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VERIFICATION MEASUREMENTS OF THE MASS AND ENRICHMENT OF URANIUM OXIDE CARD SOURCES

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ABSTRACT

Measurements were performed to determine the mass and enrichment of 10 uranium oxide (U_3O_8) card sources. The measurements and analysis were completed as a verification of the card sources in support of Oak Ridge National Laboratory's nuclear material control and accountability program. Although these cards are not nationally accredited as a nuclear standard, they are used as a working reference for measurements, such as holdup. The uranium card source measurements were taken with a broad energy germanium detector and the Genie 2000 Gamma Acquisition and Analysis software. A complete characterization of each of the 10 uranium card sources was performed using the 4 characteristic full energy peaks of ^{235}U. Using the In Situ Object Counting System software to determine the mathematical efficiency of the measurement, the mass of ^{235}U in each card was determined. The ^{235}U mass in each card ranged from 10.42 to 12.63 g with a systematic error between 0.76 and 0.95 g and a random error of 0.01 g for each card source. The Multi-Group Analysis for Uranium (MGAU) software and the Fixed-Energy, Response Function Analysis with Multiple Efficiency (FRAM) isotopic analysis software were used to determine the isotopic composition of the uranium cards. The measured enrichment was compared to the declared enrichment for each card, with uncertainties ranging from 2.7% to 4.3% for the MGAU analysis and 2.3% to 4.1% for the FRAM analysis. This is a good example of how a well-benchmarked mathematical calibration method can be useful in characterizing uranium sources.

INTRODUCTION

Measurements were performed to determine the mass and enrichment of 10 uranium oxide (U_3O_8) filter paper card sources. The measurements and analysis were completed as a verification of the card sources in support of Oak Ridge National Laboratory's (ORNL's) nuclear material control and accountability program. Although these cards are not nationally accredited as a nuclear standard, they are used as a working reference for measurements, such as holdup.

The In-Situ Object Counting System (ISOCS) software version 4.4.1 was used to calculate the efficiency calibration for each card source, which was then used, along with the measurement data and nuclear data, to calculate mass values for each source. ISOCS is used for gamma sample assay and does not require traditional calibration sources to be used for accurate efficiency calibrations. It uses the Monte Carlo N-Particle Transport (MCNP) modeling code, mathematical geometry

templates, and a few physical sample parameters input by the user. The efficiency calibration output is specific to the detector, sample, and geometry used. ISOCS is used for the quantification of nuclear and radiological materials.

Two analysis software applications were used to calculate the isotopic composition for each card source: Multi-Group Analysis for Uranium (MGAU) software version 4.2 and Fixed-Energy, Response Function Analysis with Multiple Efficiency (FRAM) isotopic analysis software version 5.2. The MGAU software was created to improve the accuracy of uranium measurements and to simplify the setup and calibration steps required for these measurements. This software uses information from the low energy region of the spectrum, which includes gamma and x-rays from 84 to 205 keV, but primarily looks at the ^{235}U and ^{238}U emissions in the 90–94 keV energy range. Several peaks in the measurement spectrum are used to develop a relative efficiency curve, and no efficiency calibration is required before taking the measurements. The FRAM isotopic analysis is typically used to determine the isotopic composition of plutonium, but it can also be used to quantify the uranium isotopic distribution with no calibration necessary. The FRAM software was developed by Los Alamos National Laboratory to analyze pulse-height spectra from measurements taken by high-resolution gamma-ray detectors. It analyzes photo peaks in the spectrum and uses this information to produce isotopic ratios; FRAM can measure uranium that has an enrichment ranging from 0.2 to >97% ^{235}U.

VERIFICATION MEASUREMENTS

Source Information

The card sources are made of uranium oxide (U_3O_8) spread on heavy paper, sealed in place with glue. The paper is folded over on itself and heat sealed in heavy plastic. The U_3O_8 was not uniformly spaced on the card sources. Each card measures 51.5 × 27.5 × 0.378 cm^3 with a 0.13 mm thick layer of plastic surrounding it. There are a total of 10 cards, labeled A–J. **Table 1** shows the declared enrichment and ^{235}U mass, as well as the measured weight of the card using a scale and the calculated density of each card, which will be used later in ISOCS.

Table 1. Information for each card source

Card	Declared ^{235}U Mass (g)	Declared Enrichment	Measured Weight (g)	Calculated Density (g/cm^3)
A	12.960	93.1704	363.8	0.679
B	11.193	93.1663	345.5	0.645
C	11.106	93.1711	345.5	0.645
D	11.100	93.1677	353.3	0.660
E	11.113	93.1673	383.3	0.716
F	11.162	93.1720	356.0	0.665
G	11.108	93.1723	363.9	0.680
H	11.148	93.1718	358.8	0.670
I	11.108	93.1723	365.0	0.682
J	11.160	93.1708	368.2	0.688

Measurement Procedure

The detector used for this procedure was a Broad Energy Germanium Detector (BEGe), along with an Inspector 2000 Digital Signal Processing Spectroscopy Workstation, both produced by Canberra Industries, now Mirion Technologies. The BEGe detector was placed on a flat surface facing the center of the object being measured. It was placed 65.5 cm away from the card source, which was far enough away to measure the entire object. Images of the setup can be seen in **Figure 1** and **Figure 2**.

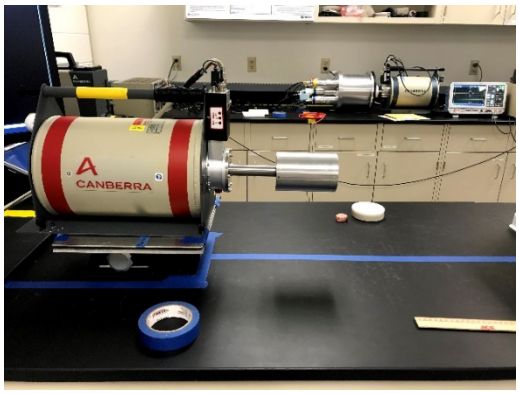

Figure 1. Side view of detector in the measurement setup. Tape was placed around the base of the detector so that it could be placed in the identical position if it were moved.

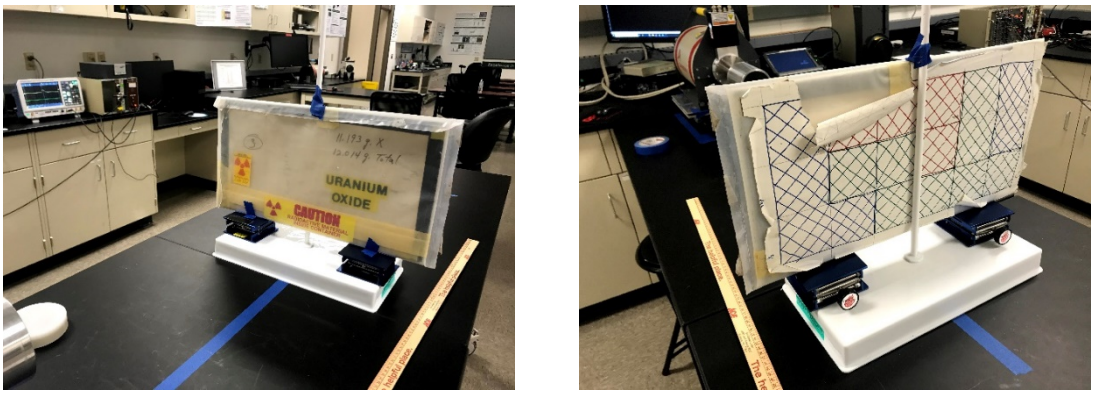

Figure 2. Front view (*left*) and back view (*right*) of card sources in the measurement setup. The card sources were placed on stands and taped so that they were centered with the detector face and placed perpendicular to the detector.

The Genie 2000 Gamma Acquisition and Analysis software was used to set the measurement parameters, defined in **Table 2**, and to start and stop the measurements. The system parameters, high voltage, and energy calibration were all verified. The live time was set to 3,600 s (1 h) for each card source measurement. Two background measurements were collected overnight for 12 h each.

Regions of interest (ROIs) were created on the measurement spectrum by selecting the entirety of a characteristic full energy peak. The ROIs were saved and loaded to each of the measurement spectra including the background spectrums. Four ROIs were created, one for each of the four characteristic

peaks of ^{235}U, which are centered at 143.760, 163.356, 185.715, and 205.316 keV. The net counts and error for each of the peaks were recorded for each spectrum and later corrected for background.

Table 2. Parameters used on the Genie 2000 Gamma Acquisition and Analysis software

Parameters	Value
HVPS	3,500 V
Course Gain	×5
Fine Gain	1.3669×
S-Fine Gain	0.999998×
Rise Time	5.6
Flat Top	0.8
PUR Guard	1.10×

Once the measurements were complete, ISOCS was used to calculate the efficiency calibration. The appropriate geometry template was selected, and the measurement setup was modeled for each of the card sources. The dimensions and material matched what was used for the measurements. When the models were complete, the geometry validity test was used to verify that the dimensions were input correctly. Once the model was validated and saved, the calibration data points were generated for the efficiency curve. The efficiency information was saved in an *.ECC file. This was done for each card source. The efficiency information and nuclear data were used to calculate the ^{235}U mass of each card, and error propagation was used to calculate the error.

The measured enrichment of the card sources was calculated using two different software applications. First, the Genie 2000 MGAU Graphical software was used. The relevant spectrum and appropriate parameters were input. The spectrum was analyzed, and the software output the measured enrichment. This was done for each card source. Second, the Genie 2000 FRAM Isotopic Analysis software was used to calculate the enrichment. Each measurement spectrum was loaded into the software, and the appropriate parameters were selected. The spectrum was analyzed, and the software output the measured enrichment. This was also done for each card source.

EFFICIENCY CALIBRATION USING ISOCS

To determine the efficiency calibration, the measurement geometry must be modeled in ISOCS. Ten ISOCS models were created, one for each card source. The simple box template was used to model the geometry, and the source-to-detector distance was set to 65.5 cm. The dimensions of the box and layers used in ISOCS are shown in **Table 3**. An example of the geometry for the card sources is shown in **Figure 3**.

Once the models were completed and validated, the efficiency curve and calibration data points were calculated for each card source model.

Table 3. Box dimensions in card source ISOCS models

Name	Value
Wall thickness	0.013 cm
Inside width	51.5 cm
Inside height	27.5 cm
Inside depth	0.378 cm
Material	Low density polyethylene
Material density	0.92 g/cm^3
Layer height	27.5 cm
Material	Cellulose ($C_6H_{10}O_5$)

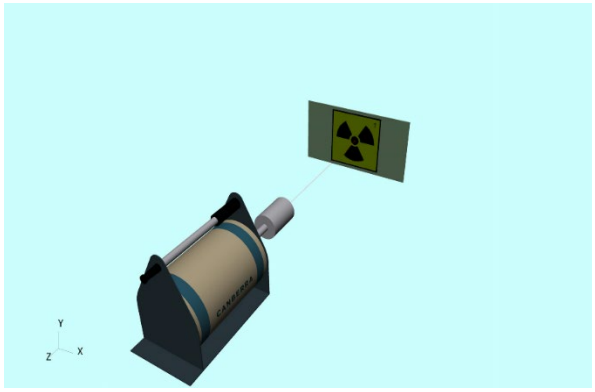

Figure 3. ISOCS geometry model of card source and detector.

MASS ANALYSIS

The ^{235}U mass was calculated with ISOCS four times for each card using information from each of the four main characteristic peaks of ^{235}U. The energies of these four peaks are 143.760, 163.356, 185.715, and 205.316 keV, with the 185.715 peak being the largest. The weighted average of the four mass calculations was used as the measured mass for each card source. The calculations that follow were completed using information about each of the four main characteristic peaks for each card source.

Mass Calculation

To determine the mass of ^{235}U present in each card, the activity first needs to be calculated using the using Equation (1).

$$A = \frac{C_{net}}{T_{live} \cdot \varepsilon \cdot Y} \quad (1)$$

In this equation, A is the activity in Becquerels, C_{net} is the net counts corrected for background, T_{live} is the live time in seconds, ε is the efficiency, and Y is the gamma yield. The net counts are the number of counts, or decays, measured in a characteristic peak of ^{235}U. This can be found using the ROIs in the Genie 2000 Gamma Acquisition and Analysis software. The ROIs were also used to find the number of background counts in a characteristic peak. The average number of background

counts in the characteristic peak per hour, or 3,600 s, was subtracted from the number of counts in the same peak that was also measured for 1 h. This corrected net count value was used for the mass calculations. The live time is the amount of time that the sample was being measured, in this case 3,600 s. The efficiency is a characteristic of the detector that is found using ISOCS and is defined as the ratio of the total number of gammas of a given energy emitted in the source volume over the total amount of counts in the associated peak. The gamma yield, or branching ratio, is the probability that when a given isotope decays it emits a gamma with a given energy. The gamma yield is available from the National Nuclear Data Center. Both the efficiency from ISOCS and the gamma yield have a known error associated with them and are different for each characteristic peak energy.

Once the activity is known, the mass in grams can be calculated using Equation (2), where M is the mass and SA is the specific activity in units of Becquerels per gram.

$$M = \frac{A}{SA} \tag{2}$$

The specific activity is a property specific to an isotope and has units of Becquerels/gram. Some values for the above-mentioned variables are the same across each calculation, and these are shown in **Table 4**. The net counts and efficiency are different for each card source and peak.

Table 4. Values used for mass analysis

Name	Values
Live time	3,600 s
Gamma yield for 143.760 keV peak	10.96% ± 6%
Gamma yield for 163.356 keV peak	5.08% ± 6%
Gamma yield for 185.715 keV peak	57% ± 6%
Gamma yield for 205.316 keV peak	5.02% ± 6%
Specific activity of ^{235}U	79,920 Bq/g

This information was used to find a mass value using each characteristic peak for each card source. Once this part of the calculation was done, there were four mass values for each card source, each one calculated using a different characteristic peak energy.

Mass Error Calculation

For all error calculations, the relative errors were used. Both the systematic and random errors were calculated for each mass value. The random error, or statistical mass error, was found using only the statistical uncertainty in the counts. The error associated with the corrected net counts (after background subtraction) was multiplied by the associated mass value to find the random error. The corrected net count error, σ_{net}, was found with Equation (3), where C_0 is the net counts before background subtraction, σ_0 is the net count error associated with C_0, σ_{bkg} is the background count error, and C_{net} is the net counts after background subtraction.

$$\sigma_{net} = \frac{\sqrt{(C_0 * \sigma_0)^2 + \sigma_{bkg}{}^2}}{C_{net}} \quad (3)$$

Using this, the random error, σ_r, was calculated with Equation (4).

$$\sigma_r = \sigma_{net} * M \quad (4)$$

The systematic mass was calculated using error propagation. The relevant error values used in the calculation were the efficiency error and the gamma yield error. If an error was known for time and specific activity, these would be used as well. The efficiency error is given by ISOCS, and the gamma yield error was 0.06. Equation (5) was used to determine systematic mass error, σ_s. In this equation, σ_ε represents the efficiency error from ISOCS and σ_Y represents the gamma yield error.

$$\sigma_s = M * \sqrt{\sigma_\varepsilon{}^2 + \sigma_Y{}^2} \quad (5)$$

This calculation was performed for each mass value. Once these error values were calculated, there were four random errors and four systematic errors associated with each card source, one of each for the four characteristic peak energies.

Weighted Average Mass Values

Once the mass and error values were calculated for each characteristic peak energy for each card, a weighted average mass value was calculated, as well as an associated systematic and random error. This was done for each card source, so each source would have one average mass value. Equation (6) was used to calculate the weighted mass average, M_{avg}.

$$M_{avg} = \frac{\sum_{i=1}^{4} \frac{M_i}{\sigma_{r_i}{}^2 + \sigma_{s_i}{}^2}}{\sum_{i=1}^{4} \frac{1}{\sigma_{r_i}{}^2 + \sigma_{s_i}{}^2}} \quad (6)$$

In this calculation, the *i* represents the values from each of the four characteristic peaks. In a similar way, the weighted average random and systematic errors were calculated. The random errors were used to calculate the weighted average random error for each card. The systematic errors were used to calculate the weighted average systematic error for each card. These weighted average values were used as the final mass and error values for each card. Equation (7) was used to calculate the weighted error values, σ_{avg}.

$$\left(\sigma_{avg} = \sqrt{\frac{1}{\sum_{i=1}^{4} \frac{1}{\sigma_i{}^2}}} \right) \quad (7)$$

Table 5 compares the declared mass values with these measured mass values of ^{235}U in each card.

Table 5. Measured mass of each card source calculated using ISOCS efficiency calibration

Card	Declared Mass of ^{235}U (g)	Measured Mass of ^{235}U (g)	Systematic Error (g)	Random Error (g)
A	12.96	12.62	0.95	0.01
B	11.19	10.70	0.77	0.01
C	11.11	10.74	0.77	0.01
D	11.10	10.42	0.76	0.01
E	11.11	10.72	0.84	0.01
F	11.16	10.76	0.79	0.01
G	11.11	10.65	0.80	0.01
H	11.15	10.74	0.80	0.01
I	11.11	10.59	0.80	0.01
J	11.16	10.81	0.82	0.01

ENRICHMENT ANALYSIS WITH MGAU

To determine the measured enrichment of each card, each spectrum was analyzed using the MGAU Gamma Acquisition and Analysis software. The declared enrichment was found by taking the ratio of the declared ^{235}U mass and the declared total uranium mass. The declared enrichment and the measured enrichment of each card are shown in **Table 6**. The uncertainty values are calculated with MGAU and represent the absolute uncertainty in the weight percent of the enrichment.

Table 6. Measured enrichment of each card source found using MGAU Gamma Acquisition and Analysis software

Card	Declared Enrichment (weight %)	Measured Enrichment (weight %)	Uncertainty
A	93.1704	95.6475	2.9791
B	93.1663	96.1691	4.0577
C	93.1711	94.0637	2.7403
D	93.1677	96.1599	3.5256
E	93.1673	93.9032	2.8580
F	93.1720	96.1660	3.6603
G	93.1723	96.1518	4.2803
H	93.1718	93.6509	2.7475
I	93.1723	96.1559	3.6081
J	93.1708	96.1566	3.6575

As seen in the table, there is a slight bias when using MGAU to calculate the enrichment of these card sources. This is not surprising because MGAU uses the low energy region of the spectrum. The card sources measured are very highly enriched, and the intensities of some of the lower energy gamma rays, such as the 92.4 and 92.8 keV gamma rays from ^{234}Th, are very low. Because of this, the accuracy and precision will not be as good.

ENRICHMENT ANALYSIS WITH FRAM

Owing to the slight bias of the measured enrichment found using MGAU, we decided to also analyze the enrichment of each card source using the FRAM isotopic analysis software. Unlike MGAU, FRAM uses higher energy gammas in its enrichment calculation.

The *UHEU_Cx_120-1010* parameter set was used to find the enrichment of the card sources. This parameter set is used for highly enriched uranium, which is greater than 20% enriched and for measurements that use a coaxial detector. The energy range used for the isotopic analysis is 120 to 1,010 keV. The energy calibration offset was set to 0.1461 keV, and the energy calibration slope was set to 0.07495 keV/Ch. These numbers were found using the Genie 2000 Gamma Acquisition and Analysis software by going to *Calibrate/Energy Show* while a measurement spectrum was loaded. The energy calibration was the same for each card source.

The measured enrichment and uncertainty using FRAM are shown in **Table 7** alongside the declared enrichment. The uncertainty values shown are the absolute standard deviation, or absolute error, arising from counting statistics of the measured enrichment value calculated by FRAM. As Table 8 indicates, there is still a slight bias for the measured enrichment of the card sources.

Table 7. Measured enrichment of each card source found using FRAM Isotopic Analysis

Card	Declared Enrichment (wt%)	Measured Enrichment (wt%)	Uncertainty
A	93.1704	93.2326	4.1031
B	93.1663	95.1424	2.8426
C	93.1711	94.9838	2.3816
D	93.1677	96.3060	2.6872
E	93.1673	94.7992	3.3439
F	93.1720	95.7545	3.1126
G	93.1723	94.6715	2.9398
H	93.1718	95.8944	2.3354
I	93.1723	96.8444	2.8786
J	93.1708	94.6449	3.5104

CONCLUSIONS

Measurements were performed to determine the mass and enrichment of 10 U_3O_8 filter paper card sources. The measurements and analysis were completed as a verification of the card sources in support of ORNL's nuclear material control and accountability program.

The uranium card source measurements were taken using a BEGe detector and the Genie 2000 Gamma Acquisition and Analysis software. A complete characterization of each of the 10 uranium card sources was performed using the four characteristic full energy peaks of ^{235}U. The ISOCS software was used to model the geometry and generate data points for an efficiency curve. Using this efficiency information, measurement data, and nuclear data, the mass of ^{235}U in each card was

calculated. The mass was calculated four times for each card source, once for each of the four characteristic full energy peaks, and then the average mass was compared to the declared mass. The ^{235}U mass in each card ranged from 10.42 to 12.63 g, with a systematic error between 0.76 and 0.95 g, and a random error of 0.01 g for each card source.

The MGAU software and the FRAM isotopic analysis software were both used to determine the isotopic composition of the uranium card sources. The measured enrichment was compared to the declared enrichment for each card. For the MGAU analysis, the measured enrichment ranged from 93.65 to 96.17 wt% with uncertainties between 2.7% and 4.3%. For the FRAM analysis, the measured enrichment ranged from 93.23 to 96.84 wt% with uncertainties between 2.3% and 4.1%. The MGAU and FRAM enrichment analyses both produced similar results. This is a good example of how a well-benchmarked mathematical calibration method can be useful in characterizing uranium sources.

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