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Scoping Study: Highly Enriched Uranium Isotopic Assessments Using Gamma Spectroscopy Relative Efficiency Analysis When No Uranium-232 Is Present

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ABSTRACT

This report documents results for a scoping study evaluating the viability of using the higher energy, low yield U-235 gamma emission at 345.9 keV to link low energy U-235 emissions to high energy U-238 emissions (rather than U-232 decay products gamma emissions) for highly enriched uranium isotopic gamma spectroscopy relative efficiency assessments when no or little U-232 is present and the U-235 signal is strong.

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ACRONYMS AND TERMS

Acronym/Term	Definition
FRAM	Fixed-Energy Response-Function Analysis with Multiple Efficiency
GADRAS	Gamma Detector Response and Analysis Software
HPGe	High-Purity Germanium
LANL	Los Alamos National Laboratory
LLNL	Lawrence Livermore National Laboratory
MGA	Multi-Group Analysis
NNDC	National Nuclear Data Center
U	Uranium metal
UF_6	Uranium hexafluoride
U_3O_8	Triuranium octoxide

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1. OVERVIEW

This report documents results for a scoping study evaluating the viability of using the higher energy, low yield U-235 gamma emission at 345.9 keV to link low energy U-235 emissions to high energy U-238 emissions (rather than U-232 decay products gamma emissions) for highly enriched uranium isotopic gamma spectroscopy relative efficiency assessments when no or little U-232 is present and the U-235 signal is strong.

1.1. Gamma spectroscopy relative efficiency analysis overview

Relative efficiency curves are used to determine radionuclide activity ratios or radionuclide mass ratios (not absolute activity or mass) from gamma spectroscopy measurements. The benefit of relative efficiency curves is that they require no measurement of calibration sources and "self-correct" for geometry and attenuation (shield attenuation and self-attenuation).

Relative efficiency curves are the fundamental basis of uranium and plutonium isotopic software programs such as Fixed-Energy Response-Function Analysis with Multiple Efficiency (FRAM) developed by Los Alamos National Laboratory (LANL) and Multi-Group Analysis (MGA) developed by Lawrence Livermore National Laboratory (LLNL).

To generate an effective relative efficiency curve, one must have a radionuclide with full-energy peaks that span the energy range of interest or multiple radionuclides with full-energy peaks with sufficient overlapping or nearly overlapping energy ranges for the energy range of interest (Figure 1). In addition, the isotopic composition throughout the sample must be the same (isotopic homogeneity) for a relative efficiency curve to be valid.

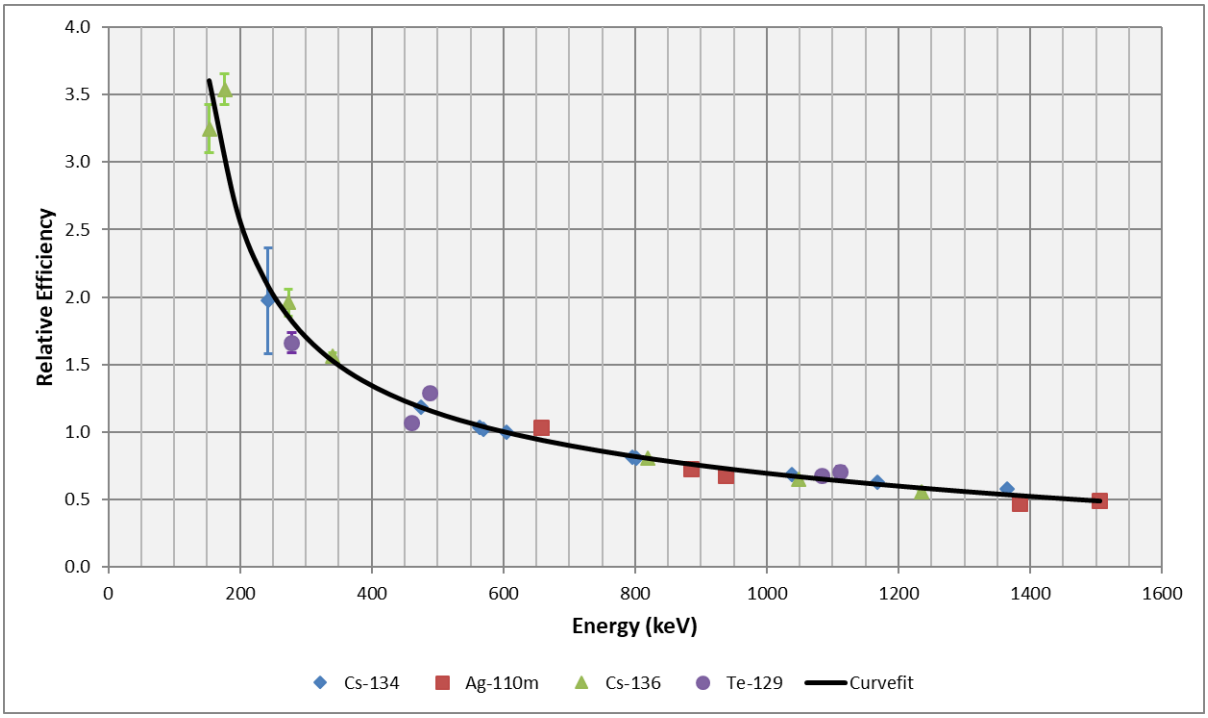

Figure 1. Example relative efficiency curve fit using multiple radionuclides with full-energy peaks with sufficient overlapping.

The shape of the relative efficiency curve is based on the detector efficiency, item geometry, shield attenuation, and self-attenuation. Accordingly, changes in detector efficiency, item geometry, shield attenuation, and self-attenuation are reflected by changes in the shape of the relative efficiency curve (Figure 2).

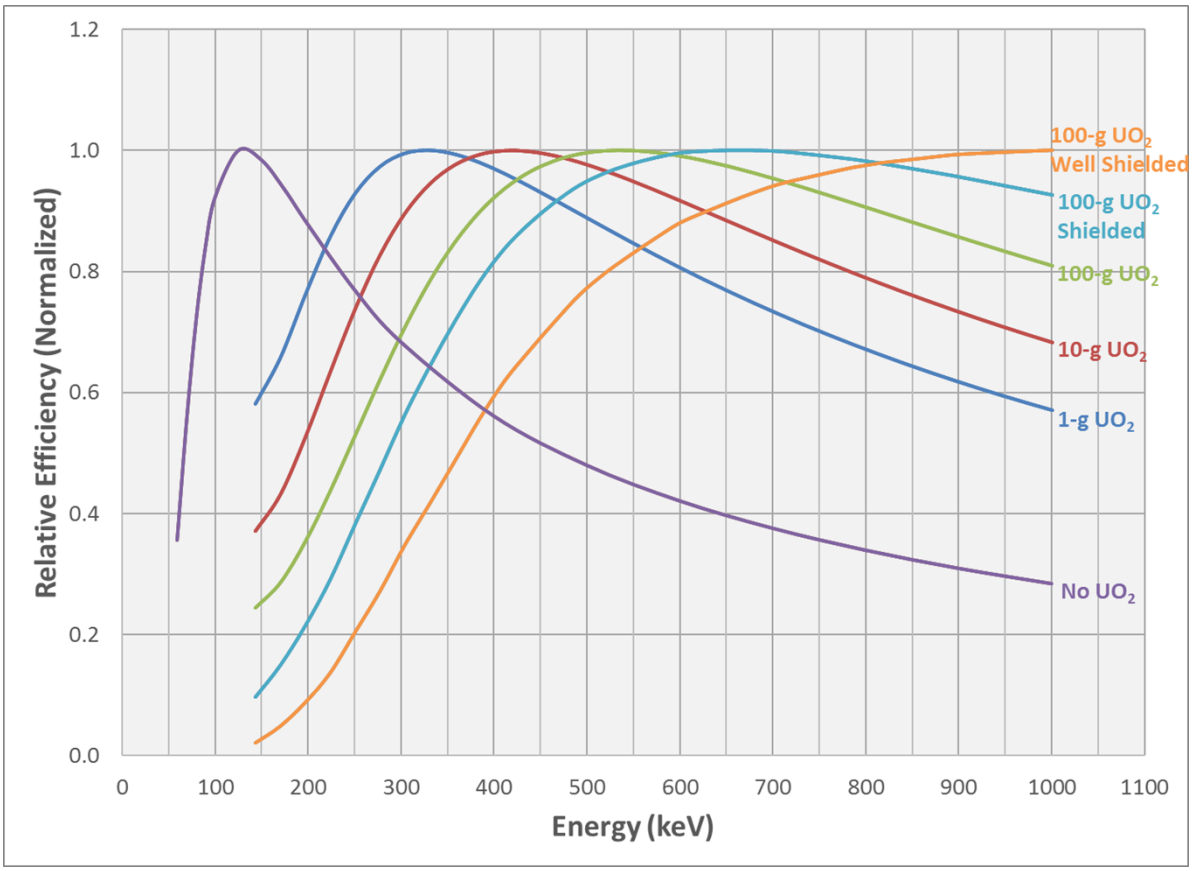

Figure 2. Example relative efficiency curve shapes based on changes in shield attenuation and self-attenuation.

Lastly, relative efficiency curves do not correct for coincidence summing effects. Accordingly, coincidence summing effects should be minimal (or corrected) for a relative efficiency curve to be valid.

For this scoping study, random coincidence summing and/or true coincidence summing of the U-235 202.1 keV and 143.8 keV gamma emissions can affect the determination of the "correct" 345.9 keV full-energy peak area and was avoided to evaluate the method.

1.1.1. Highly enriched uranium isotopic gamma spectroscopy relative efficiency assessments using U-232 decay product emissions

U-232 is produced during reactor irradiation and is present in uranium that has been reprocessed. For enriched and highly enriched uranium isotopic gamma spectroscopy relative efficiency assessments, U-232 decay product emissions are generally used to link the low energy U-235 emissions to the high energy U-238 emissions. As shown in Figure 3, the primary U-232 decay product emissions (238.6, 583.2, 727.3, and 860.6 keV) are used to bridge the gap in the relative efficiency curve between the primary U-235 low energy emissions (143.8, 163.3, 185.7, and 205.3

keV) and the primary U-238 high energy emissions (742.8, 766.4, and 1001.0 keV). In particular, the 238.6 keV U-232 decay chain full-energy peak is very important when generating relative efficiency curves to properly link to the low energy U-235 emissions to the high energy U-238 emission for highly enriched uranium samples. However, when no U-232 is present (e.g., uranium that has not been reprocessed) or little U-232 is present (e.g., uranium was reprocessed recently with little U-232 decay product in-growth), the ability to generate an accurate relative efficiency curve suffers dramatically.

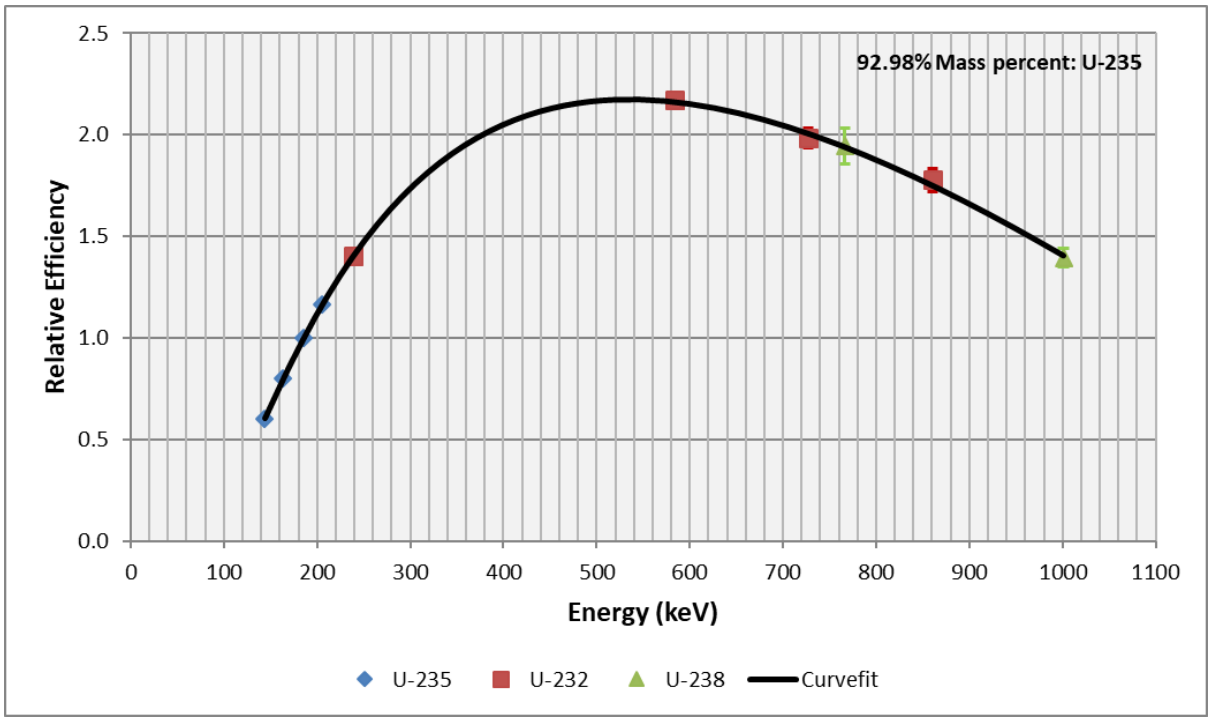

Figure 3. Example of relative efficiency curve-fit for 93.17% mass percent U-235 showing U-232 decay product emissions being used to link the low energy U-235 emissions to the high energy U-238 emissions. Analysis performed using SNL_Relative_Eff_Uiso.xlsb.

For additional detailed information related to gamma spectroscopy relative efficiency analysis, see References [1], [2], [3] and/or [4].

1.2. High-purity germanium (HPGe) detector gamma spectra data set

For the scoping study assessment, an attempt was made to identify candidate existing high-purity germanium (HPGe) detector data sets with gamma spectra from materials with well-documented isotopic distributions that could be used for this scoping study (e.g. FRAM archival test spectra, ORNL/TM-2015/370 (2015) data sets [5], International Database of Reference Gamma-Ray Spectra of Various Nuclear Materials [6]). However, no sufficient HPGe gamma spectra data set could be identified. Therefore, 61 synthetic gamma spectra (60 highly enriched uranium spectra and 1 background spectrum) were generated for evaluation using Gamma Detector Response and Analysis Software (GADRAS) 19.3.5.

An overview of the method and assumptions used to generate the GADRAS synthetic gamma spectra is provided below.

- Spherical 1-dimensional (1D) models with 100-grams of uranium metal (U) with a density of 18.95 g/cc (100% by weight U), 100-grams of triuranium octoxide (U_3O_8) with a density of 3.00 g/cc (84.8% by weight U, 15.2% by weight O), and 100-grams of uranium hexafluoride (UF_6) with a density of 4.00 g/cc (67.62% by weight U, 32.38% by weight F).
- The uranium was modeled with no shielding (bare) and with 4-mm, 8-mm, and 12-mm of stainless steel spherical shells for shielding (see Table 1).

Table 1. Modeled uranium materials/compounds, densities, and shielding.

U Type (Unitless)	Mass (g)	Density (g/cc)	Radius (cm) Bare	Radius (cm) 4-mm SS	Radius (cm) 8-mm SS	Radius (cm) 12-mm SS
U	100	18.95	1.080	1.480	1.880	2.280
UF6	100	4.00	1.814	2.214	2.614	3.014
U3O8	100	3.00	1.996	2.396	2.796	3.196

- The uranium was modeled at enrichments of 50, 60, 70, 80, 90% U-235 by weight as shown in Table 2. Adapted from Reference [7].

Table 2. Modeled uranium enrichments.

	50.00% HEU	60.00% HEU	70.00% HEU	80.00% HEU	90.00% HEU
U-234 Weight %	0.482	0.574	0.664	0.753	0.841
U-235 Weight %	50.000	60.000	70.000	80.000	90.000
U-238 Weight %	49.518	39.426	29.336	19.247	9.159

- The uranium was modeled assuming equilibrium.
  - U-238 in equilibrium with its decay products (Th-234, Pa-234m, and Pa-234).
  - U-235 in equilibrium with its decay product (Th-231).

NOTE: If equilibrium is assumed yet U-238/Pa-234m equilibrium has not been reached, the assessed uranium enrichment will be biased high.

- All spectra were modeled for a standard ORTEC Detective-X HPGe detector using 16384 channels with an upper range of 3 MeV (0.1831 keV/channel).
  - The ORTEC Detective-X contains a 65-mm diameter x 50-mm deep (±10%), p-type HPGe, coaxial construction detector.
  - The standard ORTEC Detective-X HPGe detector using 16384 channels with an upper range of 3 MeV was used since it has a lower keV/channel (finer channel bins) which is better suited for gamma spectroscopy relative efficiency assessments than the 8 MeV range FRAM version 5.2 using 16384 channels (0.4888 keV/channel).
- All spectra were modeled at a distance of 25-cm from the detector face to the center of the 1D model.
- All spectra were modeled at a height of 100-cm from the center of the 1D model to the floor.
- All spectra were modeled using a live time of 6000-seconds.

- The background spectrum was modeled using cosmic and terrestrial levels for uranium, thorium, and potassium consistent with Albuquerque, NM.
- All spectra were modeled with Poisson statistics applied.

The synthetic gamma spectra data set generated using GADRAS creates reasonable variations in shield and self-attenuation over different U-235 enrichments for the scoping study evaluation. In addition, the 1D model dimensions and distances used avoid random coincidence summing and true coincidence summing of the U-235 202.1 keV and 143.8 keV gamma emissions allowing for straightforward analysis. The absence of random coincidence summing in the synthetic gamma spectra data set was verified by ensuring the lack of a U-235 185.7 keV summation peak at 371.4 keV or ensuring the summation peak at 371.4 keV peak area was trivial compared to the 185.7 keV full-energy peak area.

Summary information for background and the uranium metal (U), triuranium octoxide (U_3O_8), and uranium hexafluoride (UF_6) synthetic gamma spectra data sets are summarized in Table 3, Table 4, and Table 5, respectively.

Table 3. Summary information for U metal synthetic spectra generated.

Record #	Title	Live Time (sec)	Real Time (sec)	Gamma (cps)	Dead Time (%)
1	Background	6000.0	6042.6	202	0.7%
2	U 100-g (50% U-235 by weight) @ 25cm H=100cm	6000.0	6187.9	881	3.0%
3	U 100-g (50% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6122.1	576	2.0%
4	U 100-g (50% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6094.4	446	1.5%
5	U 100-g (50% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6078.2	370	1.3%
6	U 100-g (60% U-235 by weight) @ 25cm H=100cm	6000.0	6211.4	990	3.4%
7	U 100-g (60% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6133.6	629	2.2%
8	U 100-g (60% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6100.9	476	1.7%
9	U 100-g (60% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6081.9	387	1.3%
10	U 100-g (70% U-235 by weight) @ 25cm H=100cm	6000.0	6235.2	1098	3.8%
11	U 100-g (70% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6145.1	682	2.4%
12	U 100-g (70% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6107.5	507	1.8%
13	U 100-g (70% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6085.7	405	1.4%
14	U 100-g (80% U-235 by weight) @ 25cm H=100cm	6000.0	6259.0	1207	4.1%
15	U 100-g (80% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6156.7	736	2.5%
16	U 100-g (80% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6114.0	538	1.9%
17	U 100-g (80% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6089.4	423	1.5%
18	U 100-g (90% U-235 by weight) @ 25cm H=100cm	6000.0	6283.1	1317	4.5%
19	U 100-g (90% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6168.4	791	2.7%
20	U 100-g (90% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6120.7	569	2.0%
21	U 100-g (90% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6093.3	441	1.5%

Table 4. Summary information for U_3O_8 synthetic spectra generated.

Record #	Title	Live Time (sec)	Real Time (sec)	Gamma (cps)	Dead Time (%)
22	U3O8 100-g (50% U-235 by weight) @ 25cm H=100cm	6000.0	6509.6	2329	7.8%
23	U3O8 100-g (50% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6282.8	1316	4.5%
24	U3O8 100-g (50% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6189.9	890	3.1%
25	U3O8 100-g (50% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6138.1	650	2.3%
26	U3O8 100-g (60% U-235 by weight) @ 25cm H=100cm	6000.0	6596.2	2707	9.0%
27	U3O8 100-g (60% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6323.9	1503	5.1%
28	U3O8 100-g (60% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6213.3	999	3.4%
29	U3O8 100-g (60% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6151.9	714	2.5%
30	U3O8 100-g (70% U-235 by weight) @ 25cm H=100cm	6000.0	6683.2	3081	10.2%
31	U3O8 100-g (70% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6365.2	1688	5.7%
32	U3O8 100-g (70% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6236.7	1106	3.8%
33	U3O8 100-g (70% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6165.7	778	2.7%
34	U3O8 100-g (80% U-235 by weight) @ 25cm H=100cm	6000.0	6772.0	3459	11.4%
35	U3O8 100-g (80% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6407.0	1875	6.4%
36	U3O8 100-g (80% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6260.4	1214	4.2%
37	U3O8 100-g (80% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6179.6	842	2.9%
38	U3O8 100-g (90% U-235 by weight) @ 25cm H=100cm	6000.0	6861.1	3834	12.6%
39	U3O8 100-g (90% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6448.9	2062	7.0%
40	U3O8 100-g (90% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6284.1	1323	4.5%
41	U3O8 100-g (90% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6193.4	907	3.1%

Table 5. Summary information for UF_6 synthetic spectra generated.

Record #	Title	Live Time (sec)	Real Time (sec)	Gamma (cps)	Dead Time (%)
42	UF6 100-g (50% U-235 by weight) @ 25cm H=100cm	6000.0	6430.3	1979	6.7%
43	UF6 100-g (50% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6244.2	1140	3.9%
44	UF6 100-g (50% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6167.4	787	2.7%
45	UF6 100-g (50% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6124.3	586	2.0%
46	UF6 100-g (60% U-235 by weight) @ 25cm H=100cm	6000.0	6501.1	2292	7.7%
47	UF6 100-g (60% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6278.1	1295	4.4%
48	UF6 100-g (60% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6186.8	876	3.0%
49	UF6 100-g (60% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6135.7	639	2.2%
50	UF6 100-g (70% U-235 by weight) @ 25cm H=100cm	6000.0	6571.8	2600	8.7%
51	UF6 100-g (70% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6312.0	1449	4.9%
52	UF6 100-g (70% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6206.0	964	3.3%
53	UF6 100-g (70% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6147.1	692	2.4%
54	UF6 100-g (80% U-235 by weight) @ 25cm H=100cm	6000.0	6643.9	2912	9.7%
55	UF6 100-g (80% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6346.2	1603	5.5%
56	UF6 100-g (80% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6225.5	1054	3.6%
57	UF6 100-g (80% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6158.5	745	2.6%
58	UF6 100-g (90% U-235 by weight) @ 25cm H=100cm	6000.0	6716.1	3222	10.7%
59	UF6 100-g (90% U-235 by weight) with 04-mm SS @ 25cm H=100cm	6000.0	6380.6	1757	6.0%
60	UF6 100-g (90% U-235 by weight) with 08-mm SS @ 25cm H=100cm	6000.0	6245.1	1145	3.9%
61	UF6 100-g (90% U-235 by weight) with 12-mm SS @ 25cm H=100cm	6000.0	6170.0	798	2.8%

1.3. U-235 345.9 keV gamma emission yield uncertainty

In addition to random coincidence summing and/or true coincidence summing issues discussed previously, another issue associated with using the U-235 345.9 keV gamma emission to link low energy U-235 emissions to high energy U-238 emissions is that published yields for the U-235 345.9

keV gamma emission (see Table 6) are not consistent [8] and would require better yield estimates with reduced uncertainty if the method is found to be viable.

Table 6. Published U-235 345.9 keV gamma emission yields.

Reference	U-235 345.9 keV Gamma Yield
ENDF/B-VIII.0	0.040% ± 0.005%
NNDC NuDat Database	0.034% ± 0.005%
FRAM 5.2	0.030%
GADRAS 19.3.5	0.038%

ENDF/B-VIII.0 Evaluated Nuclear Data Library, https://www.nndc.bnl.gov/endf-b8.0/

Decay Radiation Database at the National Nuclear Data Center (NNDC) - NuDat Database (Decay Radiation database FRAM version 5.2 of 8/8/2023), https://www.nndc.bnl.gov/nudat3/indx_dec.jsp

2. ANALYSIS METHOD, RESULTS, AND RESULTS DISCUSSION

The following sections summarize the analysis method and results.

2.1. Analysis method

The uranium metal (U), triuranium octoxide (U_3O_8), and uranium hexafluoride (UF_6) synthetic gamma spectra data sets generated using GADRAS were analyzed using FRAM version 5.2 with a modified FRAM parameter set. Specifically, the FRAM parameter set (uleu_cx_120-1010), which only uses U-234, U-235, and U-238 gamma emissions between 120 and 1010 keV and no U-232 decay product emissions, was modified to include the higher energy, low yield U-235 gamma emission at 345.9 keV in the relative efficiency curve. For consistency with the GADRAS generated synthetic spectra, a 345.9 keV gamma emission yield of 3.8E-04 gammas/disintegration was assigned in the FRAM parameter set allowing direct evaluation of the method without requiring consideration of yield uncertainty. Lastly, FRAM analysis was performed using the physical model with default parameters for the relative efficiency determination.

2.2. Analysis results

Example FRAM version 5.2 relative efficiency curve-fits, mass percent U-235, and 345.9 keV full-energy peak fits/areas for uranium metal (U), triuranium octoxide (U_3O_8), and uranium hexafluoride (UF_6) are provided in Figure 4, Figure 5, Figure 6, Figure 7, Figure 8, and Figure 9, respectively.

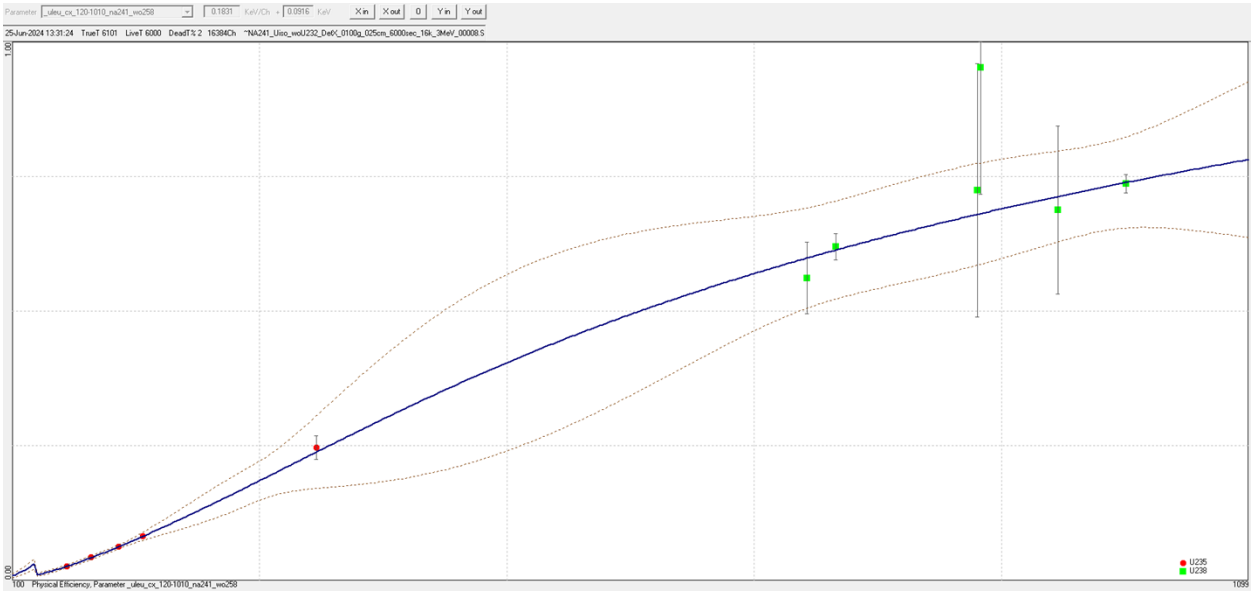

Figure 4. FRAM 5.2 relative efficiency curve-fit for U metal with 60% mass percent U-235 and 8-mm of stainless steel shielding. FRAM 5.2 result = 65.3 ± 2.87% mass percent U-235.

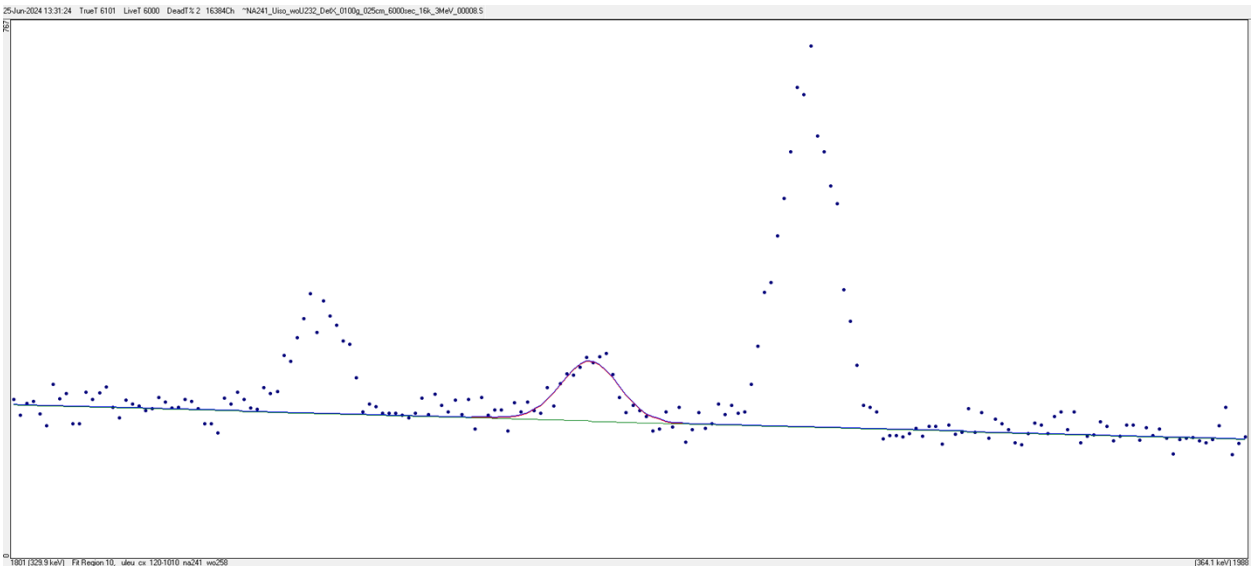

Figure 5. FRAM version 5.2 345.9 keV full-energy peak fit for U metal with 60% mass percent U-235 and 8-mm of stainless steel shielding. FRAM 5.2 result = 945 counts ± 8.81%.

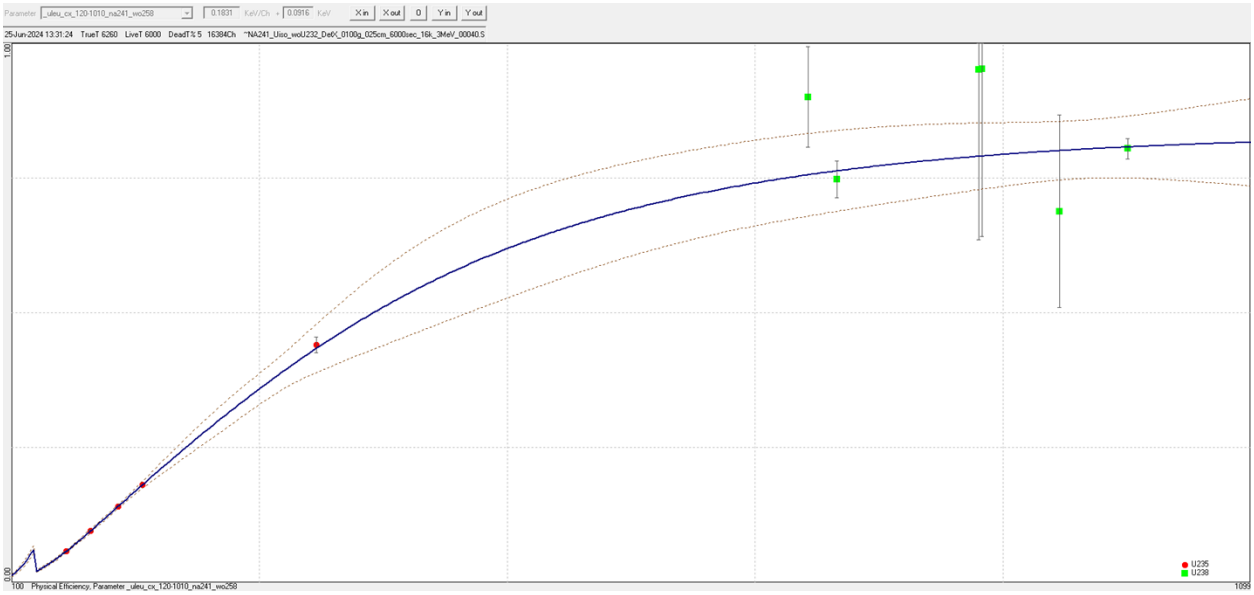

Figure 6. FRAM 5.2 relative efficiency curve-fit for U_3O_8 with 80% mass percent U-235 and 8-mm of stainless steel shielding. FRAM 5.2 result = 82.7 ± 1.11% mass percent U-235.

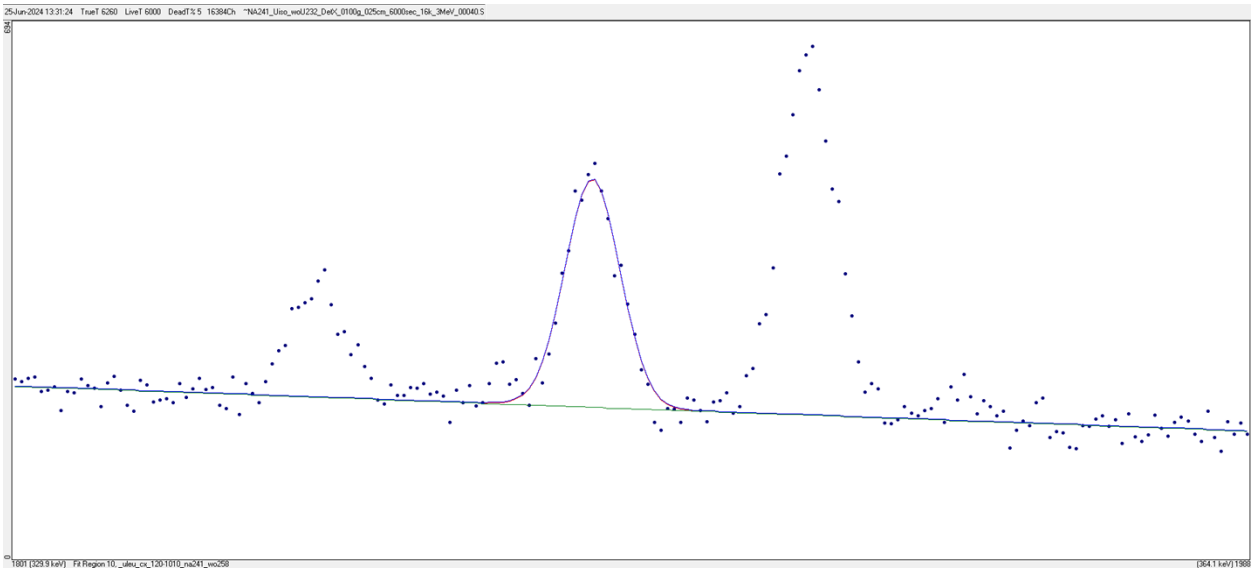

Figure 7. FRAM 5.2 345.9 keV full-energy peak fit for U_3O_8 with 80% mass percent U-235 and 8-mm of stainless steel shielding. FRAM 5.2 result = 3085 counts ± 3.22%.

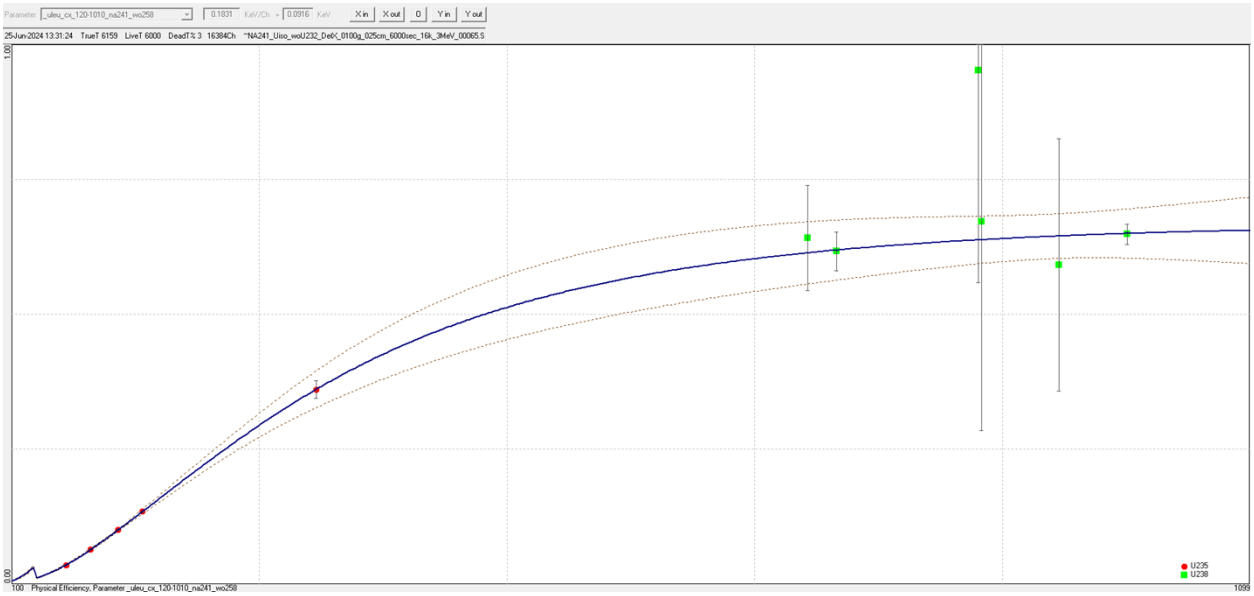

Figure 8. FRAM 5.2 relative efficiency curve-fit for UF_6 with 80% mass percent U-235 and 12-mm of stainless steel shielding. FRAM 5.2 result = 79.2 ± 1.30% mass percent U-235.

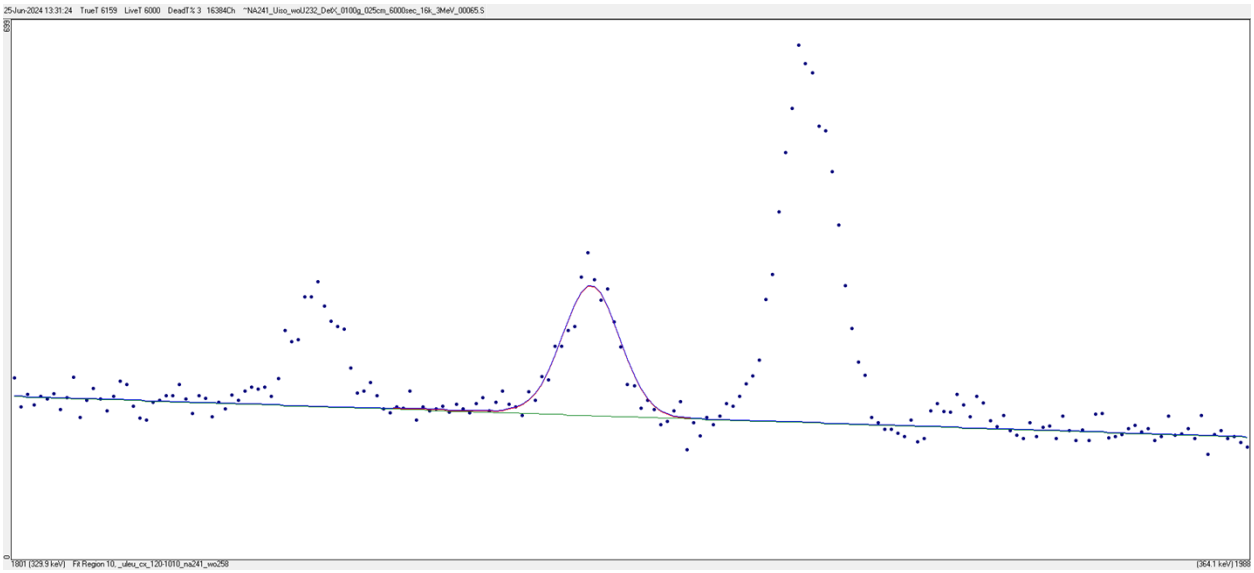

Figure 9. FRAM 5.2 345.9 keV full-energy peak fit UF_6 with 80% mass percent U-235 and 12-mm of stainless steel shielding. FRAM 5.2 result = 1857 counts ± 4.68%.

Summarized analysis mass percent U-235 and U-235 345.9 keV full-energy peak area results using FRAM version 5.2 with the modified FRAM parameter set are provided in Table 7, Table 8, and Table 9 for uranium metal (U), triuranium octoxide (U_3O_8), and uranium hexafluoride (UF_6), respectively.

Table 7. FRAM version 5.2 results with a modified FRAM parameter set: Uranium metal.

				FRAM 5.2	FRAM5.2	FRAM5.2	FRAM5.2
U compound mass (g)	U compound	Shielding	U-235 mass %	U-235 mass % w Uncertainty (w Systematic Errors)	U-235 mass % 90% Confidence Interval (w Systematic Errors)	U-235 345.9 keV Peak area w Uncertainty (counts)	U-235 345.6 keV Peak area (Sigma)
100	U	Bare	50	39.6 ± 3.51	33.8 - 45.4	1456 ± 6.23%	16.1
100	U	w 4-mm SS	50	48.7 ± 2.67	44.3 - 53.1	984 ± 8.87%	11.3
100	U	w 8-mm SS	50	50.7 ± 3.27	45.3 - 56.1	743 ± 11.55%	8.7
100	U	w 12-mm SS	50	46.3 ± 2.08	42.9 - 49.7	475 ± 16.25%	6.2
100	U	Bare	60	61.8 ± 1.51	59.3 - 64.3	1693 ± 5.28%	18.9
100	U	w 4-mm SS	60	63.2 ± 1.58	60.6 - 65.8	1122 ± 7.60%	13.2
100	U	w 8-mm SS	60	65.3 ± 2.87	60.5 - 78.6	945 ± 8.81%	11.4
100	U	w 12-mm SS	60	61.2 ± 4.72	53.4 - 69.4	700 ± 11.35%	8.8
100	U	Bare	70	70.0 ± 2.10	66.5 - 73.5	2100 ± 4.48%	22.3
100	U	w 4-mm SS	70	75.1 ± 1.26	73.0 - 77.2	1299 ± 6.43%	15.6
100	U	w 8-mm SS	70	69.3 ± 3.13	64.1 - 74.5	1020 ± 7.96%	12.6
100	U	w 12-mm SS	70	72.5 ± 3.70	66.4 - 78.6	871 ± 9.12%	11.0
100	U	Bare	80	85.8 ± 1.25	83.7 - 87.9	2342 ± 3.80%	26.3
100	U	w 4-mm SS	80	80.9 ± 1.39	78.6 - 83.2	1592 ± 5.34%	18.7
100	U	w 8-mm SS	80	85.9 ± 1.59	83.3 - 88.5	1105 ± 7.14%	14.0
100	U	w 12-mm SS	80	70.6 ± 3.80	64.4 - 76.8	837 ± 9.29%	10.8
100	U	Bare	90	89.5 ± 1.16	87.6 - 91.4	2647 ± 3.57%	28.0
100	U	w 4-mm SS	90	88.1 ± 1.61	85.4 - 94.8	1734 ± 4.91%	20.4
100	U	w 8-mm SS	90	74.2 ± 3.47	68.5 - 79.9	1118 ± 8.85%	11.3
100	U	w 12-mm SS	90	63.5 ± 4.19	56.6 - 71.4	610 ± 13.37%	7.5

Table 8. FRAM version 5.2 results with a modified FRAM parameter set: U_3O_8.

				FRAM 5.2	FRAM5.2	FRAM5.2	FRAM5.2
U compound mass (g)	U compound	Shielding	U-235 mass %	U-235 mass % w Uncertainty (w Systematic Errors)	U-235 mass % 90% Confidence Interval (w Systematic Errors)	U-235 345.9 keV Peak area w Uncertainty (counts)	U-235 345.6 keV Peak area (Sigma)
100	U3O8	Bare	50	55.2 ± 1.46	52.8 - 57.6	4206 ± 2.71%	36.9
100	U3O8	w 4-mm SS	50	52.6 ± 1.01	50.9 - 54.3	2640 ± 3.92%	25.5
100	U3O8	w 8-mm SS	50	49.5 ± 1.68	46.7 - 52.3	1834 ± 5.33%	18.8
100	U3O8	w 12-mm SS	50	53.5 ± 1.64	50.8 - 56.2	1300 ± 7.13%	14.0
100	U3O8	Bare	60	64.4 ± 1.40	62.1 - 66.7	5155 ± 2.28%	43.9
100	U3O8	w 4-mm SS	60	65.9 ± 1.92	62.7 - 69.1	3414 ± 3.04%	32.9
100	U3O8	w 8-mm SS	60	65.1 ± 1.44	62.7 - 67.5	2458 ± 4.07%	24.6
100	U3O8	w 12-mm SS	60	65.1 ± 2.13	61.6 - 68.6	1651 ± 5.46%	18.3
100	U3O8	Bare	70	75.5 ± 0.88	74.0 - 77.5	5770 ± 1.99%	50.3
100	U3O8	w 4-mm SS	70	76.1 ± 1.65	73.4 - 78.8	4087 ± 2.56%	39.1
100	U3O8	w 8-mm SS	70	73.5 ± 0.93	71.9 - 75.8	2693 ± 3.56%	28.1
100	U3O8	w 12-mm SS	70	76.7 ± 2.29	72.9 - 83.5	2084 ± 4.44%	22.5
100	U3O8	Bare	80	85.7 ± 1.09	83.9 - 87.5	7042 ± 1.65%	60.6
100	U3O8	w 4-mm SS	80	85.3 ± 1.03	83.6 - 87.0	4712 ± 2.49%	40.2
100	U3O8	w 8-mm SS	80	82.7 ± 1.11	80.9 - 84.5	3085 ± 3.22%	31.1
100	U3O8	w 12-mm SS	80	83.2 ± 2.02	79.9 - 86.5	2379 ± 3.73%	26.8
100	U3O8	Bare	90	91.6 ± 0.86	90.1 - 93.8	7764 ± 1.65%	60.6
100	U3O8	w 4-mm SS	90	91.0 ± 0.71	89.8 - 92.2	4949 ± 2.16%	46.3
100	U3O8	w 8-mm SS	90	90.2 ± 1.10	88.4 - 92.0	3578 ± 3.50%	28.6
100	U3O8	w 12-mm SS	90	91.7 ± 0.76	90.4 - 93.4	2397 ± 3.64%	27.5

Table 9. FRAM version 5.2 results with a modified FRAM parameter set: UF_6.

				FRAM 5.2	FRAM5.2	FRAM5.2	FRAM5.2
U compound mass (g)	U compound	Shielding	U-235 mass %	U-235 mass % w Uncertainty (w Systematic Errors)	U-235 mass % 90% Confidence Interval (w Systematic Errors)	U-235 345.9 keV Peak area w Uncertainty (counts)	U-235 345.6 keV Peak area (Sigma)
100	UF6	Bare	50	53.3 ± 1.55	50.7 - 55.9	3596 ± 3.03%	33.0
100	UF6	w 4-mm SS	50	56.8 ± 2.24	53.1 - 61.5	2543 ± 4.11%	24.3
100	UF6	w 8-mm SS	50	55.2 ± 2.44	51.2 - 59.2	1820 ± 5.16%	19.4
100	UF6	w 12-mm SS	50	56.4 ± 2.90	51.6 - 61.2	1356 ± 6.79%	14.7
100	UF6	Bare	60	66.2 ± 1.44	63.8 - 68.6	4385 ± 2.51%	39.8
100	UF6	w 4-mm SS	60	65.9 ± 1.68	63.1 - 68.7	2990 ± 3.37%	29.7
100	UF6	w 8-mm SS	60	61.4 ± 2.16	57.8 - 65.4	2076 ± 4.57%	21.9
100	UF6	w 12-mm SS	60	67.0 ± 2.57	62.8 - 71.2	1594 ± 5.59%	17.9
100	UF6	Bare	70	74.9 ± 1.42	72.6 - 77.2	5137 ± 2.15%	46.5
100	UF6	w 4-mm SS	70	75.3 ± 1.77	72.4 - 78.2	3351 ± 2.97%	33.7
100	UF6	w 8-mm SS	70	74.0 ± 1.00	72.4 - 75.6	2346 ± 3.99%	25.1
100	UF6	w 12-mm SS	70	72.2 ± 2.63	67.9 - 76.5	1925 ± 4.63%	21.6
100	UF6	Bare	80	85.0 ± 1.13	83.1 - 86.9	6013 ± 1.84%	54.3
100	UF6	w 4-mm SS	80	85.0 ± 1.01	83.3 - 86.7	3918 ± 2.79%	35.8
100	UF6	w 8-mm SS	80	83.6 ± 1.56	81.0 - 86.2	2873 ± 3.36%	29.8
100	UF6	w 12-mm SS	80	79.2 ± 1.30	77.1 - 81.3	1857 ± 4.68%	21.4
100	UF6	Bare	90	92.8 ± 0.74	91.5 - 94.9	6690 ± 2.17%	46.1
100	UF6	w 4-mm SS	90	91.7 ± 0.82	90.3 - 93.6	4376 ± 2.28%	43.9
100	UF6	w 8-mm SS	90	90.9 ± 0.96	89.3 - 92.5	2903 ± 3.56%	28.1
100	UF6	w 12-mm SS	90	90.0 ± 1.25	87.9 - 92.1	2214 ± 4.32%	23.1

2.3. Analysis results discussion and conclusions

Using the FRAM 5.2 calculated uncertainties with systematic errors, only 19 of the 60 calculated 90% confidence intervals contained the correct U-235 mass percent (10/20 for U metal, 4/20 for U_3O_8, and 5/20 for UF_6). It should be noted that the uranium metal (U) spectra had the least precise/worst U-235 345.9 keV full-energy peak counting statistics on average (14.6 standard deviations above background) while the uranium hexafluoride (UF_6) spectra had the most precise/best (33.8 compared to 30.5 standard deviations above background for triuranium octoxide (U_3O_8)).

Although the vast majority of calculated 90% confidence intervals did not contain the correct U-235 mass percent, the vast majority of the U-235 mass percent mean estimates were within 10% of the correct U-235 mass percent (53/60 overall: 16/20 for U metal; 20/20 for U_3O_8; and 17/20 for UF_6) with only three U-235 mass percent mean estimates deviating from the correct U-235 mass percent by more than 20% (bare 50% enriched U metal (26.3% deviation), 90% enriched U metal with 8-mm of stainless steel shielding (21.3% deviation), and 90% enriched U metal with 12-mm of stainless steel shielding (41.7% deviation)). This suggests that the method might be viable/reasonable for lower accuracy highly enriched uranium assessments (e.g., initial nuclear emergency response operations) but likely insufficient for international safeguards highly enriched uranium isotopic determinations.

Additional considerations affecting the operationally feasible for international safeguards include high count rates and/or longer count times are required due to the low yield of the U-235 emission at 345.9 keV. Further difficulties/drawbacks for international safeguards include random coincidence summing and/or true coincidence summing of the U-235 202.1 keV and 143.8 keV gamma emissions potentially affecting the determination of the "correct" 345.9 keV full-energy peak area. Lastly, the U-235 gamma emission yield uncertainty previously discussed, but not considered in this evaluation, further impacts the ability to use the 345.9 keV U-235 gamma emission for enriched uranium isotopic determinations when high accuracy is needed.

Areas for future investigation/study to be considered include better "physics measurements" to estimate the "correct" 345.9 keV U-235 gamma emission yield and the impact of reducing 345.9 keV U-235 gamma emission yield uncertainties on uranium gamma spectroscopy assessments. In addition, future assessments using highly enriched uranium gamma spectra could be generated and evaluated using a finer conversion gain of 0.125 keV/channel which is more consistent with traditional FRAM parameter set analysis in the 120 to 1010 keV range.

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