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Uranium enrichment measurement in UF6 cylinders by HRGS : an evaluation of the measurement uncertainty

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ABSTRACT

- Optimization of the traditional enrichment meter method applied to uranium enrichment measurement in 30B and 48Y UF6 cylinders.
- Calibration between 235U enrichment and net count rate in the 185.7 keV in collimated geometry with small U3O8 sources and electrically cooled high resolution gamma-ray spectrometer.
- Net count rate measured on site in non-collimated geometry and corrected from container wall attenuation, measurement geometry and physico-chemical differences before applying the inverse calibration.
- On-site measurement campaign used to calculate a calibration transfer factor, estimate measurement time , evaluate uncertainty components of each parameter of influence and total measured enrichment uncertainty.

BACKGROUND

- On-site non destructive assay of nuclear material as technical support to the Authority responsible for the protection and control of nuclear material in France.
- HRGS routinely performed by operators and inspectorates to verify 235U enrichment in large UF6 cylinders in conjunction with an ultrasonic measurement of the container wall thickness => reliable method for inspection purposes.
- Methodology and instrumentation need to be optimized to achieve reasonable inspection time based on available standards at IRSN.
- Parameters of influence and associated uncertainties need to be mastered

CHALLENGES / METHODS / IMPLEMENTATION

MEASUREMENT PRINCIPLE

$E = K * TC * CFmat * CFmT$

With K = calibration constant
TC = net count rate at 185.7 keV
CFmat = matrix material composition correction factor
CFmT = container wall correction coefficient

Practical limitation ⇔ size of the calibration reference material vs infinite thickness requirements and better compromise between parallel beam collimation and reasonable measurement time.

MEASUREMENT SYSTEM AND CALIBRATION REFERENCE MATERIALS

Typical calibration using a collimated and laterally shielded portable high resolution gamma spectrometer connected to a computer equipped with Gammavision and in-house Enrichissement software, and four U_3O_8 reference material (104-120 g. U_3O_8, 0.7-89% ^{235}U, 48 mm diameter, ≥ 26 mm height, embedded in an acetal copolymer container).

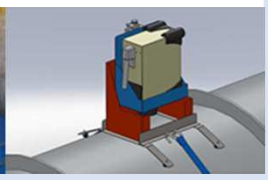
ON SITE IMPLEMENTATION

Measurements performed at the top of 4x30B and 4x48Y containers (2/12 tons of UF6, 0.3 to 5% ^{235}U, 12.9 to 16.8 mm container wall thickness) with no collimation. Important deadtime => use of shorter rise time.

UF6 235U enrichment given by DA with 0.01% uncertainty.

Container wall thickness measured with a Positector ultrasonic gauge with 0.118 mm uncertainty (calibration, resolution, paint thickness, T° effects).





30B container (D 76cm) *48Y container (D 122cm)* *Measurement config.*

OUTCOME

CALIBRATION UNCERTAINTY BUDJET (given at 1σ)

- Counting rate = TC (1%)
- TC correction due to the reference material container wall (0.03%)
- Calibration factors ⇔ weighted linear least squares regression

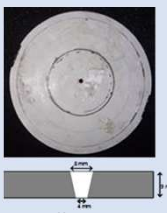
UF6 CYLINDERS VERIFICATIONS UNCERTAINTY BUDJET (given at 1σ)

- TC (1%)
- TC correction due to UF6 container wall (μρ calculated using X-MUDAT assuming ASTM A516 Grade 65 => 0.47% ; thickness unc. 0.118 mm)
- TC correction due to the non-collimated geometry:
  - Experimental (for 1 cylinder from TC with/without collimator) ⇔ 5.5%
  - MCNP (from modeled TC with/without collimator) ⇔ 9%
  - Optimization by bias minimization using 2x30B and 2x48Y containers and an average bias of 0.001% ⇔ 1% (0.01% on declared %^{235}U)
- Inverse calibration
- TC correction due to the difference in chemical composition between reference material and item to be measured ⇔ 0.24%.
- Precision (1.35% for 20 measurements with 1 container), trueness (0.37%), detector position influence (1.1% for 7 locations of 1 isolated container)
- Overall uncertainty : 5-10% (cf. table, formulas developed in the paper)
- $-0.37<\Delta/3\sigma<0.23$ for the set of 34 measurements done on 8 containers

	Type	Counting Time (s)	Ref. ^{235}U% (wt %)	Measured ^{235}U% (wt %)	Unc. (wt %)	Relative Unc. (%)	Bias (wt%)	Rel. Bias (%)
1	30B	523	1.4133	1.4486	0.082	5.68	0.0366	2.6
2	30B	90	4.1885	4.3340	0.230	5.28	0.1554	3.7
3	30B	90	4.9202	5.0237	0.264	5.24	0.1055	2.1
4	30B	90	4.1923	4.2564	0.229	5.36	0.0738	1.8
5	48Y	7203	0.3661	0.3506	0.036	10.35	-0.0176	-4.8
6	48Y	7050	0.4697	0.4663	0.039	8.48	-0.0051	-1.1
7	48Y	3060	0.7110	0.7170	0.049	6.83	0.0050	0.7
8	48Y	120	4.9240	4.9159	0.270	5.48	0.0035	-0.1
						Mean	0.0446	0.64
					Trueness Unc.		0.0258	0.37

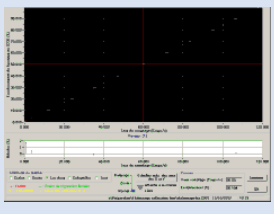
Measurement results

U3O8 reference material



Collimator 4x8x9 mm

Calibration configuration



Calibration ^{235}U Enr (%) vs TC (c/s)

CONCLUSION

- On-site verifications compatible with time inspection constraints
- Evaluation of an overall measurement uncertainty propagating uncertainty components of each parameter of influence (5-10% LEU-DU)
- Possible improvements :
  - Design of a new collimator to reduce UF6 measurements deadtime
  - Additional peak fitting algorithms to be investigated to extract net peak area from a high continuum background with better accuracy
  - Evaluation of the geometry transfer function from a larger U reference sample.