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Nuclear Material Verification Based on MCNP and ISOCS™ Techniques for Safeguard Purposes

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Abstract: *Recently, Mathematical techniques such as Monte Carlo and ISOCS™ software are being increasingly employed in the absolute efficiency calibration of gamma ray detector. Monte Carlo simulations and Canberra ISOCS™ software bring the possibility to establish absolute efficiency curve for desired energy range based on numerical simulation, with use of known or guessed geometry and chemical composition, of measured item. Broad-energy germanium (BEGe) detector was employed to perform the NDA measurements to five standard reference nuclear material (NBS, SNM-969). MC calculations were performed to calculate some factors (attenuation, geometry and efficiency) which affect the uranium isotope mass estimation. ^{235}U and ^{238}U masses are calculated based on MCNPX modeling calibration and also upon spectra analysis using ISOCS™ Calibration Software. The obtained results from the two different efficiency calibration methods were compared with each other and with the declared value for each sample. The obtained results are in agreements with the declared values within the estimated relative accuracy (ranges between -2.81 to 1.83%). The obtained results indicate that the techniques could be applied for the purposes of NM verification and characterization where closely matching NM standards are not available.*

Keywords: *Absolute efficiency, efficiency calibration, Monte Carlo simulations, Broad-Energy Germanium Detector, gamma spectrometry, ISOCS™ software, uranium isotope mass*

I. Introduction

The efficiency calibration of a system is dependent not only on the detector, but on the radiation attenuation factors in the detector–source configuration, and therefore is invalid unless all parameters of the sample assay condition are identical to the calibration condition. An alternative to source-based calibrations is to mathematically model the efficiency response of a given detector–sample configuration [1].

Several techniques can be used to determine the total efficiency such as Monte Carlo simulations, semi-empirical methods and experimental measurements. MC technique requires a good definition of the geometry and materials, including window thickness together with an accurate set of cross-sections [2].

MC method obtains answers by simulating individual particles and recording some tallies of their average behavior. The average behavior of particles in the physical system is then inferred from the average behavior of the simulated particles. MCNP code treats an arbitrary three-dimensional configuration of materials in geometric cells bounded by surfaces [3].

ISOCS™ (In Situ Object Counting System) Calibration Software brings a new level of capabilities to gamma sample assay by eliminating the need for traditional calibration sources during the efficiency calibration process. By combining the detector characterization produced by the MCNP modeling code, mathematical geometry templates, and a few physical sample parameters, ISOCS™ Calibration Software gives you the ability to produce accurate qualitative and quantitative gamma assays of most any sample type and size [4].

In this paper MCNPX code is employed to estimate the absolute full energy peak efficiency for the measuring system in order to estimate the ^{238}U and ^{235}U mass contents.

ISOCS™ calibration software was used to generate the efficiency file and to check the geometry validity. The generate efficiency file used for estimating ^{238}U and ^{235}U mass contents.

II. Materials and Methods

Standard Reference Material (NBS, SNM-969) consists of a set of five different U_3O_8 powder, with nominal ^{235}U abundances of 0.31, 0.71, 1.94, 2.95, and 4.46 mass percent, encased in aluminum cans (Aluminum type 6061 (ASTM-GS T6)) was used for non-destructive assay. Each SRM 969 subunit is made up of 200.1 ± 0.2 g of U_3O_8 powder [5]. These materials are subject to the international nuclear safeguards. Figure 1 shows the shape and example for dimensions of the assayed SRM-969 samples.

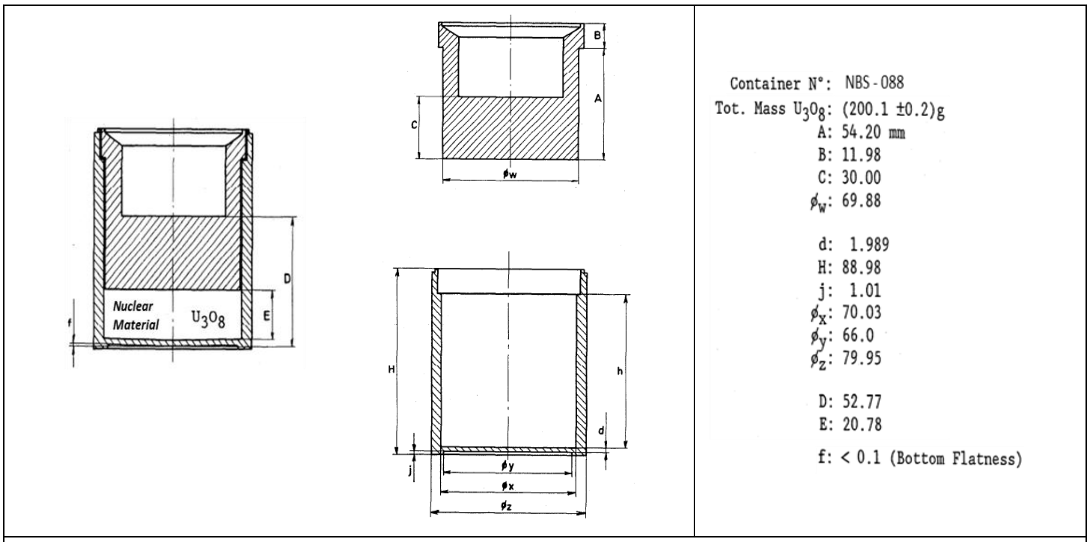

Figure 1: Dimensional control can and the shape of the measured SRM-969 samples [5]

A commercial high-Purity (HPGe) germanium gamma-ray spectrometry produced by Canberra, with a Broad-Energy germanium crystal (Model BE2830), was employed to measure the Count rates due to uranium isotopes. The data acquisition system in this work involves Genie-2000 software.

Samples have a cylindrical shape of approximately 4 cm radius and 9 cm height. The samples were placed in front of the detector as the circular base face the detector so that extended axis of symmetry of the cylinder and the HPGe crystal detector is the same. For all measurements, the samples-to-Aluminum cap of the detector distances (d = 30 cm) were adjusted and optimized in such a way to obtain the maximum count rate mean while the counting losses due to pile up and dead time were minimized. Figure 2 shows a schematic diagram for the experimental setup configuration arranged to samples measurements.

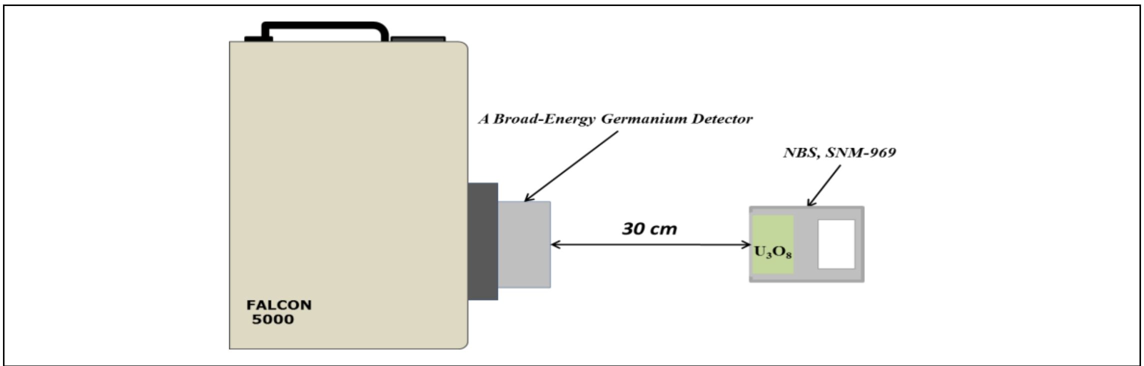

Figure 2: A schematic diagram for the configuration of SRM-969 samples during measurement.

MCNPX and ISOCS™ were used to estimate the absolute full energy peak efficiency (ε_{ab}) of the detector at both 185.7 and 1001.1 keV gamma energies. MCNPX input file was constructed by considering the following:

- The data provided by the manufacture for the detector and samples
- The command F8: P card was considered in this study, to calculate the number of photons of energy line (185.7 keV or 1001 keV) deposited in detector
- The history cutoff (NPS) card was used and selected to achieve random statistical errors of better than (2%).

ISOCS™ calibration software was employed by considering the following:

- Samples counted using a Falcon 5000 (Model BE2830) detector that has been characterized by CANBERRA [4].
- Gamma spectra obtained from the detector acquired and analyzed via the Genie 2000 Software.
- Dimensions and material composition of the container, sample matrix and source-to-detector distance inserted into the Geometry Composer (ISOCS™ Calibration Software) and generate an efficiency calibration file.
- The efficiency calibration file was used for the analysis of the spectrum collected during the sample count.

The measured count rates at 185.7 and 1001.1 keV gamma energies, the calculated absolute full energy peak efficiency, using MCNPX modeling and ISOCS^{TM} calibration software, at the same energies, and the specific activities of the measured two gamma energy lines were substituted into Eq. (1) to obtain the ^{235}U and ^{238}U mass contents in nuclear materials.

Net counting rate C_R in a gamma-peak of certain energy due to any radioactive isotope as a function of ε_{ab} could be given as [6]:

$$\mathbf{C_R = M.\ S_a.\ \varepsilon_{ab}} \text{ -------------- } (1)$$

Where,

M is the mass of the assayed isotope in grams,

$\mathbf{S_a}$ is the specific activity of the measured gamma-photons with specified energy ($g^{-1}s^{-1}$),

$\boldsymbol{\varepsilon_{ab}}$ is the absolute full energy peak efficiency of the detector at the measured gamma energy

III. Results and discussion

1.1 Count rate measurements

The count rates (C_R) with the associated percentage relative uncertainties (σ_{CR}) are given in Table (1).

Table (1): Measured count rates of 185.7 and 1001.1 keV gamma energies due to ^{235}U and ^{238}U isotopes with associated uncertainties.

Sample Id	Enrichment (declared value) E% ± (σ_E/E)%	Samples-to-detector distance (cm)	Count rate $C_R \pm (\sigma_{CR}/C_R)\%$ (s^{-1})	
			185.7 keV	1001.1keV
031	0.31± 0.02	30	4.50 ± 3.34	2.99 ± 2.63
071	0.71± 0.05		10.36 ± 1.75	2.98± 2.70
194	1.94 ± 0.14		28.37 ± 0.93	3.05± 2.64
295	2.95 ± 0.21		43.58 ± 0.74	2.95 ± 2.68
446	4.46 ± 0.32		66.09 ± 0.59	2.93 ± 2.88

Relative uncertainties are relatively large; it was expected due to the short life times (t = 10 min) of measurement to simulate in-field measurements.

1.2 Absolute efficiency estimation

The results of the absolute full energy peak efficiency estimation with the associated percentage relative uncertainties are given in Table (2) for the five samples.

Table(2): Calculated absolute full energy peak efficiency of the detector at 185.7 and 1001.1 keV gamma energies with the associated uncertainties

Sample Id	Absolute photo peak efficiency $\varepsilon_{ab} \pm (\sigma_{\varepsilon ab}/\varepsilon_{ab})\%$			
	MCNPX-based		ISOCS^{TM}-based	
	185.7 keV	1001.1keV	185.7 keV	1001.1keV
031	1.84e-4 ± 0.74	1.73E-4 ± 0.98	1.88E-4 ± 8.56	1.70E-4 ± 4.01
071	1.83e-4 ± 1.65	1.72E-4 ± 1.71	1.88E-4 ± 8.62	1.71E-4 ± 3.99
194	1.85e-4 ± 1.65	1.74E-4 ± 1.39	1.90E-4 ± 8.58	1.71E-4 ± 4.00
295	1.87e-4 ± 1.16	1.73E-4 ± 1.20	1.89E-4 ± 8.62	1.71E-4 ± 4.00
446	1.88e-4 ± 1.03	1.75E-4 ± 1.07	1.91E-4 ± 8.59	1.73E-4 ± 4.01

As the uranium enrichment increase the estimated $\boldsymbol{\varepsilon}_{ab}$ is either approximately unchanged or slightly increased. All estimated uncertainties due to MCNPX calculations for all samples were less than 2%.

1.3 Estimated masses

Table (3) presents the ^{235}U and ^{238}U masses estimated based on MCNPX and ISOCS^{TM} estimation with the associated uncertainties.

Table (3): ^{235}U and ^{238}U masses estimated by MCNPX and ISOCS™ with the associated uncertainties

Sample Id	Estimated Isotopic Mass Content M (g) ± (σ_M/M) %			
	MCNPX-based		ISOCS™-based	
	^{235}U	^{238}U	^{235}U	^{238}U
031	0.531 ± 3.38	166.037 ± 2.80	0.520 ± 3.84	168.918 ± 3.61
071	1.228 ± 2.44	166.608 ± 3.18	1.198 ± 3.58	167.144 ± 3.62
194	3.339 ± 1.88	168.513 ± 2.98	3.246 ± 2.40	171.076 ± 2.45
295	5.069 ± 1.40	163.973 ± 2.93	5.012 ± 2.09	165.459 ± 2.10
446	7.638 ± 1.17	160.635 ± 3.07	7.522 ± 2.69	162.436 ± 2.84

The estimated uncertainty of the ^{235}U and ^{238}U masses content is due to the statistical error in the counting rate (it was less than 3.34% for ^{235}U and 2.88% for ^{238}U), and errors in MCNPX estimation (less than 2%). The uncertainties of ^{235}U masses in MCNPX-based estimation are found to be in the range from 1.17% to 3.38% and the uncertainties of ^{238}U masses are in the range from 2.80% to 3.18%.

The specific activity of the 185.7 and 1001.1 keV gamma-ray line add a relatively small error contribution to the uncertainty (less than 0.83%).

Table (4) presents the percentage relative uncertainties in the estimated ^{235}U masses for MCNPX estimation, ISOCS™ estimation and declared value. The relative differences between the masses estimated using the two techniques range between -0.165 and 1.48%.

Table(4): ^{235}U mass estimated by the described method in comparison with declared values

Sample Id	^{235}U Mass Content M (g) ± (σ_M/M) %				
	Declared value	MCNPX-based		ISOCS™-based	
		^{235}U	Relative Diff. %	^{235}U	Relative Diff. %
031	0.526±0.14	0.531 ± 3.38	-0.95	0.520 ± 3.84	1.14
071	1.208±0.14	1.228 ± 2.44	-1.65	1.198 ± 3.58	0.82
194	3.295±0.14	3.339 ± 1.88	-1.33	3.246 ± 2.40	1.48
295	5.004±0.15	5.069 ± 1.40	-1.29	5.012 ± 2.09	-0.15
446	7.572±0.13	7.638 ± 1.17	-0.87	7.522 ± 2.69	0.66

Figure 3 shows the estimated ^{235}U-mass content values with their uncertainties. It is clear that the estimated masses using both methods are in agreement with declared value within the uncertainties.

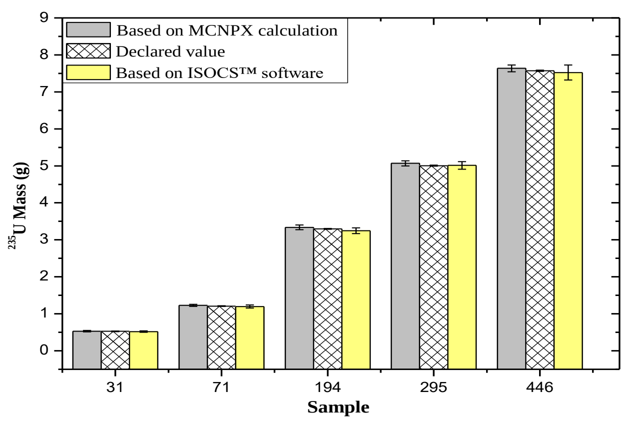

Figure 3: Estimated ^{235}U mass contents based on MCNPX calculation and ISOCS™ calibration in comparison with declared value.

Table (5) presents the percentage relative uncertainties in the estimated ^{238}U masses for MCNPX estimation, ISOCS^{TM} estimation and declared value. The relative differences between the masses estimated using the two techniques range between -2.81 and 1.83 %.

Table (5): ^{238}U mass estimated by the described method in comparison with declared values

Sample Id	^{238}U Mass Content M (g) ± (σ_M/M) %				
	Declared value	MCNPX-based		ISOCS^{TM}-based	
		^{238}U	Relative Diff. %	^{238}U	Relative Diff. %
031	169.144±0.167	166.037 ± 2.80	1.83	168.918 ± 3.61	0.13
071	168.473±0.167	166.608 ± 3.18	1.10	167.144 ± 3.62	0.78
194	166.386±0.167	168.513 ± 2.98	-1.27	171.076 ± 2.45	-2.81
295	164.677±0.167	163.973 ± 2.93	0.42	165.459 ± 2.10	-0.47
446	162.109±0.167	160.635 ± 3.07	0.90	162.436 ± 2.84	-0.20

Figure 4 shows the estimated ^{238}U-mass content values with their uncertainties. It is clear that the estimated masses using both methods are in agreement with declared value within the uncertainties.

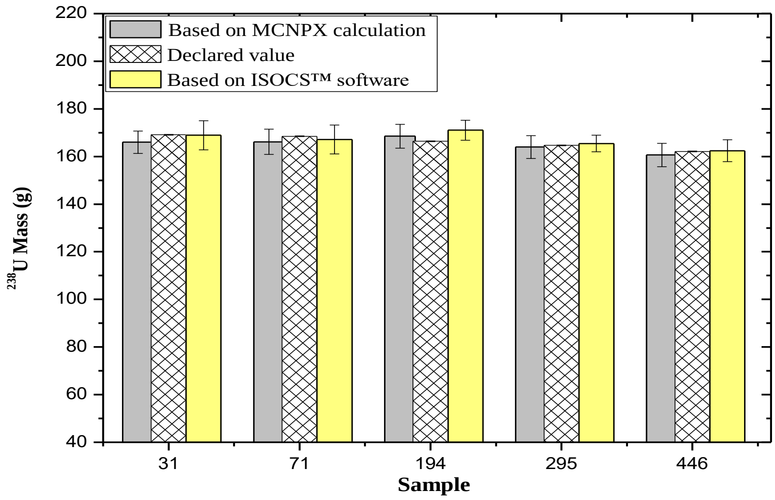

Figure 4: Estimated ^{238}U mass contents based on MCNPX calculation and ISOCS^{TM} calibration in comparison with declared value.

IV. Conclusions

This work describes a comparison of two gamma-ray efficiency estimation techniques for nuclear material verification, Canberra's ISOCS^{TM} and MCNP efficiency calibrations. the absolute full energy peak efficiency of the detector for five different U_3O_8 powder, with nominal ^{235}U abundances of 0.31, 0.71, 1.94, 2.95, and 4.46 mass percent, encased in cylindrical aluminum cans was estimated by the two different techniques.

ISOCS^{TM} geometry modeling was developed using the geometry composer feature of Canberra's Genie™ 2000 version 3.3 and Gamma Analysis version V3.3 software packages. MCNPX input files were designed to simulate each experimental setup configuration and calculate the absolute full energy peak efficiency of the detector for each verified NM.

The obtained results showed that the investigated techniques could be used to assay nuclear material samples in different enrichment with acceptable accuracy and precision. Factors that may affect the measurement or calculations were also investigated. These factors may include the measuring time which added an error of statistical nature and affects the estimated precision.

Acknowledgements

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