collimator opening: narrow (15 mm diameter) and wide (50 mm diameter). The measurements were carried with uranium material recycled around 2 years ago and the results of the measurements are shown in Table 3.

Measurement	Collimator Diameter (mm)	Wall Thickness (mm)	Dead Time
Measurement 1	15	4.7	46%
Measurement 2	50	4.7	63%
Measurement 3	15	4.7	67%
Measurement 4	50	4.7	81%
Measurement 5	50	4.7	54%
Measurement 6	50	4.7	48%
Measurement 7	Background		40%

Table 3: Dead-time parameter for different measurements of recycled uranium

With the small opening collimator the dead time is lower. However, most of the count rate comes through the shielding (and Compton scattering from high energy gamma within the detector). The wide opening collimator has a much better signal to noise ratio, but then the dead time exceed 80%. It significantly increases Real measurement time when the Live time is fixed.

In addition, laboratory test measurements with well characterized depleted and highly-enriched uranium standards showed that there is a systematic bias in the measured uranium enrichment which is rising when the dead-time parameter is growing, and approximately amounts to 8-10% when the acquisition dead-time reaches 40-45%.

The hardware settings used for the recycled uranium measurements did not allow to achieve reasonable dead-time parameter and satisfactory results. However, default settings optimized to have the best possible resolution such as low-frequency noise filter and pulse shaping time parameters can be adjusted to depress dead-time and make possible precise enrichment measurement of aged recycled uranium. Further research work is needed towards this issue.

4. Conclusion

We consider Electrically Cooled Germanium System as a perspective instrument for verification of U-235 enrichment in UF6 cylinders at enrichment and conversion plants. The ECGS pilot infield testing at various nuclear facilities showed its good performance in comparison with other instruments authorised for the Agency’s inspection activities. The instrument has a user-friendly software interface, which is intuitively ease-to-use to operate by the safeguards inspectors. The various improved corrections methods implemented in the incorporated analysis software make the verification results more accurate. However, despite of obvious benefits, ECGS has some weaknesses, such as a long cooling down time and relatively high cost, which may apply limitations on its use.
In addition, the ECGS is considered as useful tool for short-notice or unannounced inspection when a high-resolution gamma spectroscopy system is needed for verification purposes and liquid nitrogen can not be provided.

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