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Analysis of ISOCS Measurements to Establish Methodology Uncertainty

Susan Smith
Greg Nutter
Rachel Hunneke

September 2022

OAK RIDGE National Laboratory

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Nuclear Nonproliferation Division

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1. INTRODUCTION

Qualification measurements were performed at the US Department of Energy's (DOE's) Oak Ridge National Laboratory (ORNL) on a variety of measurement standards. These measurements will be used to qualify the In-Situ Object Counting System (ISOCS) measurement method with a high-purity germanium (HPGe) detector for use at ORNL. Measured items include uranium isotopic standards, filter papers coated with uranium oxide (U_3O_8), U_3O_8 line sources, and a uranium tetrafluoride (UF_4) source. The ^{235}U mass was estimated using the ISOCS calibration software for each source and compared with the declared isotope mass value.

1.1 DESCRIPTION OF ISOCS

ISOCS is a mathematical efficiency software used with gamma spectroscopy. It uses an efficiency calibration to quantify radioisotopes identified in a gamma spectrum. The efficiency of a measurement is dependent on the size, shape, and geometry of both the item and the detector; the material composition of the item; and the energy of the emitted gamma rays. The advantage of ISOCS is that a detection efficiency is determined without the need to manufacture several standards, even if such standards could be manufactured. [1, 2]

ISOCS begins with intrinsic detector efficiencies calculated by Monte Carlo simulations in all directions and over a broad range of energies (i.e., detector characterization). The Monte Carlo efficiency values are compared with eight source measurements using NIST-traceable sources. ISOCS v.4.4.1, which was evaluated for qualification, has detector efficiency values for gamma-ray energies ranging from 10 to 7,000 keV and distances ranging from on contact with the detector face out to 500 m. This version may be used with HPGe, NaI, and $LaBr_3$ detectors. [1, 2]

The user creates a measurement model starting from a basic geometry template in which details about the measurement are entered, such as dimensions, thickness, material composition, density, and fill height. The ISOCS software then uses a ray-tracing method to correct the intrinsic detector efficiency for the item characteristics and measurement geometry, determining the final efficiency of a gamma ray emitted from any location within the model.

1.2 INTENDED USE

This qualification is applicable to the use of HPGe detectors with the Genie 2000 Gamma Analysis and Acquisition software package v3.4.1 and ISOCS v4.4.1. It may be used for any gamma-emitter, but benchmarks were performed internally with only uranium items. Several publications included in the references show that the accuracy and precision of ISOCS is approximately ±10% for items with gamma-ray energies below 250 keV. At higher energies the accuracy and precision are generally 4–6%. [1, 2, 3, 4, 5, 6, 7, 8]

2. MEASUREMENT CONTROL

2.1 METHODOLOGY

Measurement control limits with a point source are established for each individual detector and shall be conducted before use. Results will be written in a registered logbook and entered in a spreadsheet for monitoring via control chart. The statistical control rules established in Department of Energy (DOE) Standard 1194-2019 [9], regarding warning and alarm limit violations, will be applied. If a control measurement exceeds the "Warning" limit, two repeat measurements will be performed. If either of the second measurements are outside the "Warning" limit, the detector owner will be contacted, and the detector will be removed from service until the issue has been identified and resolved. Similarly, if an "Alarm" limit is violated, the detector will immediately be removed from service until the issue has been identified and resolved.

2.2 MEASUREMENT GEOMETRY

A ^{137}Cs point source was used for measurement control. Two peaks were used to monitor the performance of the detector, the 32.2 keV peak, and the 661.6 keV peak. The control measurement was taken at the start of each day in which qualification measurements were taken. The source was placed 10 cm away from the face of the detector and measured for 120 s. The source was placed in a metal holder designed for a source of its size and centered with the detector face.

Figure 1. Measurement control setup.

When measurements were taken on the ISOCS cart and shield assembly, the source was placed 30 cm from the detector. To account for the reduced count rate, the count time was extended to 360 s. Figure 1 shows the setup for the control measurements taken at 30 cm.

2.3 DETERMINATION OF CONTROL LIMITS

Control limits for the HPGe detector were set according to measurable characteristics that demonstrate the functionality of the detector and electronics. Three characteristics were chosen: the peak full-width at half-maximum (FWHM), the peak centroid channel, and the peak centroid in energy. The FWHM is monitored to determine the effective operation of the HPGe detector. Monitoring of the peak centroid (i.e., in terms of energy) monitors the energy calibration and the linearity of the multi-channel analyzer. Additionally, monitoring the peak centroid channel informs the user about the stability of the electronics.

The FWHM is treated as a random variable that will follow a normal distribution. Thus, the mean and standard deviation of the peak FWHM was determined from the nine control measurements collected during the qualification measurement campaign. The warning limit was set as the mean plus 2 standard deviations and the alarm limit as the mean plus 3 standard deviations. Only upper limits are established because smaller FWHM values are not indicative of a malfunction. The control limits for the FWHM of the ^{137}Cs peaks are shown in Table 1.

Table 1. Control limits for the FWHM of ^{137}Cs peaks

Parameter	Value	
	32.2 keV peak	661.6 keV peak
Mean	0.809	1.263
Standard deviation	0.064	0.041
Warning limit	0.936	1.346
Alarm limit	1.000	1.387

In general, germanium detectors and their multi-channel analyzers are fairly stable. Therefore, the position of a gamma ray in the multi-channel analyzer does not fluctuate in a random fashion and control limits can be set as fixed upper and lower bounds instead of using the 2x and 3x standard deviation control limits. The control limits are selected based on analysis settings to ensure gamma rays of interest can be identified by the identification routines; the peak centroid must be within 1 keV of the nominal peak energy. Therefore, the control limits have been established so that the warning limits are the nominal peak energy ±0.5 keV and the alarm limits are ±1 keV. The control limits for the peak centroid in channels have been established based on simply the conversion of these limits into channels using the energy calibration. The control limits on the peak centroid, in both energies and channels, are presented in Table 2 and Table 3.

Table 2. Control limits for the peak centroid in energy (keV)

Nominal peak energy	Lower control limits		Upper control limits	
	Alarm	Warning	Warning	Alarm
32.2 keV	31.19	31.69	32.69	33.19
661.6 keV	660.66	661.16	662.16	662.66

Table 3. Control limits for the peak centroid in channels

Nominal peak energy	Expected channel	Lower control limits		Upper control limits	
		Alarm	Warning	Warning	Alarm
32.2 keV	87.89	85.15	86.52	89.26	90.63
661.6 keV	1,811.04	1,808.30	1,809.67	1,812.40	1,813.77

3. QUALIFICATION MEASUREMENTS

Qualification measurements were taken using a liquid nitrogen–cooled broad energy germanium detector (BEGe), serial number 13211, and an Inspector 2000 Digital Signal Processing Portable Spectroscopy Workstation, both produced by Canberra Industries (now Mirion Technologies). Data acquisition and

analysis for qualification measurements followed an internal operating procedure, summarized in Section 3.1.

Several measurement standards of varying forms and enrichments were used for the qualification measurements. Measured items included uranium enrichment standards, filter papers coated with U_3O_8, U_3O_8 line sources, and a UF_4 source. The known characteristics of the standards were used in the development of the ISOCS models.

3.1 OVERVIEW OF MEASUREMENT PROCEDURE

Measurements were taken using the BEGe detector, the Inspector 2000, and the Genie 2000 Gamma Acquisition and Analysis Software package. The hardware settings used for these measurements are shown in Table 4. The energy calibration was performed using three sources: ^{241}Am, ^{137}Cs, and ^{60}Co.

Table 4. Inspector 2000 settings used for qualification measurements

Parameter	Value
High voltage	+3,000 V
Conversion gain	8,192
Course gain	×10
Fine gain	1.3361
Input polarity	Negative
Pole zero	3,103

The detector was placed on a flat surface facing the center of the source being measured. A control measurement was performed each day for 120 s with a ^{137}Cs point source before any qualification measurements were taken. Most measured items were placed either 10 cm or 30 cm away, with a few exceptions, depending on the number of counts, dead time, and size of the item. The average dead time was under 25% for each item. Each qualification measurement was acquired for a live time of 600 s. The parameters for each measurement are defined in the following sections. Several background measurements were taken overnight for 12 h each.

After the measurements, the geometries were modeled with ISOCS using the template that best matched the measurement setup. An ISOCS model was created to match each measurement setup and source. The models should match the measurement geometry and item dimensions, material composition, and density. For each model, the energies at which efficiency values will be calculated are defined to include the primary gamma rays of interest. After the model is saved, the geometry is validated to verify that the dimensions were input correctly. The Geometry Composer Report can be used to check parameters for accuracy before the data points for the efficiency curve are generated.

To analyze the spectra, each measurement spectrum was analyzed using the Genie 2000 Gamma Acquisition and Analysis Software package. The following steps and parameters were used. Steps 1 and 2 were completed for each background spectrum.

1. Analyze | B Peak Locate |1 Unidentified 2nd Diff
    a. Set Start Channel to 100 and set Significance Threshold to 5.00. Execute.
2. Analyze | C Peak Area | Sum/Non-Linear LSQ Fit

a. Set Start Channel to 100, select 95% Critical level test, select Display ROIs, and select reject zero area peaks. Execute.

3. Analyze | E Area Correction | 1 Standard deviations. Bkg. Subtract

    a. Check the Yes option, select the corresponding background file, select 95% Critical level test. Execute.

4. Analyze | G Efficiency Correction | 2 ISOCS

    a. Select the corresponding ISOCS model. Execute.

5. Analyze | I Nuclide Identification | NID w/ Interf. Corr

    a. Set Start Channel to 100, select the desired NID library, select Generate Report. Execute.

6. Analyze | N Post ID Processing | 1 LACE

    a. Select Display Results and select Generate Report. Execute.

    b. If there is a noticeable trend in the LACE results (less than −0.5 or greater than 0.5), the ISOCS model should be adjusted and the previous steps repeated, starting at step 4.

7. Analyze | O Reporting | 1 Standard

    a. Select MASS.TPL for Template Name, select <All> for Section Name, and select New File. Execute.

Step 6 above refers to the Line Activity Consistency Evaluator (LACE) algorithm in Genie 2000. LACE calculates the ratio of the gamma peak activity to the weighted mean activity for all the gamma peaks of an isotope when more than one peak is present in the spectrum. The ratio and gamma energy are transformed to the natural log of each parameter. A linear fit is performed on the transformed data. The closer the slope of the line is to zero, the more accurate the ISOCS model will be [10].

All reports were saved. These analysis steps were repeated for every measurement spectrum with the corresponding ISOCS geometries.

3.2 MEASUREMENTS OF URANIUM ISOTOPIC REFERENCE STANDARDS

3.2.1 Description of Standards

The New Brunswick Laboratory (NBL) and National Bureau of Standards uranium standards used have enrichments ranging from depleted uranium to highly enriched uranium. These standards contain U_3O_8 in sealed aluminum cans. These Certified Reference Materials and Standard Reference Materials are intended for use for calibration and evaluation of gamma-ray counting for nondestructive assay of uranium isotopic analysis in uranium bulk materials. The associated certificates issued by New Brunswick Laboratory [11, 12] include certified values for the isotopic fractions of all the uranium isotopes, as well as their masses. Information on these standards is shown in Table 5, Figure 2, and Table 6.

Table 5. Material enrichment, mass, fill height and density of the uranium isotopic reference standards

Name	Enrichment (wt %)	^{235}U mass (g)	U_3O_8 fill height (mm)	U_3O_8 density (g/cm^3)

Standard A	0.31	0.52	20.8	2.5
Standard B	0.71	1.2	20.8	2.5
Standard C	1.94	3.28	20.8	2.5
Standard D	2.95	4.99	20.8	2.5
Standard E	4.46	7.54	15.8	3.29
Standard F	20.11	39.10	15.8	3.78
Standard G	52.49	101.72	15.8	3.78
Standard H	93.17	181.15	15.8	3.78

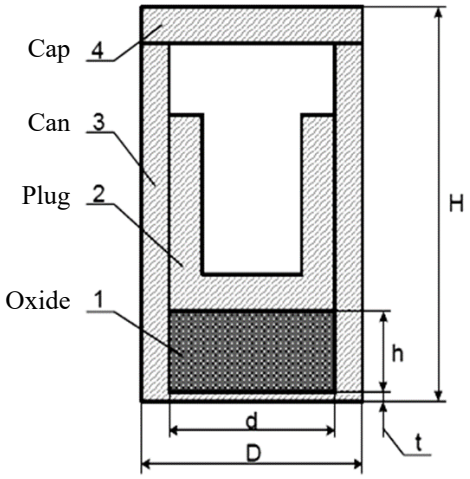

Figure 2. Dimensional drawing of the isotopic standards. The entire can is made of aluminum. The dimensions indicated by letters are H = total can height; h = powder chamber height; D = can diameter; d = powder chamber diameter; and t = bottom thickness.

Table 6. Dimensions of isotopic standards as stated in their certificates

Reference material	Can dimensions [a] (mm)				
	H	h	D	d	t
Standards A–D	100	20.8	80	70	2
Standard E	100	15.8	80	70	2
Standards F–H	100	15.8	80	70	2

[a] H = total can height, h = powder chamber height, D = can diameter, d = powder chamber diameter, and t = bottom thickness.

3.2.2 Measurement Geometries

Standards C and F were chosen to be measured 30 times each and the remaining 6 standards were measured once each. Each measurement had a live time of 600 s. Standards with enrichments below 20% were placed 10 cm away from the face of the detector. Standards with enrichments above 20% were placed 30 cm away. This method helped to keep the average dead time below 25%. The measurement setup for Standards F–H is shown in Figure 3.

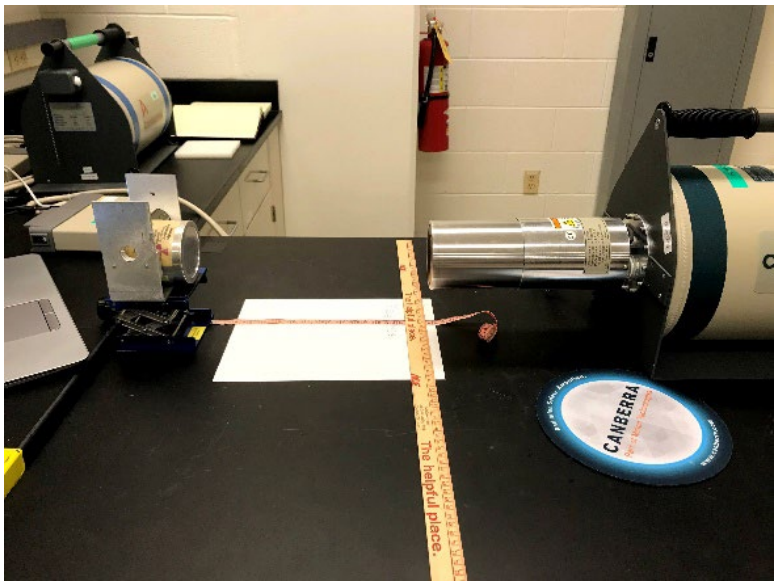

Figure 3. Side view of measurement setup for NBL Isotopic Standards F, G and H.
The source-to-detector distance is 30 cm.

3.2.3 ISOCS Models

The ISOCS models for the uranium isotopic reference standards, which are shaped like cans, were created using the circular plane template. Information and dimensions that remained the same for each of these models are shown in Table 7. There are three models for the reference standards: one for Standards A–D placed 10 cm away from the detector, one for Standards F–H placed 30 cm away from the detector, and a third for Standard E because the U_3O_8 layer differs from the other standards. The ISOCS geometry models are shown in Figure 4 and Figure 5. Appendix A includes the ISOCS model report.

Table 7. Dimensions for the uranium isotopic reference standards ISOCS models

Name	Value
Template	Circular plane
Side wall, layer 1	Aluminum (density of 2.7 g/cm^3)
Side wall thickness	5 mm
Side wall inside diameter	70 mm
Layer 1 thickness	2 mm
Layer 2 thickness	15.8 mm
Layer 1 relative concentration	0.00
Layer 2 relative concentration	100.00

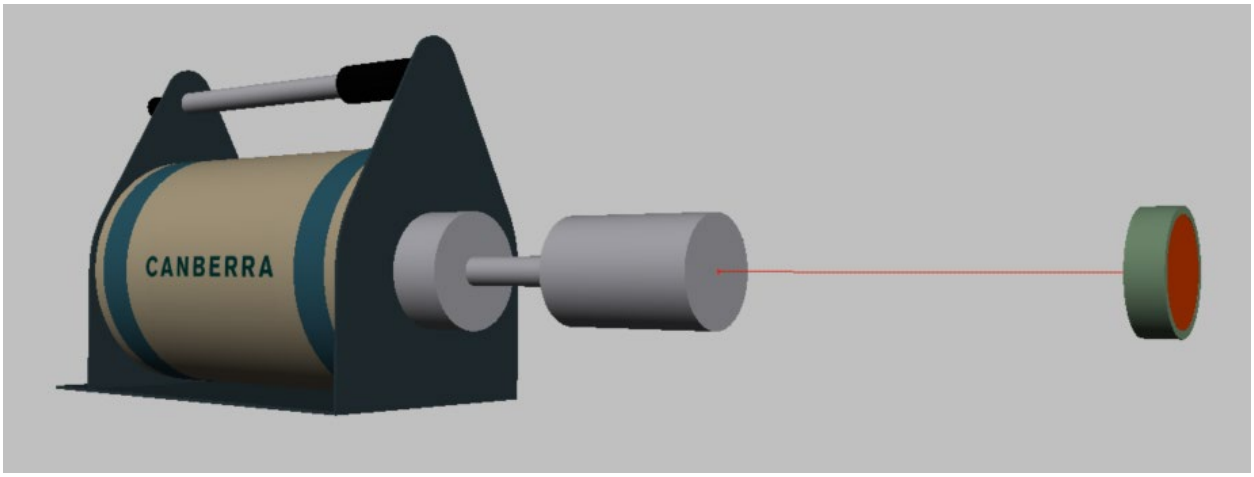

Figure 4. 3D visualization of the ISOCS model for Standards F, G, and H at 30 cm.

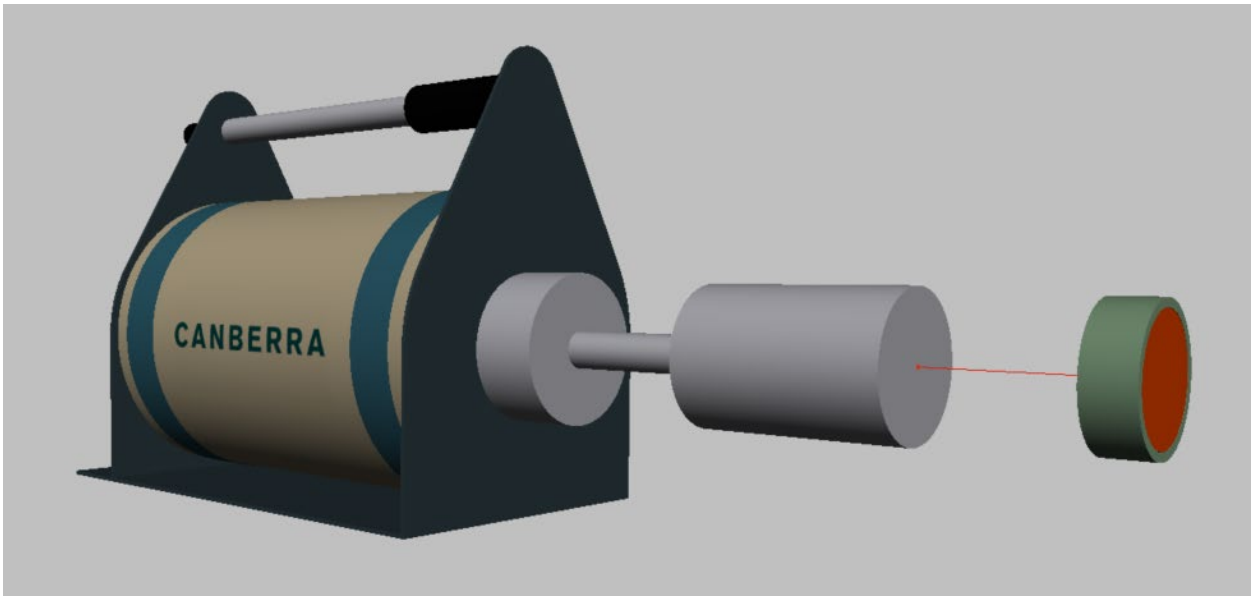

Figure 5. 3D visualization of the ISOCS models for Standards A–D at 10 cm.

3.3 UF_4 WORKING STANDARD

3.3.1 Description of Standard

The UF_4 working standard is a plastic vial containing UF_4 mixed with a slow-setting epoxy. The plastic vial is wrapped in ~2 mm of plastic because sealed sources require two layers of encapsulation. The vial is ~7 cm tall and 2 cm in diameter. This source has an enrichment of 93.17% and its total U mass is 11.372 g.

3.3.2 Measurement Geometry

The UF_4 vial was placed on a stand 10 cm directly in front of the detector, as shown in Figure 6. It was measured 30 times for a live time of 600 s each.

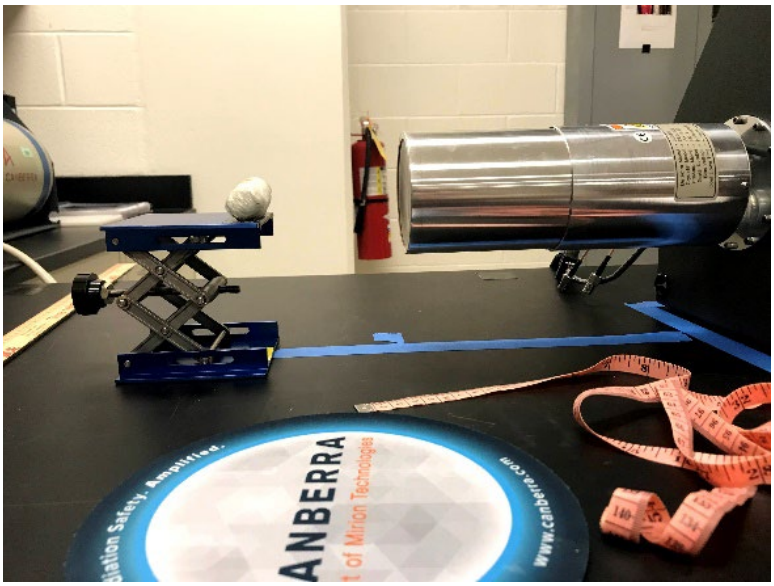

Figure 6. Measurement setup for the UF_4 working standard. The source-to-detector distance was 10 cm.

3.3.3 ISOCS Model

In ISOCS, the simple cylinder template was used to model the UF_4 vial. The model's dimensions are shown in Table 8. An absorber was placed between the source and the detector to represent the plastic surrounding the vial, as shown in Figure 7.

Table 8. Dimensions for the UF_4 working standard ISOCS model

Name	Value
Template	Simple cylinder
Source-to-detector distance	10 cm
Container wall thickness	0.25 cm
Container inside diameter	1.5 cm
Container inside height	2.5 cm
Container material	Polypropylene, C_3H_6 (density 0.91 g/cm^3)
Source height	2.5 cm
Source material	UF_4 (density 2.9 g/cm^3)
Source relative concentration	1.00
Absorber thickness	0.2 cm
Absorber material	Low-density polyethylene, C_2H_4 (density 0.2 g/cm^3)

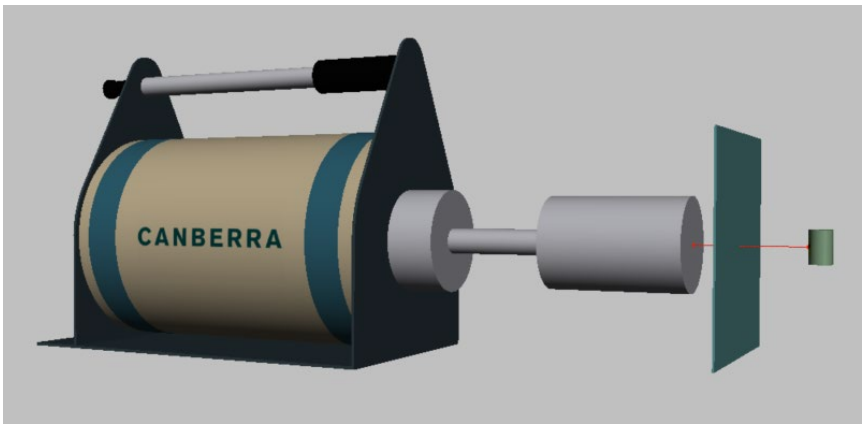

Figure 7. 3D visualization of the UF_4 working standard ISOCS model.

3.4 65% ENRICHED LINE SOURCES

3.4.1 Description of Standard

Several U_3O_8 line sources were used in different measurement setups. These long, thin sticks consist of U_3O_8 sealed in a plastic tube. Each line source is either 93.17% or 65.75% enriched and 30 cm long. Figure 8 shows an image of the 93.17% enriched line sources. The 65.75% enriched line sources look nearly identical.

Figure 8. Image of ten 93.17% enriched line sources.

3.4.2 Measurement Geometry

Three 65.75% enriched U_3O_8 line sources were placed in a line inside a copper pipe. The line sources were slightly overlapped so that the activity distribution looked like a continuous line. The pipe was centered horizontally in front of the detector. The pipe was 89 cm long and placed 50 cm away from the detector. A standard ISOCS shield/collimator assembly was used for this setup. For this item, 30 measurements were taken for a live time of 600 s each. Figure 9 shows the setup using the copper pipe.

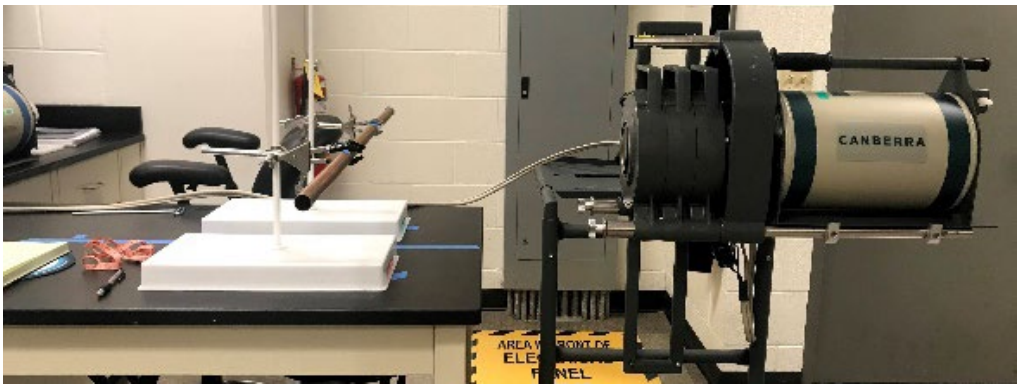

Figure 9. Measurement setup for copper pipe with U_3O_8 line sources.

3.4.3 ISOCS Model

The ISOCS model for the copper pipe setup used the pipe analysis template. The polypropylene casings of the sources were represented as a source layer coating the inside of the pipe with the relative concentration set to zero. The line sources were represented as an independent cylinder within the pipe and situated on the bottom using a radial and angular offset. The ISOCS shield is predefined by Mirion in the ISOCS software as "50mm-180d new". This collimator was applied to this model. The dimensions used in this geometry model are shown in Table 9 and the ISOCS geometry is shown in Figure 10.

Table 9. Dimensions for the 65%-line sources in the copper pipe ISOCS model

Layer	Dimension	Value
Model	Template	Pipe analysis
	Source-to-detector distance	50 cm
Pipe	Wall thickness	0.1 cm
	Inside diameter	1.705 cm
	Height above reference point	51 cm
	Height below reference point	51 cm
	Material	Copper (density of 8.96 g/cm^3)
Source 1	Thickness	0.1015 cm
	Height above reference point	45 cm
	Height below reference point	45 cm
	Material	Low-density polyethylene, C_2H_4 (density of 0.92 g/cm^3)
	Relative concentration	0.00

Table 9. Dimensions for the 65%-line sources in the copper pipe ISOCS model (continued).

Layer	Dimension	Value
Source 2	Diameter	0.432 cm
	Height above reference point	45 cm
	Height below reference point	45 cm
	Material	U_3O_8 (density of 0.55 g/cm^3)
	Relative concentration	1.00
Collimator	Cylindrical	50 mm–180d new

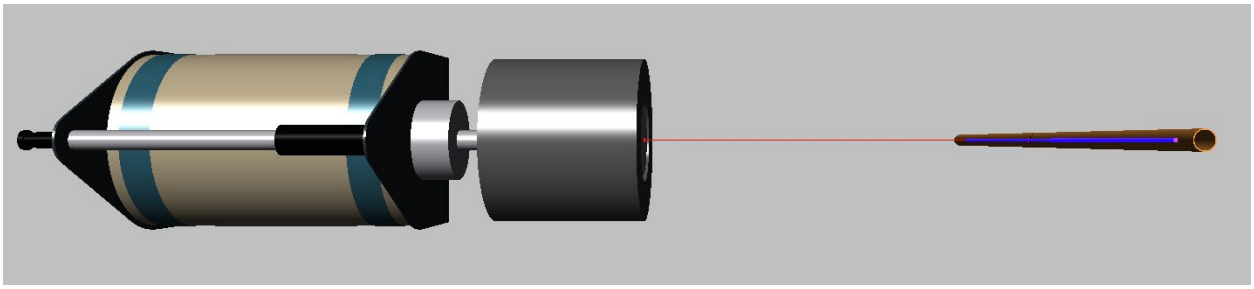

Figure 10. 3D visualization of the copper pipe ISOCS model. The small purple line represents the line sources within the copper pipe.

3.5 URANIUM FILTER PAPER STANDARDS

3.5.1 Description of Standards

Ten U_3O_8 filter papers, or cards, were stacked and measured together. The card sources were made of U_3O_8 that was spread on heavy paper, sealed in place with glue and paper folded over on itself, and heat-sealed in heavy plastic. The average enrichment of these cards was 93.17%, and the enrichment and ^{235}U mass of each card is shown in Table 10. The measured dimensions of each card were 51.5 × 28 × 0.3048 cm^3. The U_3O_8 did not cover the entire size of the card; its dimensions were 46 cm × 23 cm centered on each card. A representation of filter card deposition is shown in Figure 11.

Table 10. Enrichment and ^{235}U mass for each uranium oxide filter paper standard

Name	Enrichment (wt %)	^{235}U mass (g)
Card 1	93.1704	12.960
Card 2	93.1663	11.193
Card 3	93.1711	11.106
Card 4	93.1677	11.100
Card 5	93.1673	11.113
Card 6	93.1720	11.162
Card 7	93.1723	11.108
Card 8	93.1718	11.148
Card 9	93.1723	11.108
Card 10	93.1708	11.160

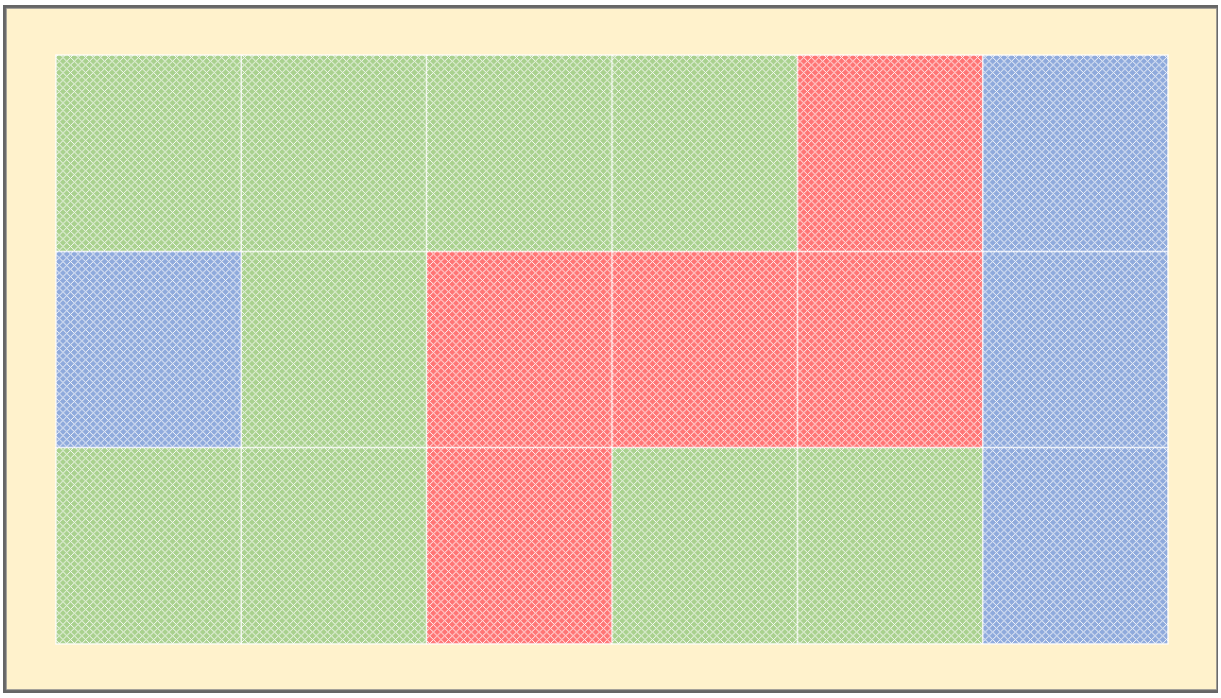

Figure 11. Representation of filter card deposition. The colors represent the concentration of U_3O_8 on each section of the card. Red is the highest concentration and green the lowest concentration. Notably, the U_3O_8 does not extend to the edges of the card.

3.5.2 Measurement Geometry

The ten filter paper standards were stacked together in numerical order and centered a distance away from the detector. The side and top views of the measurement setup are shown in Figure 12 and Figure 13. Two measurements were taken with this item: one at 37 cm from the front filter card to the detector face and the other at 50 cm.

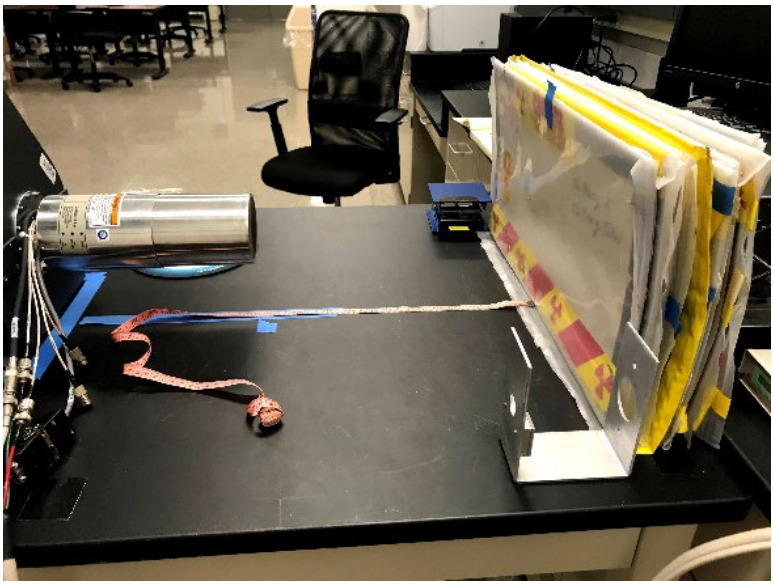

Figure 12. Side view of measurement setup for U_3O_8 filter paper standards at a source-to-detector distance of 37 cm.

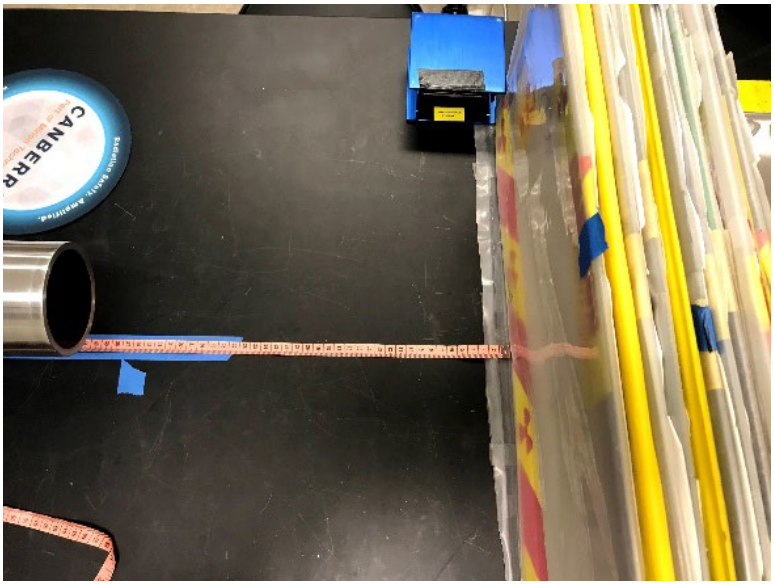

Figure 13. Top view of measurement setup of U_3O_8 filter paper standards at a source-to-detector distance of 37 cm.

3.5.3 ISOCS Model

The rectangular plane template was used to model the uranium filter paper standards in ISOCS. All ten card sources were added as a layer. The relative concentration for each layer represents the fraction of the total mass contained in that card. An absorber was placed between the source and the detector to model the plastic covering of the sources. Two models were created for this geometry: one with a source-to-detector distance of 37 cm and the other with a source-to-detector distance of 50 cm. The remaining dimensions for these models are shown in Table 11, and the 3D visualization of the ISOCS geometry for this setup is shown in Figure 14.

Table 11. Dimensions for the uranium filter paper standards ISOCS model

Layer	Material	Density (g/cm^3)	Relative concentration	Thickness (cm)	Inside width (cm)	Inside height (cm)
Side Walls	—	—	—	—	51.5	28
Absorber 1	Low-density polyethylene (C_2H_4)	0.92	—	0.08446	—	—
Layer 1	Cellulose ($C_6H_{10}O_5$)	0.9835	0.11	0.3048	—	—
Layer 2	Cellulose ($C_6H_{10}O_5$)	0.9858	0.10	0.3048	—	—
Layer 3	Cellulose ($C_6H_{10}O_5$)	0.9859	0.10	0.3048	—	—
Layer 4	Cellulose ($C_6H_{10}O_5$)	0.9859	0.10	0.3048	—	—
Layer 5	Cellulose ($C_6H_{10}O_5$)	0.9859	0.10	0.3048	—	—
Layer 6	Cellulose ($C_6H_{10}O_5$)	0.9858	0.10	0.3048	—	—
Layer 7	Cellulose ($C_6H_{10}O_5$)	0.9859	0.10	0.3048	—	—
Layer 8	Cellulose ($C_6H_{10}O_5$)	0.9858	0.10	0.3048	—	—
Layer 9	Cellulose ($C_6H_{10}O_5$)	0.9859	0.10	0.3048	—	—
Layer 10	Cellulose ($C_6H_{10}O_5$)	0.9858	0.10	0.3048	—	—

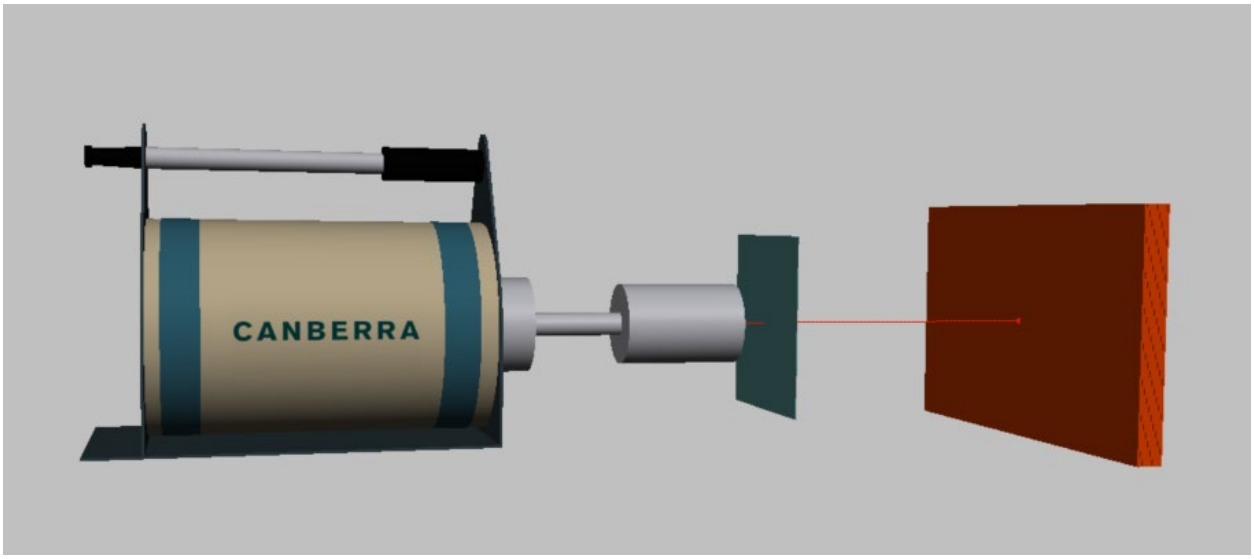

Figure 14. 3D visualization of uranium filter paper card standards ISOCS model.

3.6 URANIUM DISC STANDARD

3.6.1 Description of Standard

A uranium enrichment standard in the shape of a disc was measured. This uranium disc standard is typically used as a holdup standard. The disc consists of uranium metal alloy, with a radius of 1.6828 cm, sealed with epoxy in a 2.381-cm-radius stainless steel cavity behind a 0.159-cm-thick stainless steel window. There is a piece of stainless steel welded to the lip of the cavity to secure the epoxy seal and U_3O_8. The total diameter of the disc is 3 in. The uranium has an enrichment of 93.16% and a ^{235}U mass of 10.89 g. A diagram of this standard is shown in Figure 15.

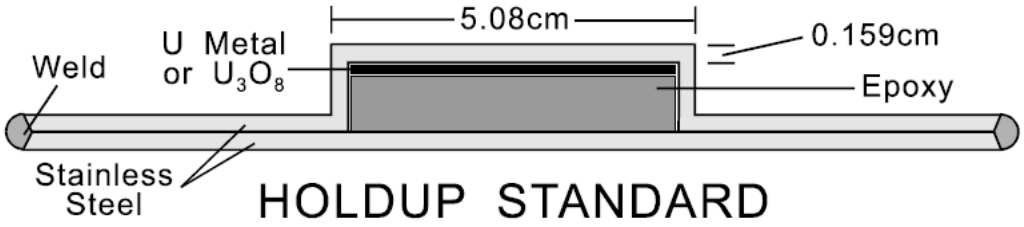

Figure 15. Side view diagram of the uranium disc standard.

3.6.2 Measurement Geometry

The uranium disc was placed on a stand and centered 15 cm directly in front of the detector. One measurement was taken for this item with a live time of 600 s. Figure 16 shows the measurement setup for the uranium disc.

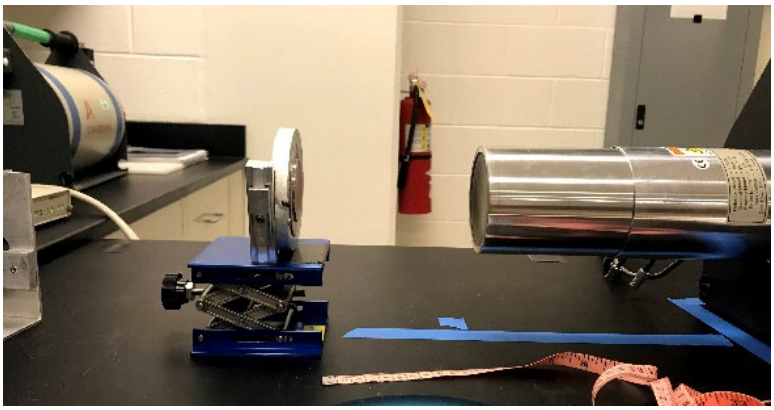

Figure 16. Measurement setup for the uranium disc standard. The source-to-detector distance is 15 cm.

3.6.3 ISOCS Model

The circular plane template was used to model the uranium disc standard in ISOCS. The source-to-detector distance was 15 cm. Layer 1 and the side walls were the stainless steel outside of the standard. Layer 2 was the uranium alloy metal. The dimensions for this ISOCS model are shown in Table 12 and the ISOCS geometry is shown in Figure 17.

Table 12. Dimensions of the uranium disc standard ISOCS model

Layer	Material	Density (g/cm^3)	Relative concentration	Thickness (cm)	Inside diameter (cm)
Side walls	Type 304 stainless steel[a]	8.02	—	0.8572	4.762
Layer 1	Type 304 stainless steel[a]	8.02	0.00	0.159	—
Layer 2	Al (0.10%), U (99.90%)	18.759	1.00	0.070134	—

[a] Type 304 stainless steel consists of the following: C (0.08%), Cr (19.00%), Fe (71.92%), and Ni (9.00%).

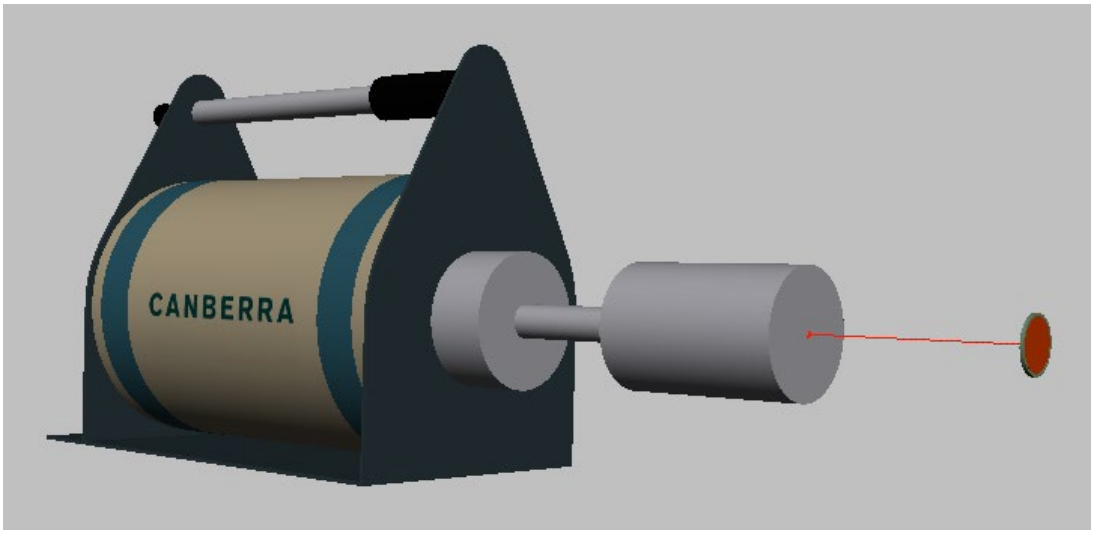

Figure 17. 3D visualization of the uranium disc standard ISOCS model.

3.7 PERFORMANCE

The gamma spectra were analyzed using Genie 2000 algorithms. The reported ^{235}U mass was compared with the declared or expected value from certificates or manufacturer descriptions. The measurement error was then calculated to determine how well the results agree. The measurement error serves as a parameter to demonstrate the accuracy of the analysis.

3.7.1 Initial Results

Replicate measurement results are graphed with error bars of one standard deviation and a line showing the declared or expected value. A second graph shows the measurement error from the expected mass value and the reported measurement uncertainty. These graphs are presented in Figure 18, Figure 19, Figure 20 and Figure 21. Table 13 and Table 14 present the measured mass, reported uncertainty, measurement error, and resulting bias. Full results are included in Appendix A.

Table 13. Average mass of UF_4 source, U_3O_8 standards, and U_3O_8 line sources in copper pipe and pipe array

Measurement ID	Declared ^{235}U mass (g)	Avg. measured ^{235}U mass (g)	Reported uncertainty (%)	Avg. measurement error (%)	Bias (g)
Standard C at 10 cm	3.295	3.51	5.1	6.58	0.22
Standard F at 30 cm	39.12	42.15	5.1	7.66	3.03
UF_4 vial	11.37	11.29	5.0	0.28	−0.08
Copper pipe	4.07	4.24	5.2	4.01	0.16

Table 14. Mass of uranium oxide standards, filter card sources, and uranium disc standard

Measurement ID	Declared ^{235}U mass (g)	Measured ^{235}U mass (g)	Reported uncertainty (%)	Measurement error (%)
Standard A	0.54	0.54	6.4	0.90
Standard B	1.21	1.35	5.3	11.61
Standard D	5.00	5.50	5.1	9.90
Standard E	7.57	8.21	5.0	8.41
Standard G	101.72	109.88	5.0	8.02
Standard H	181.15	191.80	5.0	5.88
U_3O_8 cards at 37cm	113.16	112.15	5.0	−0.89
U_3O_8 cards at 50 cm	113.16	111.00	5.0	−1.91
Uranium disc	10.89	12.30	5.0	12.86

The measured results for the ^{235}U mass were within twice the reported uncertainty of the declared values with only two exceptions. Standard B and the U disc measured results were within three times the reported uncertainty of the declared values.

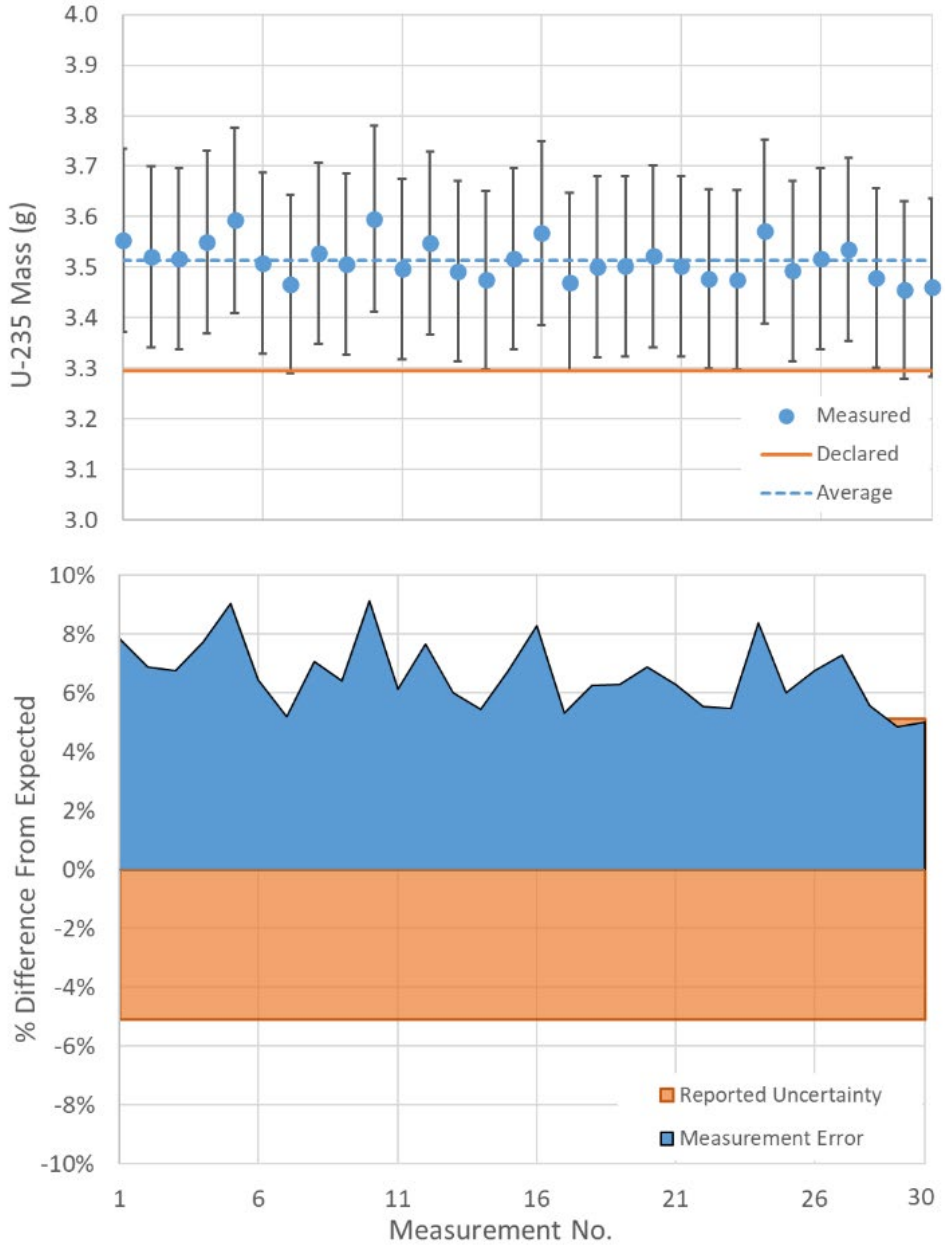

Figure 18. Results from replicate measurements of Standard C at 10 cm.

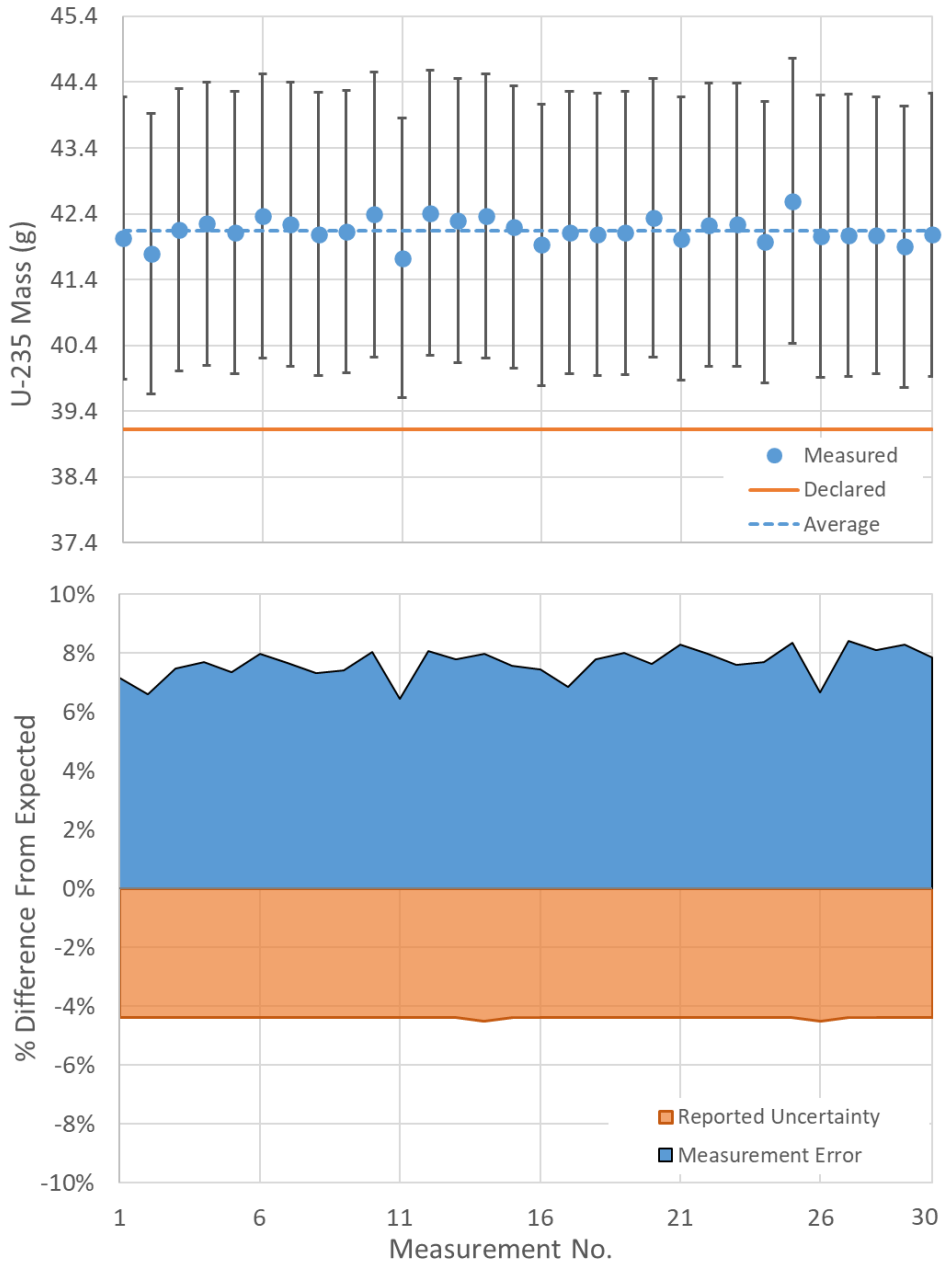

Figure 19. Results from replicate measurements of Standard F at 30 cm.

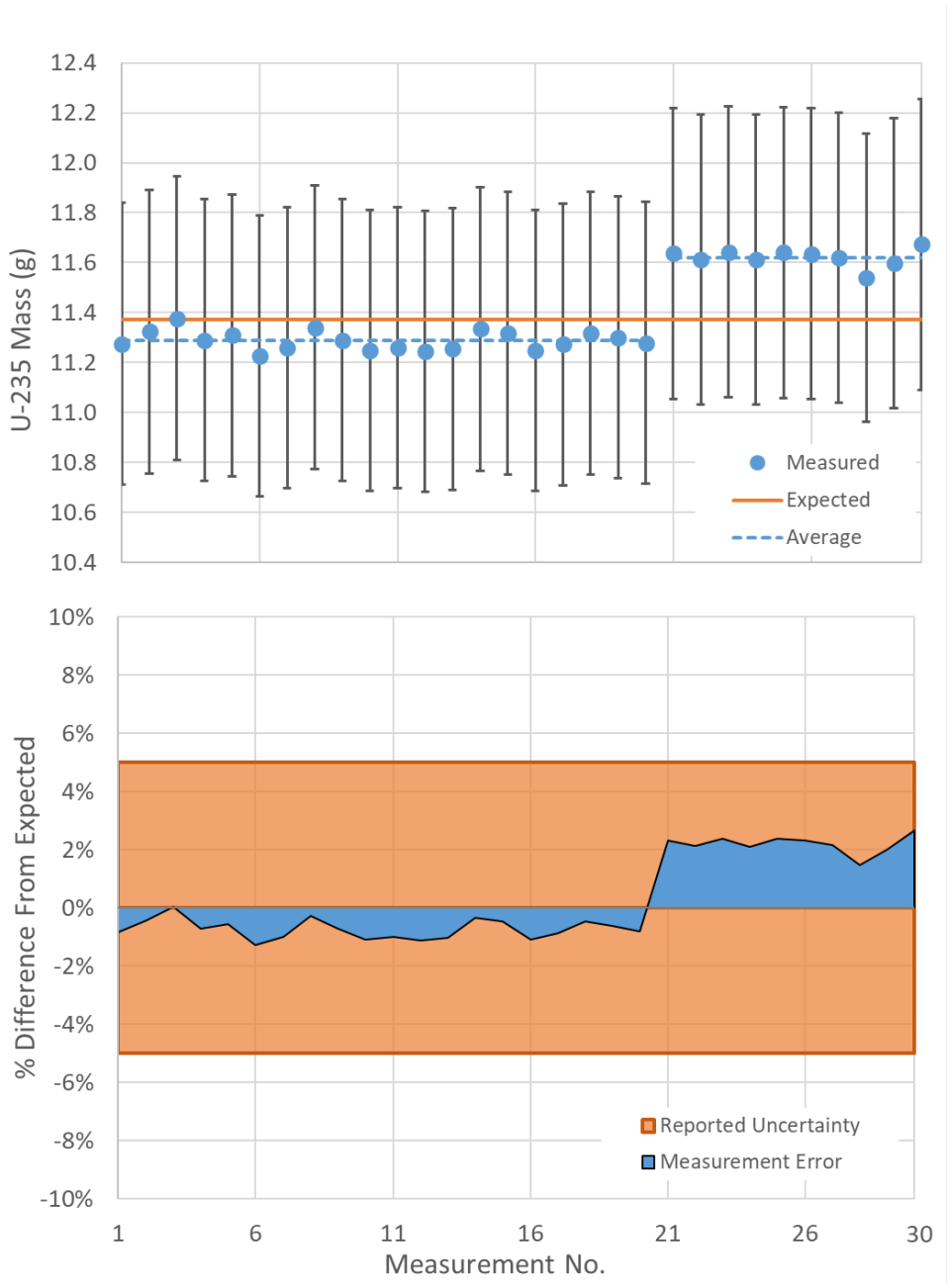

Figure 20. Results from replicate measurements of UF_4 vial.

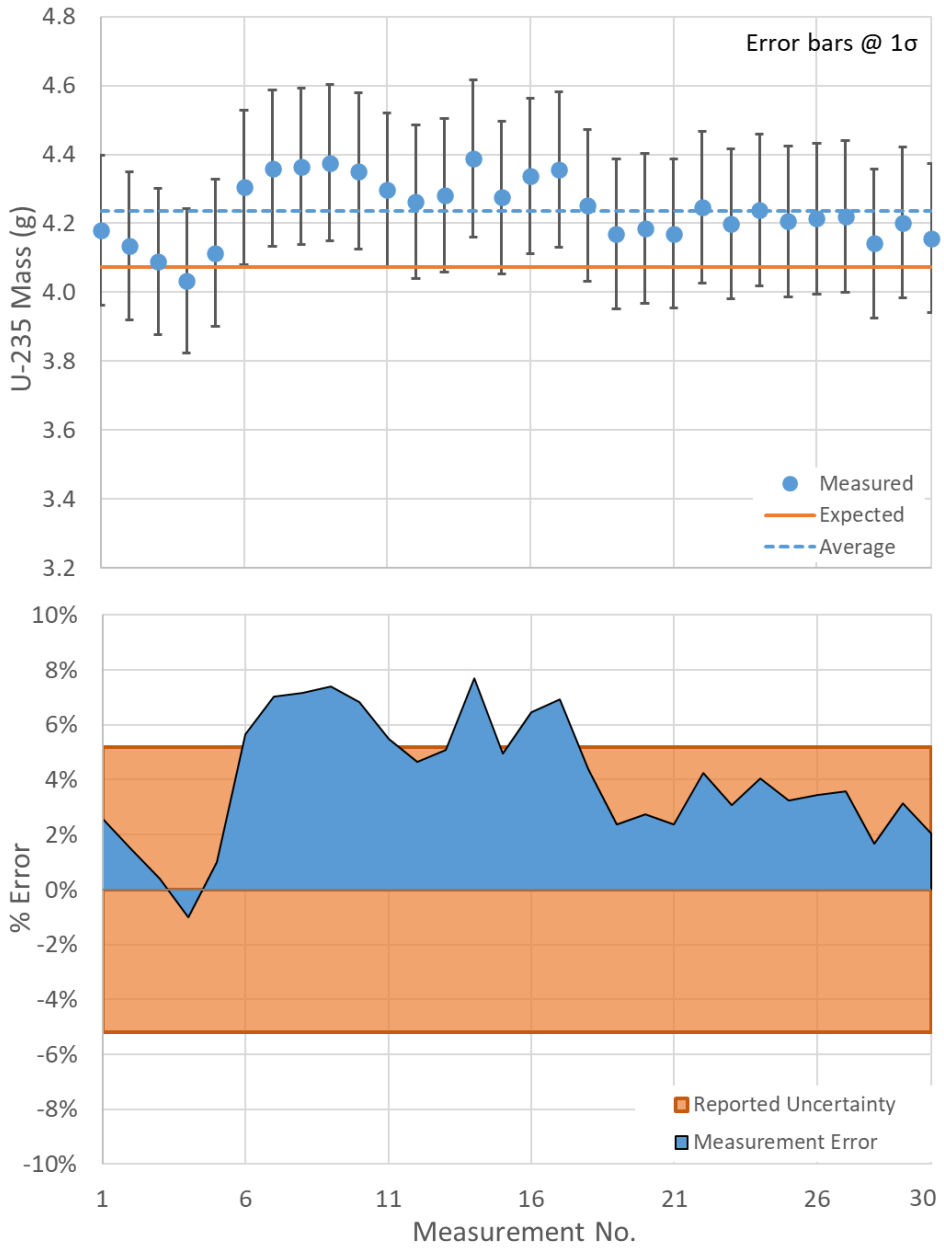

Figure 21. Results from replicate measurements of copper pipe.

3.7.2 Model Refinement

Review of the measurement results showed larger measurement errors than anticipated across most of the items measured repeatedly. Investigation into the measurement details and the fabrication of the isotopic reference standards revealed two additional pieces of information leading to the refinement of the ISOCS models for the UF_4 vial, Standard C and Standard F.

3.7.2.1 UF_4 Vial Source

In the case of the UF_4 vial, the percentage error of the measured ^{235}U mass was ~1% or less for the first 20 measurements but jumped to ~2% for the remaining measurements. In addition, the first 20 results were biased lower than the accepted value of 11.37 g, but the remaining 10 had a greater bias (see Figure 20).

This result was considerably different from the random scatter of measured values about the accepted value that was expected. The parameters chosen for the ISOCS model were taken from the measurement log and held constant for all analyses, and the measurement records were examined for a likely cause for the error. Logbook entries revealed that the first 20 measurements were taken during a single shift, and the remaining 10 were taken the following day. There was no indication that the detector had been moved during the 2 days or that the sample was moved after it had been set for the measurements. Because of the step change in the bias, the team aimed to find out if the sample was not set at the same distance from the detector on the second day. The distance between the detector face and the sample in the model was varied in the ISOCS model and the final 10 measurements were reanalyzed. If the measurement distance was changed from 10 cm to 9.8 cm, then the isotope mass results would agree better. This measurement distance was constant for all 10 final measurements, and no parameters were changed for the first 20 measurements (see Figure 22).

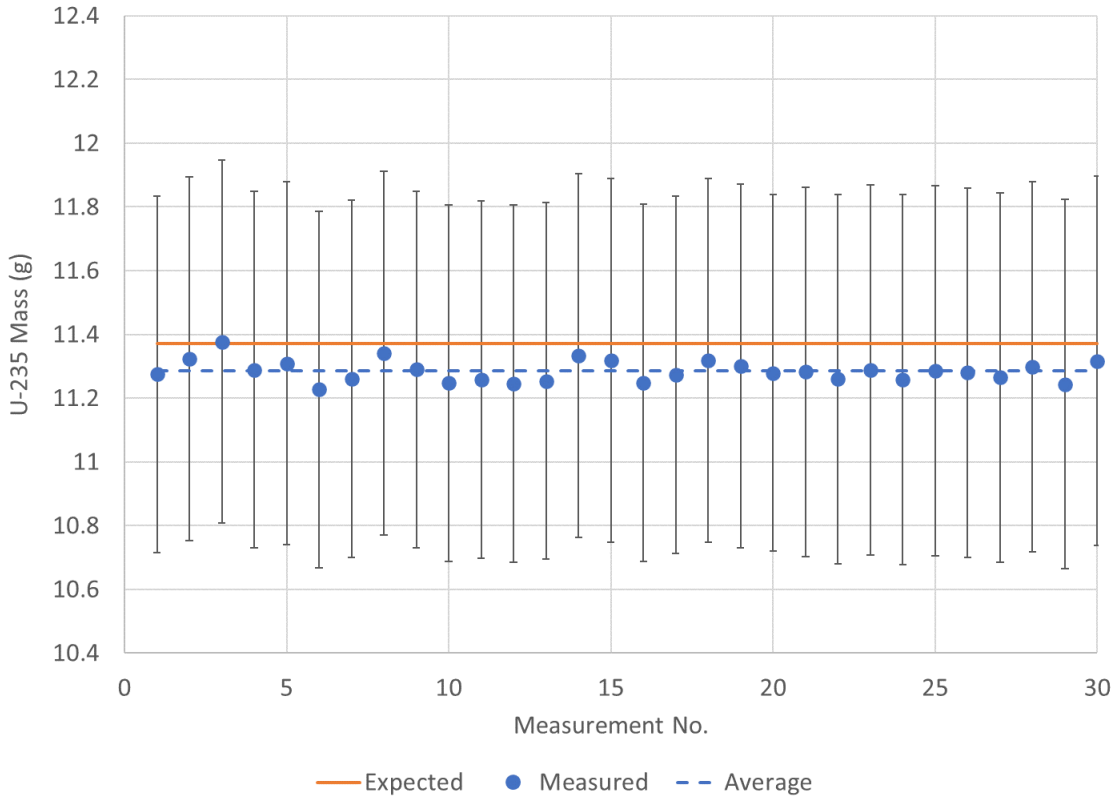

Figure 22. Analysis results using second ISOCS model for UF_4 vial.

The overall measurement bias changed from 0.03 to −0.09 when the source-to-detector distance was changed from 10 cm to 9.8 cm. However, the standard deviation of the measurement results dropped from 0.1656 to 0.0334, and the variation dropped from 0.0274 to 0.0011. This result supported the proposition that the measurement distance on the second day was different from that of the first day.

The source of the measurement bias remains unresolved. A difference of only 0.2 cm in the placement of the sample seemed to lead to the distinct difference in measurement results between the 2 days. A measuring tape was used to set the measurement distance. If the distance was scaled incorrectly, then it is possible that the sample was not set accurately on either day. The measurement distance was changed again in the model, and all the results were reanalyzed with the intention of reducing biases and errors. A measurement distance of 10.04 cm was selected for the first 20 measurements and 9.85 cm for the last 10 measurements. The results of the isotope mass calculations were distributed more evenly about the true value, and the expected improvements in the bias and error were achieved (see Figure 23). The average measurement error dropped to 0.08%, the bias dropped to −0.01, and the variance remained essentially unchanged.

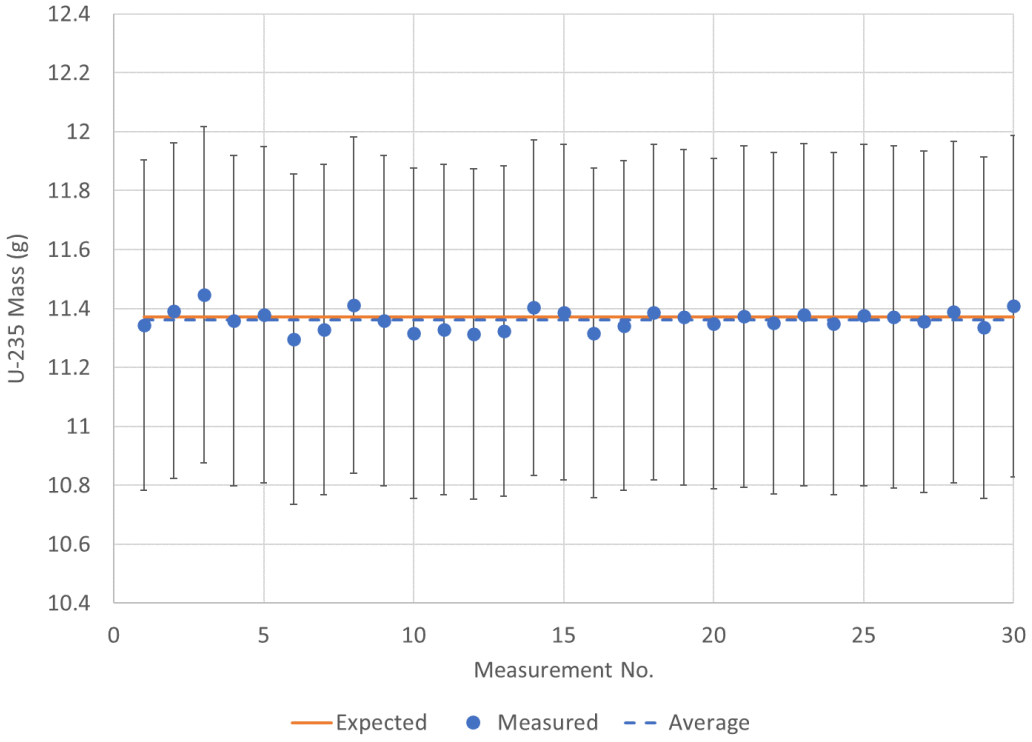

Figure 23. Analysis results using third ISOCS model UF_4 vial.

Table 15. Summary of refined ISOCS analysis for UF_4 vial source

Analysis	Declared ^{235}U mass (g)	Avg. measured ^{235}U mass (g)	Reported uncertainty (%)	Avg. measurement error (%)	Bias (g)
Initial	11.37	11.29	5.0	0.29	−0.08
Second	11.37	11.28	5.0	−0.75	−0.09
Third	11.37	11.36	5.0	−0.08	−0.01

3.7.2.2 Uranium Isotopic Reference Standards

The other analyses indicating large measurement errors were examined next. The team assumed that using the same measuring tape could easily lead to inaccurate measurement standoff. The standoff parameter was altered for the ISOCS models that were used for the isotopic standards and the uranium disc. The correct isotope mass could be calculated by changing the standoff parameter in the model by about a centimeter or sometimes less. This was determined to be well within a user error of ±2 cm using the flexible tape. These results reinforced the necessity of accurately determining the sample's position relative to the detector. This point is clearly demonstrated by the refinement to the uranium disc model from a measurement distance of 15 cm to 13.9 cm. By reducing the source-to-detector distance, the measured mass is now 10.92 g and there is a measurement error of 0.25%.

However, other factors necessary for a reliable analysis had not been addressed. Accurately accounting for attenuation of the gamma energies of interest was one of the most consequential of these factors. Success in attenuation correction is indicated by the slope of the LACE plot of selected energies of the isotope. As a rule of thumb, the closer this slope is to zero, the better the model reflects the real sample and the more reliable the analysis. Few standards are as well characterized as the isotopic reference standards (i.e., Standards A–H); that meticulous characterization is the reason they were chosen for reanalysis with the intent of decreasing the LACE slope. In the first reanalysis of Standard F, the average of the LACE slopes for all 30 measurements was 0.054 with a variability of 0.00197 and a standard deviation of 0.04436. Adjusting the position of the standard in relation to the detector could have corrected the measurement bias, but the adjustment had negligible effects on the LACE slopes unless the position parameters were set at values that would reflect irresponsible sample placement by the operator. The design and construction of the standards was examined to determine if it could improve the ISOCS model.

All these standards are constructed of a machined aluminum can (70 mm I.D.) wherein the U_3O_8 powder is deposited. An aluminum spacer is set on top of the U_3O_8 powder and a machined aluminum plug. A hydraulic hand press is used to uniaxially compress the assembly, which is welded together to set the geometry and seal in the U_3O_8 powder. The dimensions of the aluminum pieces are documented in the certificate of analysis from NBL. They are easily modeled and leave little freedom for adjusting the attenuation correction.

The density of the U_3O_8 powder was considered next. The initial model assumed homogeneous density throughout the U_3O_8 mass. Previous research studies on the dry pressing of ceramic powders for sintering indicated that this might not be an accurate assumption. Powder pressing processes are well established, and many precautions are taken to avoid macroscopic density gradients in the powder compact. Two of these precautions recommended by Ewsuk et.al. [13], (i.e., using a small surface area and avoiding use of a single-action uniaxial press) were apparently not followed in the fabrication of these standards. A uniaxially pressed ceramic powder cylinder with a smaller diameter than that of the standards was analyzed at Sandia National Laboratories (SNL), and distinct density gradients were found throughout the body of the powder compact [14]. Therefore, a model with a more complex internal density was developed to determine if it could yield any gain in attenuation compensation.

It was not practicable to try to replicate anything like the density gradients discovered at SNL in the U_3O_8 powder of the standards. Researchers at SNL used ultrasonic scanning to map a surprisingly complex pattern in their sample [14], but such scanning was not available to the team nor does ISOCS have the capability of replicating such complexity. A simple model that has denser zones near the plug and window ends of the can was developed instead. The depth of the three zones and the density of the middle zone were adjusted to maintain the bulk density of the material at the certified values. This model was tested using all the isotopic reference standard measurements. The average of the LACE slopes decreased

by nearly 19% with this change. A slight bias in the measurement results also developed, so the measurement distance was altered slightly to compensate for it. The average of the LACE slopes remained positive, indicating further improvements were possible. A simpler model was created with only a single larger and denser area adjacent to the window and applied to the Standard F measurements. The dimensions of the two areas and the density of the larger area were again adjusted to maintain the listed bulk density of the material. This approach yielded a larger drop in the average LACE slope of about 27%. No further improvements were attempted, but the average of the slopes remained slightly positive. The dimensions of the final ISOCS model are presented in Table 16.

Table 16. Dimensions for the refined ISOCS models of the uranium isotopic reference standards

Name	Value
Template	Circular plane
Side wall, layer 1	Aluminum (density of 2.7 g/cm^3)
Side wall thickness	5 mm
Side wall inside diameter	70 mm
Layer 1 thickness	2 mm
Layer 2 thickness	0.02 mm
Layer 3 thickness	15.78 mm
Layer 1 relative concentration	0.00
Layer 2 relative concentration	0.20
Layer 3 relative concentration	99.80

Table 17 lists results that support incorporating complexity in the material density of the model will result in those substantial gains in attenuation correction if appropriate to the item. Guidance for approximating that complexity can be found in the knowledge base of other disciplines.

Table 17. Mass of U_3O_8 standards

Measurement ID	Declared ^{235}U mass (g)	Measured ^{235}U mass (g)	Reported uncertainty (%)	Measurement error (%)
Standard A	0.54	0.54	6.4	0.57
Standard B	1.21	1.22	5.0	0.89
Standard C	3.30	3.29	5.0	−0.17
Standard D	5.00	5.03	5.0	0.55
Standard E	7.57	7.54	5.0	−0.34
Standard F	39.10	39.15	5.0	0.12
Standard G	101.72	102.09	5.0	0.36
Standard H	181.15	181.44	5.0	0.16
U Disc	10.89	10.92	5.0	0.24

Based on this analysis, the density profile of the material can contribute ~11% measurement uncertainty.

4. SUMMARY

The measurements performed at ORNL will be used to qualify the ISOCS measurement method with an HPGe detector. Several items were used for the measurements, including uranium isotopic standards, filter papers coated with U_3O_8, U_3O_8 line sources, and a UF_4 source. The ISOCS calibration software was used to estimate the ^{235}U mass for each source and was compared with the declared isotope mass value.

Using the available information on the construction and characteristics of available uranium standards, as well as the documented measurement geometry, the measured mass agreed with the declared mass within twice the reported uncertainty for most items. The largest measurement error was ~12.5%.

The ISOCS models were refined when the initial analysis determined that the measurement geometry was not always consistent with the documented distance. The material density profile for the isotopic standards was also modified after research determined that powder is not uniform, depending on how it is compacted. After these refinements, the largest measurement error was <2%.

The recommended accuracy of ISOCS has been previously identified as ±10% for gamma-ray energies lower than 250 keV, based on published data, and this work supports this generalization. It is recommended that the measurement uncertainty be recorded as two times the reported uncertainty. Furthermore, this work demonstrates that when ISOCS is used, the accuracy is only as good as the information available on the item and measurement geometry.

5. REFERENCES

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APPENDIX A. TABLES OF RESULTS

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Table 18. Full measured isotope mass results for the UF_4 vial source—initial analysis

Measurement number	Measured isotope mass (g)	Measurement relative uncertainty (%)	Measurement relative error (%)
1	11.28	5.0	−0.85
2	11.32	5.0	−0.43
3	11.38	5.0	0.04
4	11.29	5.0	−0.73
5	11.31	5.0	−0.55
6	11.23	5.0	−1.28
7	11.26	5.0	−0.98
8	11.34	5.0	−0.27
9	11.29	5.0	−0.72
10	11.25	5.0	−1.10
11	11.26	5.0	−1.00
12	11.25	5.0	−1.12
13	11.25	5.0	−1.04
14	11.33	5.0	−0.33
15	11.32	5.0	−0.47
16	11.25	5.0	−1.09
17	11.27	5.0	−0.87
18	11.32	5.0	−0.47
19	11.30	5.0	−0.62
20	11.28	5.0	−0.82
21	11.64	5.0	2.33
22	11.61	5.0	2.13
23	11.64	5.0	2.38
24	11.61	5.0	2.11
25	11.64	5.0	2.37
26	11.64	5.0	2.31
27	11.62	5.0	2.17
28	11.65	5.0	2.47
29	11.60	5.0	1.99
30	11.67	5.0	2.65

Table 19. Full measured isotope mass results for Standard F—initial results

Measurement number	Measured isotope mass (g)	Measurement relative uncertainty (%)	Measurement relative error (%)
1	42.03	4.4	7.17
2	41.79	4.4	6.61
3	42.16	4.4	7.48
4	42.25	4.4	7.69
5	42.11	4.4	7.36
6	42.37	4.4	7.97
7	42.24	4.4	7.66
8	42.09	4.4	7.32
9	42.13	4.4	7.40
10	42.39	4.4	8.02
11	41.73	4.4	6.45
12	42.41	4.4	8.08
13	42.29	4.4	7.79
14	42.37	4.5	7.97
15	42.20	4.4	7.56
16	41.93	4.4	7.44
17	42.12	4.4	6.83
18	42.09	4.4	7.77
19	42.11	4.4	8.00
20	42.34	4.4	7.64
21	42.02	4.4	8.30
22	42.23	4.4	7.97
23	42.24	4.4	7.59
24	41.97	4.4	7.68
25	42.60	4.4	8.35
26	42.06	4.5	6.67
27	42.07	4.4	8.42
28	42.07	4.4	8.11
29	41.90	4.4	8.30
30	42.08	4.4	7.86

Table 20. Full measured isotope mass results for Standard C—initial results

Measurement number	Measured isotope mass (g)	Measurement relative uncertainty (%)	Measurement relative error (%)
1	3.55	5.1	7.84
2	3.52	5.1	6.86
3	3.52	5.1	6.76
4	3.55	5.1	7.73
5	3.59	5.1	9.04
6	3.51	5.1	6.46
7	3.47	5.1	5.20
8	3.53	5.1	7.07
9	3.51	5.1	6.40
10	3.60	5.1	9.14
11	3.50	5.1	6.12
12	3.55	5.1	7.66
13	3.49	5.1	5.99
14	3.47	5.1	5.44
15	3.52	5.1	6.74
16	3.57	5.1	8.27
17	3.47	5.1	5.33
18	3.50	5.1	6.24
19	3.50	5.1	6.28
20	3.52	5.1	6.89
21	3.50	5.1	6.29
22	3.48	5.1	5.53
23	3.47	5.1	5.46
24	3.57	5.1	8.37
25	3.49	5.1	6.00
26	3.52	5.1	6.74
27	3.54	5.1	7.29
28	3.48	5.1	5.57
29	3.45	5.1	4.85
30	3.46	5.1	5.01

Table 21. Full measured isotope mass results for 65% enriched line sources in a copper pipe

Measurement number	Measured isotope mass (g)	Measurement relative uncertainty (%)	Measurement relative error (%)
1	4.18	5.2	2.58
2	4.13	5.2	1.49
3	4.09	5.2	0.39
4	4.03	5.2	−1.00
5	4.11	5.2	1.00
6	4.30	5.2	5.67
7	4.36	5.2	7.03
8	4.37	5.2	7.16
9	4.38	5.2	7.41
10	4.35	5.2	6.82
11	4.30	5.2	5.48
12	4.26	5.2	4.64
13	4.28	5.2	5.10
14	4.39	5.2	7.72
15	4.28	5.2	4.94
16	4.34	5.2	6.46
17	4.36	5.2	6.93
18	4.25	5.2	4.38
19	4.17	5.2	2.36
20	4.18	5.2	2.73
21	4.17	5.2	2.37
22	4.25	5.2	4.24
23	4.20	5.2	3.07
24	4.24	5.2	4.04
25	4.21	5.2	3.26
26	4.21	5.2	3.45
27	4.22	5.2	3.59
28	4.14	5.2	1.66
29	4.20	5.2	3.16
30	4.16	5.2	2.05

Table 22. Full measured isotope mass results for the UF_4 vial source—final results

Measurement number	Measured isotope mass (g)	Measurement relative uncertainty	Measurement relative error
1	11.34	5.00	−0.23
2	11.39	5.00	0.19
3	11.45	5.00	0.68
4	11.36	5.00	−0.11
5	11.38	5.00	0.08
6	11.30	5.00	−0.65
7	11.33	5.00	−0.36
8	11.41	5.00	0.36
9	11.36	5.00	−0.10
10	11.32	5.00	−0.47
11	11.33	5.00	−0.37
12	11.31	5.00	−0.49
13	11.32	5.00	−0.41
14	11.40	5.00	0.29
15	11.39	5.00	0.15
16	11.32	5.00	−0.47
17	11.34	5.00	−0.25
18	11.39	5.00	0.15
19	11.37	5.00	0.00
20	11.35	5.00	−0.19
21	11.37	5.00	0.03
22	11.35	5.00	−0.18
23	11.38	5.00	0.08
24	11.35	5.00	−0.18
25	11.38	5.00	0.06
26	11.37	5.00	0.01
27	11.36	5.00	−0.13
28	11.39	5.00	0.16
29	11.34	5.00	−0.31
30	11.41	5.00	0.33

Table 23. Full measured isotope mass results for Standard F—final results

Measurement number	Measured isotope mass (g)	Measurement relative uncertainty (%)	Measurement relative error (%)
1	39.02	5.00	−0.20
2	38.83	5.00	−0.70
3	39.14	5.00	0.10
4	39.23	5.00	0.34
5	39.09	5.00	−0.02
6	39.33	5.00	0.59
7	39.22	5.00	0.31
8	39.10	5.00	0.00
9	39.13	5.00	0.08
10	39.38	5.00	0.72
11	38.77	5.00	−0.85
12	39.41	5.00	0.79
13	39.28	5.00	0.47
14	39.37	5.00	0.68
15	39.18	5.00	0.21
16	38.94	5.00	−0.42
17	39.13	5.00	0.07
18	39.08	5.00	−0.05
19	39.15	5.00	0.12
20	39.34	5.00	0.61
21	39.07	5.00	−0.08
22	39.23	5.00	0.34
23	39.25	5.00	0.39
24	38.99	5.00	−0.29
25	39.55	5.00	1.16
26	39.09	5.00	−0.03
27	39.08	5.00	−0.04
28	39.08	5.00	−0.06
29	38.91	5.00	−0.50
30	39.11	5.00	0.03

Table 24. Full measured isotope mass results for Standard C—final results

Measurement number	Measured isotope mass (g)	Measurement relative uncertainty (%)	Measurement relative error (%)
1	3.33	5.00	0.99
2	3.30	5.00	0.05
3	3.29	5.00	−0.05
4	3.32	5.00	0.83
5	3.36	5.00	2.08
6	3.28	5.00	−0.35
7	3.25	5.00	−1.49
8	3.30	5.00	0.29
9	3.28	5.00	−0.36
10	3.37	5.00	2.21
11	3.27	5.00	−0.62
12	3.32	5.00	0.80
13	3.27	5.00	−0.78
14	3.25	5.00	−1.27
15	3.29	5.00	−0.06
16	3.34	5.00	1.36
17	3.25	5.00	−1.38
18	3.28	5.00	−0.54
19	3.28	5.00	−0.48
20	3.30	5.00	0.09
21	3.28	5.00	−0.49
22	3.26	5.00	−1.17
23	3.25	5.00	−1.25
24	3.34	5.00	1.48
25	3.27	5.00	−0.76
26	3.29	5.00	−0.04
27	3.31	5.00	0.47
28	3.26	5.00	−1.14
29	3.23	5.00	−1.83
30	3.24	5.00	−1.69