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Assessment of Operator's Declaration from the Nuclear Safeguards Prescriptive

H. I. Khedr[1], M. Abdelati[1], K.M. El Kourghly[1]
[1](*Nuclear Safeguards and Physical Protection department/ Egyptian nuclear and Radiological Regulatory Authority (ENRRA) - Cairo, Egypt)*
Corresponding Author: H. I. Khedr

Abstract: *Recently, Nuclear Fuel Fabrication Plant (NFFP) decided to produce uranium mini-plates (targets) with chemical composition U_3O_8 to be used at Radioisotope Production Facility (RPF). Assessment of operator's declaration to verify Nuclear Material [NM] presence in the fabricated targets is of high importance for both national and international nuclear safeguards (SG). HPGe detector was used to determine isotopic ratios as well as uranium mass based on Multi-Group Analysis software (MGAU) and efficiency calibration method, respectively. Measurements were performed at different axial sample to detector distances (5, 10 and 20 cm) from the front facet of the detector for measuring time 120 s. The obtained results were compared with the declared values, the relative difference in the estimated ^{235}U mass based on MGAU was ranging from -6.20 % to 7.15 % and from 0.3 % to 5.49 % based on efficiency calibration method.*

Keywords: *Uranium mini-plate, efficiency calibration, uranium mass.*

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I. Introduction

Nuclear Fuel Fabrication Plant (NFFP) decide to investigate the feasibility of fabrication low enrichment uranium mini-plates (hereinafter referred to as "targets") with chemical composition U_3O_8 and dimensions suitable for using at molybdenum production facility.

The physical inventory verification of nuclear materials includes the conformity assessment activities to check whether nuclear operator's declarations and accounting report and operating records, including Items List of the Inventory, correctly reflect the physical reality [1].

On-site nuclear material verification activities, for nuclear material accounting purpose, is the primary basis to detect any undeclared processing or production of nuclear material at facilities or location out facilities (LOFs) [2]. In nuclear fuel fabrication plants, nuclear material is handled in item and bulk forms. Various safeguards measures, both non-destructive and destructive assay, can be applied to both forms for nuclear material verification. Standard sources are needs, when using a non-destructive assay (NDA) system, to allow efficiency calibration and these sources should match the geometry and physical property of the sample. However, these sources are not always obtainable. Efficiency calibration using the ISOCS mathematical efficiency calibration software eliminates the need of radioactive sources for efficiency calibration by the use of a Monte-Carlo based mathematical efficiency computing method [3, 4]. ISOCS could be used for quantitative analysis in a wide range of applications (scrap materials, solid wastes, fuel elements … etc.).

Multi-Group Analysis for Uranium (MGAU) application analyses the HPGe detector spectra of uranium samples, and reports the relative abundances of the uranium isotopes in the sample [5]. Before make the analysis for the gamma-rays spectrum the energy calibration and the efficiency calibration are required, and sequentially call several analyses to perform the analysis. MGAU only requires that the spectra be energy calibrated, and it performs its entire analysis in one step. For peaks used to build the relative efficiency curve MGAU generates and stores various parameters such as: peak energy, peak counts, relative efficiency and relative efficiency uncertainty. MGAU Output report contains the values of these parameters.

The main purpose of this paper is to assess and verify operators' declarations of uranium mass in natural and low enrichment uranium targets at NFFP.

II. Materials and method

2.1 Material specification

A homogeneous mixture of aluminum and uranium powder (U_3O_8) is pressed to form uranium compact, considered as the core of the target (hereinafter referred to as "meat"). The final dimensions of the target (thickness, width and length) are result from the cladding of the compact with aluminum and performing the rolling operation. In order to check the position of the meat in the target and verify its dimension, length and

width, X-ray radiography technique is performed. Targets are designed to be suitable for irradiation into the core of the MTR research reactor. Five targets were selected randomly in order to verify the declared data. An example of the dimensions and shape of the assayed targets is shown in Fig. 1. Table (1) provides the specifications of the selected targets according to the operator declaration.

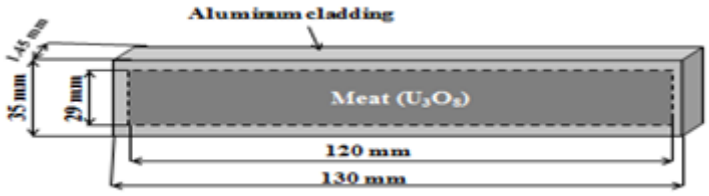


Fig. 1: shape and example for dimensions of the assayed targets

Table 1: Targets Specification provided by operator

Target Id			T1	T2	T3	T4	T5
Enrichment			NU	LEU	LEU	LEU	LEU
Dimensions (mm)	Plate	Thickness	1.525	1.448	1.447	1.464	1.458
		Length	130.5	130.05	130.1	130	130
		width	35.15	35.05	35.07	35.15	35
	Meat*	Length	116.5	121	120	114.75	116.75
		width	28.25	29.25	29	29.25	29.50
		Average Thickness	0.7	0.7	0.7	0.7	0.7
Mass (g)	U_3O_8		7.87	7.89	7.88	7.72	7.79
	U_{Total}		6.67	6.68	6.68	6.54	6.59
	U-235		0.0474	1.32	1.32	1.29	1.30

* Meat dimensions estimated by using radiography technique

2.2 Measuring device

Gamma-ray spectrometer based on electrically cooled Broad-Energy Germanium Detector (BEGe) with an active volume 89.00 cc, 3.09 cm length, 6.08 cm diameter, ≤1 keV FWHM at 0.122 MeV, ≤ 2 keV at 1.33 MeV, and relative efficiency ≥ 20%, a built in Multi-channel Pulse-Height Analyzer [Inspector, Model IN2K], for sorting and collecting the gamma-ray pulses coming from the main amplifier, an adjustable High Voltage Power Supply [HVPS], provides a negative voltage of 3300V which is necessary for detector operation, The measuring system is combined with In Situ Operating Counting System (ISOCS) Software used for efficiency calibration and with Canberra multi-group analysis software MGAU (version S507c) [6].

2.3 Measurements

Targets were assayed by placing samples axially in the front facet of the HPGe detector. Targets are measured at different source-detector distances (5, 10 and 20 cm), three times each, and measuring time (120 s). Distances were adjusted and optimized to obtain the maximum count rate mean while the counting losses due to pile up and dead time were minimized. Count rates of 185.7 keV and 1001.2 keV gamma lines relevant to ^{235}U and ^{238}U uranium isotopes, respectively, have been registered. Fig. 2 shows a schematic diagram for the experimental setup configuration arranged to measurements of the targets.

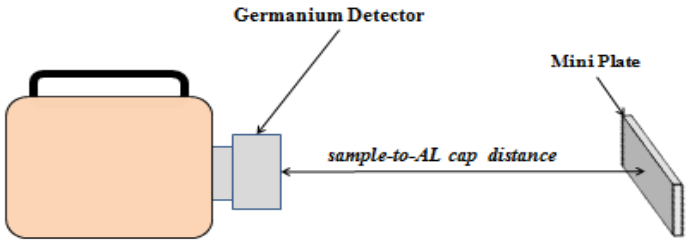


Fig. 2: A schematic diagram for the configuration of targets during measurement.

The ratio of ^{235}U enrichment is an important value in characterizing the nuclear material from the nuclear safeguards point of view. It can be given by dividing the ^{235}U weight by the total uranium weight as the following [7]:

$$\varepsilon_w = \frac{weight\ of\ ^{235}U}{weight\ of\ U_t} \quad \text{---------- (1)}$$

Where U_t is the total weight of uranium,

From this equation it is clear that ^{235}U weight can be estimated whenever the enrichment and the total uranium mass in a given sample are known.

GENIE 2000 software has been used to find and estimate the area under each peak (185.7 & 1001.2 KeV) in the spectrum and, after that, uranium enrichment value (^{235}U ratio) for each target has been estimated using the Canberra multi-group analysis software (MGAU) combined with the measuring system.

The efficiency calibration has been performed by using the ISOCS software to estimate the activity of ^{235}U and ^{238}U isotope. The first step of the efficiency calibration using ISOCS is the generation a geometry model for the experimental set-up. The geometry model for each target is based on the dimensions data, provided by the operator, and the distance between the target-frontal face of the detector. The built in rectangular plane template has been used to generate an efficiency calibration file and perform the ISOCS efficiency calibration (absolute calibration). The ISOCS efficiency calibration file for each target was applied to its spectrum. The gamma peaks were identified in addition to the isotopes weighted mean activities. Normally, the modeling and numerical calibration consumes several minutes. Fig. 3 shows a schematic diagram for the model set-up generated for each target.

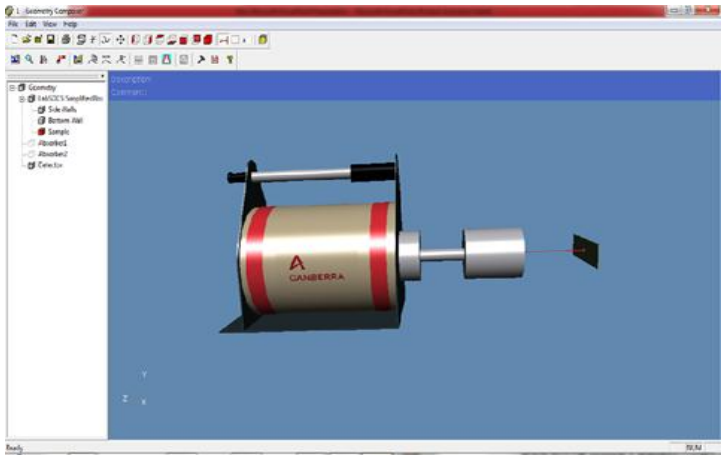


Fig. 3: a schematic diagram for the model set-up generated for each target

III. Results and Discussion

Uranium mass content in each target has been estimated using two different methods as the following:

a. Based on ^{235}U enrichment estimation

Uranium total and isotopic mass contents in each target have been obtained from equation (1) by multiplying the results obtained for uranium isotopic ratio using the MGAU software by the total uranium mass provided by operator.

b. Based on efficiency calibration

After the calculation of the absolute efficiency curve, it has been used for the quantitative analysis of the isotopes of interest by using peak intensities derived from the acquired spectrum. The isotope weighted mean activities obtained (at 185.7 or 1001.2 keV gamma energies) are divided by the activity per unit mass (specific activity) of gamma energy line in order to obtain the isotope mass contents in each target.

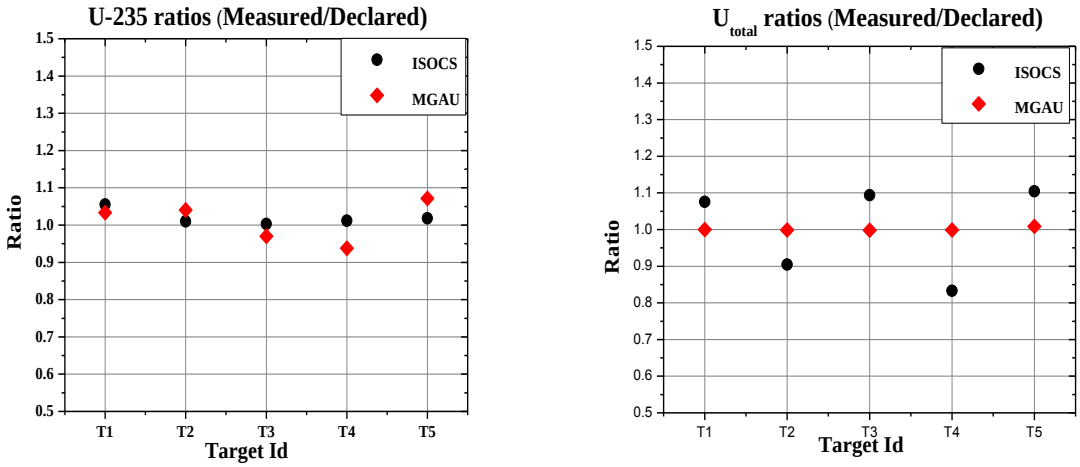
Uranium masses obtained from ISOCS and those obtained from the MGAU were compared to the declared value. Table (2) provides the uranium mass estimated with the associated percentage relative uncertainties.

Table (2): Uranium mass estimated by the two methods in comparison with declared value

No.	Uranium	Target Id	Declared Uranium mass (g)	ISOCS™		MGAU	
				Mass ± σ_M	diff %	Mass ± σ_M	diff %
1	^{235}U	T1	0.0474	0.050 ± 0.005	5.49	0.049 ± 0.002	3.38
2		T2	1.32	1.333 ± 0.117	0.98	1.374 ± 0.068	4.09
3		T3	1.32	1.324 ± 0.116	0.30	1.280 ± 0.063	-3.03

4		T4	1.29	1.305 ± 0.114	1.16	1.210 ±0.044	-6.20
5		T5	1.30	1.324 ± 0.116	1.85	1.393 ±0.070	7.15
6	U_{total}	T1	6.67	7.174 ± 0.336	7.56	6.669 ± 0.003	-0.016
7		T2	6.68	6.042 ± 0.997	-9.55	6.671 ± 0.097	-0.130
8		T3	6.68	7.308 ± 1.102	9.40	6.668 ± 0.089	-0.178
9		T4	6.54	5.447 ± 0.792	-16.71	6.533 ± 0.063	-0.113
10		T5	6.59	7.279 ± 1.207	10.47	6.647 ±0.100	0.872

For data results presented in Table (2), the relative differences between the declared uranium mass and that obtained by using the MGAU method ranged from -6.20 to 7.15 for ^{235}U and from -0.178 to 0.872 for uranium total mass. The essential source of this difference is the measurement time, it was short (120 s) to simulate the inspection case inside the facility. Also, it is clear the obtained mass is in agreement within the uncertainties with the declared value. For mass obtained from ISOCS method the difference ranged from 0.3 to 5.49 and from -16.71 to 10.47 for ^{235}U and uranium total, respectively. The short time of the measurement and differences between the actual and the modeled geometry are essential sources of the difference between the obtained and declared values. Differences between the actual and modeled geometry may include distribution of the uranium within the target, location of the target to the detector and the physical properties of the target (clad thickness and shape, material composition.....etc.).
The ratios of the obtained uranium mass value to the uranium declared mass value are displayed in Fig. 4.



(a) ^{235}U Ratios (b) Utotal Ratios

Fig. 4: Ratios of the measured value to the declared value of uranium mass

Fig.4 shows that, for uranium masses obtained from ISOCS, the ratios of ^{235}U mass (measured/declared) range from 1.00 to 1.05 and Utotal mass ratios range from 0.83 to 1.10. While, for uranium masses obtained from MGAU, the ratios of ^{235}U mass range from 0.93 to 1.07 and Utotal mass ratios range from 0.99 to 1.00.

IV. Conclusion

Nuclear material verification activity was performed at the facility, to verify the operators' declarations values, by using gamma-ray spectrometry. Uranium mass contents in each target has been estimated by two methods, one of them based on isotopic fraction estimation by MGAU and the second, based on modeling the experimental setup for efficiency calibration using ISOCSTM. The obtained value of uranium mass has been compared with the value of uranium mass declared by the operator for each target. Because of the short time of measurement the difference between the obtained value of uranium mass and the declared value was relatively large in some cases. Inaccurate modeling for actual experimental geometry can be contributed to increase this difference also.

Acknowledgements

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