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Cooperation on Improved Isotopic Identification and Analysis Software for Portable, Electrically Cooled High-Resolution Gamma Spectrometry Systems Final Report

J. G. Dreyer, T. F. Wang, D. T. Vo, P. F. Funk, A. . Weber

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Disclaimer

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FINAL REPORT

Action Sheet 4

Cooperation on Improved Isotopic Identification and Analysis Software for Portable, Electrically Cooled, High-Resolution Gamma Spectrometry Systems

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Prepared by
Tzu-Fang Wang, Jonathan Dreyer, LLNL[a]
Duc Vo, LANL[b]
Pierre Funk, Anne-Laure Weber, IRSN

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[b] This work was performed under the auspices of the U.S. Department of Energy by Los Alamos National Laboratory under Contract DE-AC52-06NA25396.

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1. Overview

Under a 2006 agreement between the Department of Energy (DOE) of the United States of America and the Institut de Radioprotection et de Sûreté Nucléaire (IRSN) of France, the National Nuclear Security Administration (NNSA) within DOE and IRSN initiated a collaboration to improve isotopic identification and analysis of nuclear material [i.e., plutonium (Pu) and uranium (U)]. The specific aim of the collaborative project was to develop new versions of two types of isotopic identification and analysis software: (1) the fixed-energy response-function analysis for multiple energies (FRAM) codes and (2) multi-group analysis (MGA) codes. The project is entitled *Action Sheet 4 – Cooperation on Improved Isotopic Identification and Analysis Software for Portable, Electrically Cooled, High-Resolution Gamma Spectrometry Systems* (Action Sheet 4).

FRAM and MGA/U235HI are software codes used to analyze isotopic ratios of U and Pu. FRAM is an application that uses parameter sets for the analysis of U or Pu. MGA and U235HI are two separate applications that analyze Pu or U, respectively. They have traditionally been used by safeguards practitioners to analyze gamma spectra acquired with high-resolution gamma spectrometry (HRGS) systems that are cooled by liquid nitrogen. However, it was discovered that these analysis programs were not as accurate when used on spectra acquired with a newer generation of more portable, electrically cooled HRGS (ECHRGS) systems.

In response to this need, DOE/NNSA and IRSN collaborated to update the FRAM and U235HI codes to improve their performance with newer ECHRGS systems. Lawrence Livermore National Laboratory (LLNL) and Los Alamos National Laboratory (LANL) performed this work for DOE/NNSA. Specifically, LLNL, LANL, and IRSN collaborated on the following tasks:

- **Collect gamma spectra** using ECHRGS systems and then analyze the data using FRAM and MGA/U235HI software codes.
- **Compare the FRAM and MGA/U235HI analysis results** for spectra collected using ECHRGS systems with spectra collected using HRGS systems that are cooled by liquid nitrogen in order to determine areas where the codes failed to analyze or inaccurately estimated the isotopic abundance for data collected using ECHRGS systems.
- **Develop improved versions of FRAM and U235HI** codes that reliably provide a more accurate estimate of the isotopic abundance of U and Pu when using the ECHRGS systems. Under the original Action Sheet, the MGA software was to be adapted for this application; however, the U235HI was found to be more suitable for this application and was modified instead.
- **Test and evaluate** the new versions of FRAM and U235HI.
- **Release improved versions of the codes** to be used by the international safeguards community.

An additional objective under Action Sheet 4 was to examine the rapid HRGS radionuclide identification software (RADID) developed at LLNL and embedded in the Ortec DETECTIVE® ECHRGS system. This project included the implementation of minor modifications to allow background subtraction capabilities and to enable IRSN to explore opportunities to improve the capabilities of RADID to properly identify radionuclides.

This final report discusses improvements made in isotopic analysis tools for use with portable ECHRGS systems. It also includes the results of testing RADID and the recommendation not to use RADID for categorization of U or Pu. Additionally, the report presents a summary of the improvements to FRAM and U235HI for data collected using ECHRGS systems, such as the Ortec DETECTIVE® and Canberra Falcon®. These improved tools will enable the use of ECHRGS systems in a wider range of safeguards scenarios by providing more accurate and effective verification measurements for ECHRGS detection systems.

2. Background

IRSN has extensive experience performing on-site, standard HRGS measurements for nuclear safeguards purposes. This experience consists of identifying, characterizing, and quantifying nuclear materials [i.e., U, Pu, and thorium (Th)] that are present in facilities in various physiochemical forms and packaging. In addition, at IRSN, the laboratory for safeguards nondestructive assay is equipped with two hand-held ECHRGS instruments (Ortec DETECTIVE® and MICRODETECTIVE®) to implement controls in facilities where IRSN performs inspection activities. Furthermore, this equipment is also used during annual inventories in IRSN facilities. These two ECHRGS systems improve the efficiency of first-level safeguards inspections through enhanced performance and applications compared with traditional handheld scintillator instruments and improved ease of use compared with traditional liquid nitrogen–cooled HRGS (LNCHRGS) systems.

However, the width of the individual peaks for a spectrum generated using the ECHRGS is broader than the peak width of standard LNCHRGS detection systems. Thus, biases are expected in ECHRGS-generated results of U and Pu isotopic composition measurements because the basic analysis algorithms implemented in the software analysis codes are tailored to the use of HRGS. An objective of Action Sheet 4 was to revise FRAM and U235HI codes to provide accurate measurements when used with ECHRGS systems, which will allow safeguards inspectors to independently verify a larger number of items within the time allotted in the safeguards approach for a facility.

Another issue of specific interest to IRSN was to better understand the RADID identification software embedded in the DETECTIVE® package and to determine limitations in its ability to properly identify radionuclides according to measurement conditions at a facility and to then improve these capabilities. IRSN has a variety of nuclear material and has developed a custom software application called Reconnaissance for difficult background situations. In addition to the DETECTIVE® package, IRSN currently uses a DETECTIVE® instrument in conjunction with in-house dedicated Reconnaissance nuclear material identification software.

Action Sheet 4 sought to minimize analysis biases from the individual codes and to improve the capability of RADID software to accurately identify radionuclides present in the spectrum. The goals for this work were (1) to provide IRSN with a capability to perform more efficient, effective, and accurate verification measurements and (2) to provide the international safeguards community with documented improvements in FRAM and U235HI when coupled with the use of portable ECHRGS systems.

3. Scope of Work

An overview of the scope of work under Action Sheet 4 is provided in Table 1 below.

Table 1. Overview of scope of work for Action Sheet 4.

Tasks	Description	Report Section
4	Collect spectra using ECHRGS systems.	4
5	Compare spectra collected using ECHRGS systems with spectra collected using LNCHRGS. Conduct isotopic analysis using FRAM and MGA/U235HI software to identify algorithm deficiencies.	5
6	Improve FRAM and MGA/U235HI performance with ECHRGS.	6
7 and 8	Test and evaluate the updated FRAM and MGA/U235HI software. Refine and release the final version of the software.	6
2 and 3	Test the capability of RADID software to accurately identify radionuclides present in the spectrum.	7

Collect and Compare Data. IRSN, LANL, and LLNL collaborated to obtain new sets of U, Pu, and mixed U and Pu (i.e., mixed oxide fuel MOX) reference material spectra using ECHRGS. This new data (collected using ECHRGS) was then compared with data previously collected using LNCHRGS.

Compare Analysis Results. IRSN, LANL, and LLNL benchmark spectra were used to evaluate the performances of FRAM v5.2, U235HI v0.9, and the French-developed IGA v7.0[c] codes. Based on the results of the data comparison, LANL and LLNL worked to improve the deconvolution algorithms to reduce some of the biases related to the poorer energy resolution of the ECHRGS systems (compared to the LNCHRGS systems).

Improve FRAM and MGA/U235HI. The spectra acquired under Task 4 were used by LANL, LLNL, and IRSN to adapt FRAM and U235HI algorithms for improved performance with different ECHRGS systems.

Test and Evaluate Updated FRAM and MGA/U235HI. Following the initial FRAM and MGA software improvements (FRAM v5.2 and U235HI v0.9), these analysis tools were provided to IRSN for testing. The test results identified areas that needed further improvement. Based on this work, additional modifications were made to address software deficiencies (discussed in Section 6 of this report).

[c] IGA is a French acronym standing for actinides gamma isotopy.

In activities covered under *Action Sheet 6 – Cooperation on the International Working Group on Gamma Spectrometry Techniques (IWG-GST)* (Action Sheet 6), DOE/NNSA and IRSN collaborated to build a reference database of spectra that covers the range of measurement conditions encountered on-site. Moreover, the spectral database that was prepared by the Action Sheet 4 team to identify and correct algorithm deficiencies was also used under Action Sheet 6. The spectral database includes the following collection of measurement conditions:

- A broad range of matrix and nuclear material sample densities and sizes
- A variety of sample containers with various thickness and materials of construction
- A variety of measurement geometries and attenuating material thicknesses
- A variety of mixtures of U and Pu and/or other nuclear material with impurities

The joint work performed under Action Sheet 4 provided information regarding the capability of the revised codes to (1) provide accurate results for a variety of sample containers with various material compositions and wall thicknesses, (2) generate accurate correction factors, and (3) accurately determine the measurement error.

Test RADID Software. This report also summarizes the joint evaluation of the "offline" software for rapid identification of gamma-ray emitters and recommended improvements for the identification process.

Table 2. INSEP-IRSN Action Sheet 4 Participants

Name	Organization
Jessica Rahim	DOE/NNSA
Risa Haddal	SNL
Duc T. Vo	LANL
Jonathan Dreyer	LLNL
Tzu-Fang Wang	LLNL
Pierre Funk	IRSN
Nicolas Pépin	IRSN
Anne-Laure Weber	IRSN

4. ECHRGS Data Collection

IRSN, LANL, and LLNL worked to develop a set of spectra using both LNCHRGS and ECHRGS instruments. These spectra were used to determine how the reduced energy resolution of ECHRGS effected isotopic analysis software.

4.1. IRSN ECHRGS Data Collection

The ECHRGS data collection was performed at IRSN using a MicroTransSpec (Ametek/Ortec) equipped with a type P coaxial high-purity germanium (HPGe) detector (50 mm diameter, 35 mm length, 15% relative efficiency, and average resolution about 1.2 keV at 122 keV), connected to a dedicated computer in order to make it compatible with FRAM and IGA for U and Pu isotopic

composition measurements. IGA is an isotopic analysis code developed by the French Alternative Energies and Atomic Energy Commission (CEA). IRSN used this code in its work and compared the results of IGA with the results of the FRAM v5.1, FRAM v5.2, and U235HI v0.9 codes. The instrument operates with a fixed conversion gain of 0.3662 keV/channel up to 3 MeV on 8192 channels.

IRSN performed an experiment in its laboratory using certified U_3O_8 powder sources covering the U enrichment range from depleted U (DU) (0.3%) up to high-enriched U (HEU) (89%). The objective of this measurement campaign was to acquire spectra in order to later evaluate the performances of the FRAM v5.1, FRAM v5.2, and U235HI v0.9 codes (accuracy against the reference 235U enrichment value, comparison with the IGA code, and reported code uncertainty) under varying conditions, such as 1) a range of 235U enrichment (DU to HEU), 2) counting statistics (up to 5.0 X 106 total counts), 3) source/detector distance (a few mm to a few tens of cm to evaluate the impact of coincidence summing), and 4) presence of a stainless steel shielding (2, 5, or 10 mm simulating the container walls that might be encountered on site and might have an impact on the spectrum shape and consequently on the efficiencies of the codes). The ^{235}U enrichment from the sources used in this project is certified with an uncertainty between 0.01 and 0.036% depending on the source (with k = 2). These uncertainties result from mass spectrometry characterization that is based on certified reference materials traceable to an international reference base.

The acquisitions were launched using the IRSN AutoISO_PLUM software by selecting a time interval for spectra recording and FRAM v5.2 (or IGA v7.0 or U235HI v0.9) auto-incremented analysis. AutoISO_PLUM is software used for the acquisition and analysis of γ (U and Pu) spectrometry measurements. It was developed to facilitate measurement discontinuation by observing the evolution of a result (isotopes and mass) over time. AutoISO_PLUM is used with systems connected to Ortec analysers.

The features of the software are as follows:

- An interface for acquisition and analysis
- Graphic tools to visualise measurement results
- Excel compatible summary tables

This code includes multiple automatic calculations of the isotopic composition of Pu or U using all available codes (MGA, MGA ++, IGA, and PC/FRAM).

These acquisitions were done up to a minimum of 5.0 X 10^6 counts in the spectrum (recommended value by the code developer) and were stopped when the result stabilized. The evolution of the enrichment value according to the acquisition time was compared with the reference value in the measurement configurations defined above. It was then possible to check if the enrichment given by the code converged toward the reference value after some counting statistics. It was also essential to check that the uncertainty given by the code was realistic. The used method was developed in the scope of *Action Sheet 3 – Cooperation on testing the Gamma-Ray Isotopic Analysis Software FRAM v5 and MGA++ v1.1*. It is described in [6].

4.2. LANL ECHRGS Data Collection

The spectra were acquired with the DETECTIVE SN258. The average resolution of the detector during the acquisition was about 1.3 keV at 122 keV and 2.0 keV at 1.33 MeV. LANL measured

one wide-range U set, one wide-range Pu set, and one MOX set. For each item, 16 spectra of 15 minutes real time were acquired. The samples were placed as close to the detector as possible such that the dead time was not more than 55%. The U set consisted of five low-enriched uranium (LEU) items of the New Brunswick Laboratory NBL Certified Reference Material (CRM) 969 set and the three HEU items of the NBL CRM 146 set with the enrichment ranging from 0.31% to 93.18% ^{235}U. These samples were approximately 200 g each. The Pu set consisted of four of the seven samples from the Pu isotopic determination inter-comparison exercise (PIDIE) set: PIDIE-1, PIDIE-3, PIDIE-5, and PIDIE-7. For this set, the ^{240}Pu ranges from 6% to 26%. These samples were small (only 0.4 g each). For the MOX set, five samples with a U/Pu ratio ranging from 0.3 to 38 were measured. For this set, the nuclear materials are 93% ^{235}U and 93% ^{239}Pu. The total nuclear mass of each sample was about 4 g.

4.3. LLNL ECHRGS Data Collection

A set of twelve 1 g U gamma-ray standards (ranging from DU to HEU) from the National Bureau of Standards (NBS) was used in this study, along with six Pu gamma-ray standards from the PIDIE set.

5. Isotopic Analysis Comparison: LNCHRGS vs. ECHRGS

5.1. Isotopic Analysis Using MGA++/U235HI

The goal of this portion of the work was to understand the limitations/biases of MGA++ for Pu analysis and U235HI for U analysis when applied directly to the detection systems that have energy resolution slightly (50–350 eV) outside the recommended (< 600 eV) energy resolution specified by the software. LLNL and LANL used the following detection systems to conduct this comparison:

- Two CANBERRA Falcon 5000 ECHRGS systems (120-keV energy resolution of ~820 eV and ~920 eV, respectively).
- A safeguards type LNCHRGS system, the Ortec DETECTIVE SGD-GEM-5030P4 with an energy resolution of ~650 eV at 120 keV.

U sources were placed in front of the detection systems for multiple 1200-second (real time) measurements. For the Canberra Falcon ECHRGS detector, associated internal electronics were used. For the safeguards HPGe system, integrated DSPECPro electronics were used. Cadmium absorbers of thicknesses ranging from 0.1–0.2 mm were placed in between the six PIDIE standards (PIDIE #2–#7), and the detection systems attenuated the strong 59-keV gamma-rays from the decay of ^{241}Am. The same measurements with 1200-second real time were used as in the Pu data collection, and the source-detector distance was 2 in. A total of 1200 spectra were collected, and, given time constraints, a subset of 400 were analyzed. The Pu and U isotopic ratios were analyzed using MGA++ v1.06 and U235HI, respectively. The analysis results are illustrated in **Figures 1 and 2**.

From this study without absorbers, the slight (50–350 eV) departure from the required MGA++ resolutions at 120 keV resulted in 5–10% deviation in ^{235}U for some of the U samples and 2–6% deviation in ^{239}Pu for higher burn-up Pu samples.

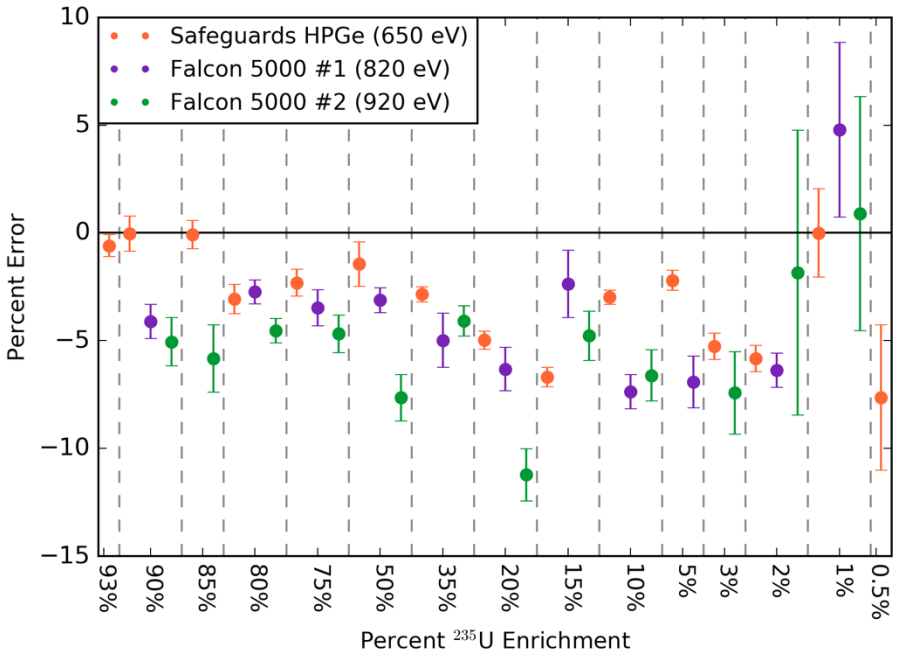

Figure 1. Results of the analysis of isotopic ratios. Percent deviation from the reference values for the three detection systems with respect to various 235U enrichments using the U235HI routine. This analysis showed a systematic bias in isotopic analysis of data taken using the Falcon ECHRGSs.

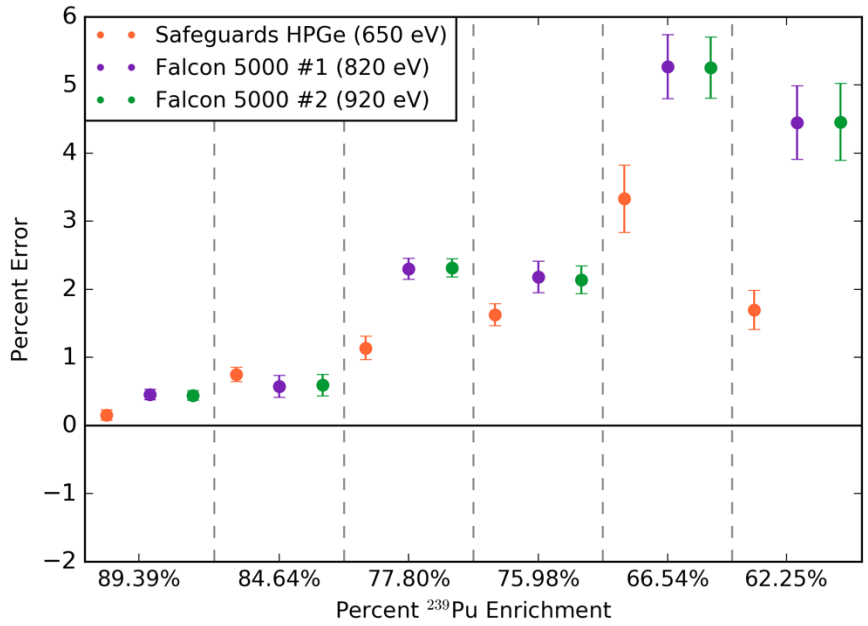

Figure 2. Results of the analysis of isotopic ratios. Deviations from the ^{239}Pu reference values using MGA++ v1.06 analysis for the three detection systems. The analysis code shows an increasing bias in the isotopic analysis with decreasing ^{239}Pu concentration.

5.2. Isotopic Analysis Using FRAM

The FRAM analysis includes two distinct components: the FRAM code and the parameter set. FRAM v5.1 was released in 2011. This version of the code did not have built-in parameter sets to analyze spectra collected using the Ortec DETECTIVE®. Therefore, LANL created a new parameter set "u_detective_121-1001" to analyze the U spectra obtained with the Ortec DETECTIVE®. LANL then analyzed the data collected using the Ortec DETECTIVE® for the eight U reference standards with the enrichment ranging from 0.31% to 93.18% ^{235}U of the NBL CRM 969 and NBL CRM 146 sets. The results were then compared with the results of the analysis of the spectra obtained with a 25% relative efficiency coaxial HPGe detector on the same eight items. **Figure 3** shows the comparison. Each data point from both the Ortec DETECTIVE® and the coaxial HPGe is the average of 16 results of 16 spectra.

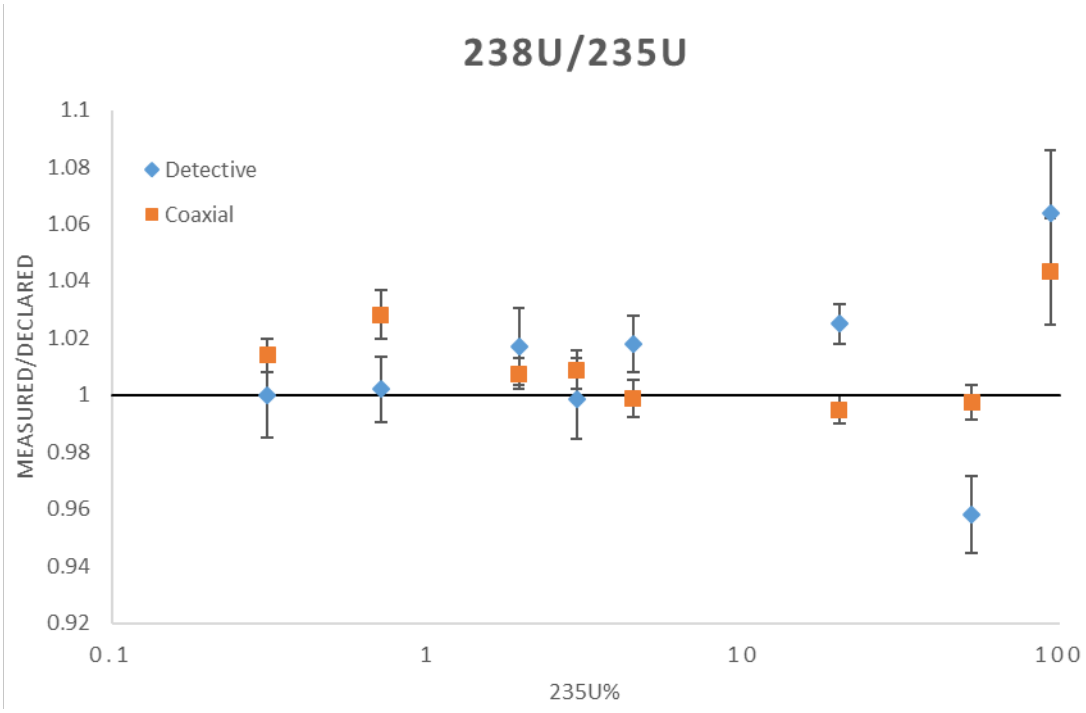

Figure 3. Results from the comparison of the $^{238}U/^{235}U$ ratios of the Ortec DETECTIVE® with those of the coaxial HPGe detector systems. The chart shows the measured $^{238}U/^{235}U$ ratios divided by the declared $^{238}U/^{235}U$ ratios. The error bars are relative uncertainties of the measured $^{238}U/^{235}U$ ratios.

There was no Pu comparison since there were no parameter sets for FRAM v5.1 to analyze the Pu spectra of the Ortec DETECTIVE®.

LANL released FRAM v5.2 with some upgrades from FRAM v5.1 in 2013, but FRAM v5.2 did not have the ECHRGS parameter sets built in. Thus, for this project LANL created the ECHRGS parameter sets and imported them into the FRAM database to improve the capability of FRAM v5.2 to analyze spectra obtained using ECHRGS systems. LANL created four parameter sets for the Ortec DETECTIVE® that can be used to analyze various types of U and Pu spectra [4]:

1. "pu_detective_120-460" to analyze Pu using the middle-energy region
2. "pu_detective_180-1010" to analyze Pu using the high-energy region
3. "uleu_detective_120-1010" to analyze LEU, natural U (NU), and DU using the high-energy region
4. "uheu_detective_120-1010" to analyze HEU using the high-energy region

For the Canberra Falcon, since its resolution is comparable to that of a typical coaxial detector, the standard built-in parameter sets of FRAM v5.2 developed for the coaxial detector can be used. Additionally, for a good Falcon with above-average resolution (< 800 eV at 122 keV), the parameter sets ULEU_Plnr_060-250 and UHEU_Plnr_060-250 employing the X-ray region can be used to analyze both LEU and HEU.

6. Updated FRAM and U235HI Algorithms

6.1. FRAM and U235HI Algorithm Evaluation

The results of the IRSN tests performed on the LANL, LLNL, and IRSN databases are presented in the tables below.

Table 3 presents two types of results obtained by analyzing LANL and LLNL CRM sources using FRAM v5.2, U235HI v0.9, and IGA 7.0 codes:

- Mean relative difference between the code result and ^{235}U% reference value (Δ%)
- Standard deviation obtained on the ^{235}U enrichment value when repeating the measurement between 16 and 80 times

The discrepancies for the U235HI analysis were discussed during a workshop held at LLNL in April 2017 that included all parties involved in Action Sheet 4, and it was stated that LANL did the gamma-ray acquisition using a 0.5 mm Cd absorber. The consequence was the absorption of gamma lines in the X-ray region that are used by U235HI. U235HI performed well in the LLNL measurement campaign where no Cd absorber was present. FRAM v5.2 performed well for all enrichments, whereas IGA v7.0 had various performances with underestimated uncertainties for HEU.

Table 4 **and Table 5** present the results obtained by analyzing certified U_3O_8 powder material available at IRSN, again with the FRAM v5.2, U235HI v0.9, and IGA v7.0 codes. The main difference between Tables 2 and 3 is the presence of an additional steel absorber between the U sample and the detector for Table 3.

Table 3. Results obtained by analyzing LANL and LLNL CRM sources using FRAM v5.2, U235HI v0.9, and IGA v7.0 codes.

Measurement Number	Certified value for U235	Software	Δ%	σ deviation	Number of measurements	Total counts	Percent Successful Analysis
u0031 (LANL)	0.3166	FRAM (5.2)	0%	0.02	17	3880671	100%
		U235HI (0.9)	326%	0.37	17	3880671	100%
		IGA (7.0)	-4%	0.01	17	3880671	100%
u0071 (LANL)	0.7119	FRAM (5.2)	-1%	0.03	16	4064066	100%
		U235HI (0.9)	205%	0.33	16	4064066	100%
		IGA (7.0)	-4%	0.03	16	4064066	100%
u0071 (LLNL)	0.7119	FRAM (5.2)	-3%	0.69	36	1.8E+07	100%
		U235HI (0.9)	1%	0.00	36	1,8E+07	100%
		IGA (7.0)	-8%	0.09	36	1,8E+07	100%
u0194 (LANL)	1.9421	FRAM (5.2)	-3%	0.12	16	4519483	100%
		U235HI (0.9)	169%	0.39	16	4519483	100%
		IGA (7.0)	-2%	0.07	16	4519483	100%
u0194 (LLNL)	1.9421	FRAM (5.2)	-4%	0.10	41	4662539	100%
		U235HI (0.9)	0%	0.02	41	4662539	100%
		IGA (7.0)	-13%	0.12	41	4662539	100%
u0295 (LANL)	2.9492	FRAM (5.2)	-2%	0.15	16	4820418	100%
		U235HI (0.9)	146%	0.42	16	4820418	100%
		IGA (7.0)	3%	0.11	16	4820418	100%
	2.9492	FRAM (5.2)	-3%	0.16	30	4841264	100%

Measurement Number	Certified value for U235	Software	Δ%	σ deviation	Number of measurements	Total counts	Percent Successful Analysis
u0296 (LLNL)		U235HI (0.9)	-1%	0.02	30	4841264	100%
		IGA (7.0)	-18%	0.11	30	4841264	100%
u0446 (LANL)	4.4623	FRAM (5.2)	-4%	0.17	16	5526652	100%
		U235HI (0.9)	193%	12.57	16	5526652	100%
		IGA (7.0)	2%	0.23	16	5526652	100%
u446 (LLNL)	4.4623	FRAM (5.2)	-3%	0.27	40	4791115	100%
		U235HI (0.9)	-1%	0.03	40	4791115	100%
		IGA (7.0)	-17%	0.22	40	4791115	100%
u2006 (LANL)	20.07	FRAM (5.2)	-1%	0.60	16	8925413	100%
		U235HI (0.9)	173%	58.68	16	8925413	100%
		IGA (7.0)	-8%	2.03	16	8925413	100%
u201 (LLNL)	20.1	FRAM (5.2)	-2%	0.77	80	5552539	100%
		U235HI (0.9)	1%	0.25	80	5552539	100%
		IGA (7.0)	-9%	1.68	80	5552539	100%
u5256 (LANL)	52.488	FRAM (5.2)	3%	2.18	16	693867	100%
		U235HI (0.9)	61%	11.62	16	693867	100%
		IGA (7.0)	-23%	13.28	16	693867	100%
u540 (LLNL)	54	FRAM (5.2)	-7%	2.90	40	6373968	100%
		U235HI (0.9)	-1%	12.48	40	6373968	98%
		IGA (7.0)	-3%	5.81	40	6373968	100%
u9318	93.17	FRAM (5.2)	0%	0.51	16	9506793	100%

Measurement Number	Certified value for U235	Software	Δ%	σ deviation	Number of measurements	Total counts	Percent Successful Analysis
(LANL)		U235HI (0.9)	7%	65.04	16	9506793	100%
		IGA (7.0)	2%	1.36	16	9506793	100%
u931 (LLNL)	93.17	FRAM (5.2)	-1%	0.67	80	8504342	100%
		U235HI (0.9)	-21%	95.99	80	8504342	93%
		IGA (7.0)	2%	1.36	80	8504342	100%

Table 4. Results obtained by analyzing certified U_3O_8 powder material available at IRSN using FRAM v5.2, U235HI v0.9, and IGA v7.0 codes.

Measurement Number	Certified value for ^{235}U	Software	Δ%	σ deviation	Number of measurements	Total counts	Percent Successful Analysis
8426-1	0.34	FRAM (5.2)	13%	0.04	5	5.00E+06	100%
		U235HI (0.9)	36%	0.00	5	5.00E+06	100%
		IGA (7.0)	5%	0.03	5	5.00E+06	100%
8426-2	0.54	FRAM (5.2)	2%	0.06	2	5.00E+06	100%
		U235HI (0.9)	37%	0.01	2	5.00E+06	100%
		IGA (7.0)	0%	0.01	2	5.00E+06	100%
8426-20	0.71	FRAM (5.2)	-7%	0.02	4	5.00E+06	100%
		U235HI (0.9)	13%	0.01	4	5.00E+06	100%
		IGA (7.0)	-7%	0.04	4	5.00E+06	100%
07U005	0.71	FRAM (5.2)	-4%	0.10	2	5.00E+06	100%
		U235HI (0.9)	8%	0.03	2	5.00E+06	100%
		IGA (7.0)	-15%	0.13	2	5.00E+06	100%
8426-4	1.532	FRAM (5.2)	2%	0.11	2	5.00E+06	100%
		U235HI (0.9)	23%	0.01	2	5.00E+06	100%
		IGA (7.0)	4%	0.14	2	5.00E+06	100%
8426-5	3.04	FRAM (5.2)	-4%	0.14	4	5.00E+06	100%
		U235HI (0.9)	12%	0.03	4	5.00E+06	100%
		IGA (7.0)	-5%	0.27	4	5.00E+06	100%

Measurement Number	Certified value for ^{235}U	Software	Δ%	σ deviation	Number of measurements	Total counts	Number of successful analysis %
LM-5-1-LM-5-2	3.25	FRAM (5.2)	-4%	x	1	5.00E+06	100%
		U235HI (0.9)	11%	x	1	5.00E+06	100%
		IGA (7.0)	-8%	x	1	5.00E+06	100%
8426-6	5.49	FRAM (5.2)	6%	0.45	3	5.00E+06	100%
		U235HI (0.9)	18%	0.02	3	5.00E+06	100%
		IGA (7.0)	3%	0.35	3	5.00E+06	100%
8426-8	11.304	FRAM (5.2)	4%	0.91	4	5.00E+06	100%
		U235HI (0.9)	13%	0.20	4	5.00E+06	100%
		IGA (7.0)	9%	0.24	4	5.00E+06	100%
8426-9	14.21	FRAM (5.2)	-15%	0.15	2	5.00E+06	100%
		U235HI (0.9)	15%	x	1	5.00E+06	100%
		IGA (7.0)	-1%	x	1	5.00E+06	100%
8426-10	21.9	FRAM (5.2)	-2%	0.65	2	5.00E+06	100%
		U235HI (0.9)	16%	0.19	2	5.00E+06	100%
		IGA (7.0)	2%	2.26	2	5.00E+06	100%
8426-11	29.187	FRAM (5.2)	-1%	1.07	3	5.00E+06	100%

Measurement Number	Certified value for ^{235}U	Software	Δ%	σ deviation	Number of measurements	Total counts	Number of successful analysis %
		U235HI (0.9)	10%	0.24	3	5.00E+06	100%
		IGA (7.0)	-12%	1.70	3	5.00E+06	67%
8426-12	45.11	FRAM (5.2)	7%	2.93	2	5.00E+06	100%
		U235HI (0.9)	20%	2.07	2	5.00E+06	100%
		IGA (7.0)	x	x	2	5.00E+06	0%
8426-18	45.11	FRAM (5.2)	15%	0.34	4	5.00E+06	50%
		U235HI (0.9)	19%	0.55	4	5.00E+06	100%
		IGA (7.0)	18%	x	4	5.00E+06	25%
8426-13	57.04	FRAM (5.2)	4%	0.40	2	5.00E+06	100%
		U235HI (0.9)	25%	0.85	2	5.00E+06	100%
		IGA (7.0)	x	x	2	5.00E+06	0%
8426-14	68.043	FRAM (5.2)	0%	0.40	1	5.00E+06	100%
		U235HI (0.9)	19%		1	5.00E+06	100%
		IGA (7.0)	4%	x	1	5.00E+06	100%
8426-15	89.3	FRAM (5.2)	-3%	3.33	4	5.00E+06	100%
		U235HI (0.9)	5%	2.96	4	5.00E+06	100%
		IGA (7.0)	-4%	12.49	4	5.00E+06	100%

Table 5. Results obtained by analyzing certified U_3O_8 powder material available at IRSN using FRAM 5.2, U235HI 0.9, and IGA 7.0 codes with an additional stainless steel absorber (2, 5, or 10 mm) between the U sample and the detector.

Measurement Number	Certified value for ^{235}U	Software	Δ%	σ deviation	Number of measurements	Total counts	Percent Successful Analysis
8426-1	0.34	FRAM (5.2)	16%	0.05	5	5.00E+06	100%
		U235HI (0.9)	308%	0.76	5	5.00E+06	100%
		IGA (7.0)	2%	0.10	5	5.00E+06	100%
8426-2	0.54	FRAM (5.2)	7%	0.03	2	5.00E+06	100%
		U235HI (0.9)	406%	0.12	2	5.00E+06	100%
		IGA (7.0)	-30%	x	2	5.00E+06	50%
8426-20	0.71	FRAM (5.2)	4%	0.07	4	5.00E+06	100%
		U235HI (0.9)	118%	0.39	4	5.00E+06	100%
		IGA (7.0)	-8%	0.17	4	5.00E+06	100%
8426-4	1.532	FRAM (5.2)	6%	0.05	2	5.00E+06	100%
		U235HI (0.9)	175%	0.44	2	5.00E+06	100%
		IGA (7.0)	19%	0.07	2	5.00E+06	100%
8426-5	3.04	FRAM (5.2)	-3%	0.11	4	5.00E+06	100%
		U235HI (0.9)	108%	1.28	4	5.00E+06	100%
		IGA (7.0)	-10%	0.12	4	5.00E+06	100%
8426-6	5.49	FRAM (5.2)	-7%	x	1	5.00E+06	100%
		U235HI (0.9)	117%	x	1	5.00E+06	100%
		IGA (7.0)	-5%	x	1	5.00E+06	100%
8426-8	11.304	FRAM (5.2)	1%	0.27	4	5.00E+06	100%
		U235HI (0.9)	51%	0.89	4	5.00E+06	100%

Measurement Number	Certified value for ^{235}U	Software	Δ%	σ deviation	Number of measurements	Total counts	Percent Successful Analysis
		IGA (7.0)	9%	3.28	4	5.00E+06	100%
8426-9	14.21	FRAM (5.2)	2%	0.09	2	5.00E+06	100%
		U235HI (0.9)	48%	0.00	2	5.00E+06	100%
		IGA (7.0)	-12%	0.56	2	5.00E+06	100%
8426-10	21.9	FRAM (5.2)	5%	x	1	5.00E+06	100%
		U235HI (0.9)	54%	x	1	5.00E+06	100%
		IGA (7.0)	x	x	1	5.00E+06	0%
8426-11	29.187	FRAM (5.2)	-5%	1.85	4	5.00E+06	100%
		U235HI (0.9)	26%	1.37	4	5.00E+06	100%
		IGA (7.0)	4%	0.60	4	5.00E+06	75%
8426-12	45.11	FRAM (5.2)	3%	2.62	4	5.00E+06	100%
		U235HI (0.9)	31%	5.72	4	5.00E+06	100%
		IGA (7.0)	-8%	x	2	5.00E+06	50%
8426-18	45.11	FRAM (5.2)	5%	3.78	4	5.00E+06	100%
		U235HI (0.9)	14%	6.00	5	5.00E+06	100%
		IGA (7.0)	35%	5.12	5	1,00E+00	80%
8426-13	57.04	FRAM (5.2)	-1%	1.80	2	5.00E+06	100%
		U235HI (0.9)	23%	4.80	2	5.00E+06	100%
		IGA (7.0)	x	x	2	5.00E+06	0%
8426-15	89.3	FRAM (5.2)	-3%	1.32	3	5.00E+06	100%
		U235HI (0.9)	-6%	18.53	3	5.00E+06	100%
		IGA (7.0)	1%	5.55	3	5.00E+06	100%

6.2. U235HI Modifications

The U235HI program was developed under Action Sheet 4. It differs from the traditional ^{235}U analysis of the gamma-ray region below 200 keV due to the poor resolution of the Ortec DETECTIVE® when compared with a planar HPGe detector. Alternatively, the U235HI code uses

the gamma-ray region from 90 keV to 1 MeV to perform the analysis of the U spectrum. This program uses the original MGA methodology, which calculates the detector efficiency, absorber thickness, and U thickness and then obtains the three U isotope ratios: ^{234}U, ^{235}U, and ^{238}U [5]. To perform calibration to this software, a set of U spectra was collected using an Ortec MicroDetective with both NBL CRM 969 and NBL CRM 146 gamma-ray standards. In the IRSN tests, using all available spectra under AutoISO_PLUM, there was good agreement with the LLNL set of the MicroDetective spectra; however, U235HI showed significant biases in the rest of the pool spectra. Under Action Sheet 4 Task 6, U235HI was modified to allow the user to include the presence of a cadmium filter to address this deficiency. Using this filter reduces both the instrument dead time and the detector response at energy below approximately 200 keV. With this correction, the analysis of the LANL data listed in

Table 6 shows improved agreement. However, the relative difference between the measured versus declared values is still quite large for the U235HI v1.1 code, and this difference should be considered when performing independent verifications of operator materials.

Table 6. Comparison of U235HI v1.0 with the inclusion of cadmium filter added in v1.1. The results from v1.1 are the averages for five spectra from the international database for that enrichment standard.

Percent ^{235}U Enrichment	U235Hi Version	
	v0.9	v1.1
0.31	1.079	0.36
0.71	2.053	0.81
1.94	3.33	2.10
2.95	4.44	3.24
4.46	7.87	4.72
20.06	20.8	18.84

7. RADID Nuclide Identification Software

7.1. RADID Software Description

The RADID rapid automatic identification code for HPGe detectors was developed at LLNL by Thomas Gosnell et al. [1,2,3]. This gamma-ray spectrum analysis program for a rapid screening, rule-based heuristic system can identify well over 200 radioactive sources of interest in addition to U and Pu. The execution time is about one second with minimal user interactions. For Action Sheet 4, RADID was modified to allow for the subtraction of background from the spectra so that users could correct background interferences under conditions of high radiation background from the environment. The analysis routines were updated to account for the changes in the spectra when performing the peak identification and analysis.

The software was developed using mostly simulated spectra, with only a few field-collected spectra. The IRSN collaborators used the RADID software with the Ortec DETECTIVE® and identified the following issues with the software: (1) the inability to perform weak peak fitting regardless of different signal to noise input; (2) the inability to perform full-width-half-maximum (FWHM) calculations even when the FWHM-fitting results at the two different energies are available; and (3) the inability to complete the final identification in some cases even when the

nuclide was identified in the top-10 list, such as with Cd-109. IRSN has made these spectra available to LLNL for reference.

7.2. RADID Software Testing and Results

LLNL RADID nuclide software identification capabilities were tested at IRSN based on three spectra databases:

- A database of spectra acquired from reference sources under laboratory measurement conditions using two IRSN instruments: Ortec DETECTIVE® n°258 and MicroTransSpec n°7309 (HPGe crystal dimensions: 50 mm diameter and 35 mm length). Both instruments are shielded by 4 mm of tungsten and set with the energy calibration required by the Ortec DETECTIVE® identification software (~0.3662 keV/channel on 8192 channels) in order to cover the energy range from 0 to 3 MeV. The Ortec DETECTIVE® n°258 has a resolution of 1.4 keV at 122 keV, and the MicroTransSpec has a resolution of 1.2 keV at 122 keV. The database contains 150 spectra collected on radioactive sources such as ^{60}Co, ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{241}Am, ^{252}Cf, ^{66m}Ho, ^{237}Np, ^{226}Ra, ^{88}Y, ^{51}Cr, ^{57}Co, ^{75}Se, ^{85}Sr, ^{109}Cd, ^{113}Sn, ^{139}Ce, ^{244}Cm, and mixed radionuclide sources, but also on U samples (DU up to HEU), Th samples, mixtures of U and ^{133}Ba or ^{137}Cs or mixed radionuclide or Th samples, and shielded U and Th samples. All these spectra were primarily analyzed using the embedded Ortec DETECTIVE® software.
- A database of spectra acquired by IRSN using its two instruments (Ortec DETECTIVE® n°258 and MicroTransSpec n°7309) set with the energy calibration required by the DETECTIVE® identification software, during on-site inspections in order to test the environmental background subtraction capabilities of RADID. All these spectra were analyzed with IRSN Reconnaissance software. A background measurement is available for each acquisition. The database contains 60 spectra collected on nuclear materials during on-site measurement campaigns such as MOX, Pu, U (DU, NU, LEU, HEU, and ^{233}U), mixtures of Pu and ^{137}Cs or ^{237}Np or U and Th, mixtures of U with Th, ^{241}Am, and ^{243}Am.
- A database of theoretical (obtained by numerical simulation) environmental spectra used as entry data for nuclear accident gamma spectrometry inter-comparison exercises, in order to test the robustness of the code on complex spectra representative of radionuclides that would be rejected in the environment in case of a nuclear accident that would impact either a nuclear power plant or a nuclear fuel cycle facility. The database contains several synthetic spectra generated from environmental radionuclide rejections according to different nuclear accident scenarios.

The evaluation was made based on the following identification result indicators:

- The detection of all present radionuclides
- One or several present radionuclides not detected and/or a radionuclide suspected but not definitively detected
- None of the present radionuclides detected
- One or several that was/were not present where the radionuclide was detected

In addition to the identification result evaluation, the accuracy of U and Pu categorization was also recorded in terms of U enrichment range (DU, NU, LEU, or HEU) and Pu type (reactor grade or weapon grade). Three U or Pu category result indicators were considered: category ok, no category announced, or erroneous category announced.

In terms of user-friendliness of the RADID software for an Ortec DETECTIVE® spectrum automatic identification, the useful amount of reported information in the ID details window, as well as the visualization of the checked lines, was clearly beneficial. The energy calibration, FWHM, and peak-fitting options proved to be powerful tools, making the identification more robust, but a certain practice is needed to have them appropriated. The software could be improved by allowing the storage of findings and results data in a dedicated result file.

In terms of identification performances, the RADID software properly identified the present radionuclides in 85% of the cases for laboratory measurements and in 62% of the cases for in-situ measurements when using RADID as a black box to compare the identification results with the results from the embedded Ortec DETECTIVE® identification software (i.e., no false negatives or positives). Some of the identification problems encountered by IRSN were solved by correcting the energy calibration using the interactive Ecal functionality of the RADID software. For instance, in the case of a pure Th source spectra acquired over a two-day period, the RADID recognized ^{212}Bi and possibly Th X-rays but not ^{228}Ac, whereas the Ortec DETECTIVE® software found naturally occurring radioactive material (NORM)-Th. This case was discussed during an April 2017 workshop at LLNL and showed the importance of the RADID user checking and refining the energy calibration. Once the energy calibration was completed, the result of the identification was improved; ^{212}Pb, ^{212}Bi, ^{228}Ac, and likely ^{232}Th were found. In the case of a ThO_2 sample associated with a U_3O_8 powder sample (DU), the Ortec DETECTIVE® software found both NORM-Th and U. The RADID software identified several potential sources, including: "^{232}Th background likely", "^{238}U", and "likely processed U"; however, RADID also indicated that "uranium not manifest."

For samples with low counting statistics the RADID software either suspects or does not identify the radionuclide of interest. For instance, for MOX cases encountered on-site, the presence of U and/or Pu is not always manifest, whereas the gamma lines of ^{235}U at 185.7 keV and/or ^{241}Pu at 208 keV are statistically present in the spectrum (net area > 4 net area uncertainty when using the region of interest analysis report from GammaVision®). IRSN Reconnaissance software uses a statistical criterion to directly compare the count rate in a gamma line of interest to the one evaluated in the same region of interest during the background measurement where no sample is present. The count rate of each gamma line of interest, selected by the user, is calculated based on a region of interest analysis. The Reconnaissance software is nevertheless adapted to nuclear material identification measurements held in facilities and is limited to 16 gamma lines of interest, whereas RADID has 200 radionuclides it its library. When using the background subtraction option, existing radionuclides that are detected and mentioned as manifest in the "HPGe ID details" are displayed as not manifest in the "HPGe ID result output" window of the software. Such an option has to be carefully used, as it is a channel by channel subtraction of the background.

In some cases, one or several non-present radionuclides were detected, these results are summarized in Table 7. For example, the RADID analysis indicated the presence of ^{239}Pu, which was not actually present, in a pure ^{237}Np sample. In several cases the software indicated the presence of medical radionuclides that did not exist in the sample. For instance, in the case of a pure ^{152}Eu source, the software indicated the presence of ^{82}Rb/^{82}Sr, ^{47}Ca, and ^{110m}In in addition to ^{152}Eu. In the case of the AmLi source, the software indicated the presence of ^{115m}In, ^{237}Np, ^{51}Cr, and ^{7}Be in addition to ^{241}Am. In the case of a pure ^{133}Ba source, the software indicated the presence of ^{123m}Te and ^{47}Sc in addition to ^{133}Ba. In the case of a pure Th source, the software indicated the presence of ^{149}Pm, ^{172}Lu, ^{195m}Pt, ^{50}V, ^{207}Bi, and ^{7}Be in addition to ^{212}Bi. In the case of a pure ^{75}Se

source, the software indicated the presence of ^{203}Pb and ^{133m}Ba in addition to ^{75}Se. In the case of a pure Pu source, the software indicated the presence of ^{135m}Ba in addition to ^{239}Pu. IRSN presumes that these false positives came from the application of heuristic rules to a very large number of radionuclides (200) with only a small number of gamma lines for each of them.

In the masking scenario of HEU with ^{266m}Ho, ^{235}U was not recognized.

Table 7. Comparison of RADID source identification for various radioactive materials.

Source Material	Material Identified
^{237}Np	^{239}Pu
^{152}Eu	^{2}Rb/^{82}Sr, ^{47}Ca, ^{110m}In,^{152}Eu
AmLi	^{115m}In, ^{237}Np, ^{51}Cr, ^{7}Be, ^{241}Am
^{133}Ba	^{123m}Te and ^{47}Sc in addition to ^{133}Ba
Th	^{149}Pm, ^{172}Lu, ^{195m}Pt, ^{50}V, ^{207}Bi, ^{7}Be, ^{212}Bi
^{75}Se	^{203}Pb, ^{133m}Ba, ^{75}Se
Pu	^{135m}Ba, ^{239}Pu

Following these tests, LLNL and IRSN discussed the spectrum analysis process and identification result expression (i.e., gamma-ray photopeak presence criteria, detection limit, and confidence level associated with the identification result) during the April 2017 *INSEP–IRSN AS-4, AS-6 Meeting* at LLNL. IRSN feedback and findings were discussed during this workshop. Some difficult cases were analyzed together with Tzu-Fang Wang, and additional information about the code functionalities was discussed. IRSN feedback on the use of the RADID software was transmitted to LLNL RADID developers (T. Gosnell, J.C Chavez) for review and further information was requested to gain a better understanding regarding the performance of the code. The developer responses (LLNL-TR-735478) addressed several of the questions regarding the user interface, the methodology for determining the signal-to-noise ratio, and the rationale for the selection of certain gamma-ray energies for analysis.

Some problems were encountered by IRSN when considering the categorization of U and Pu with an Ortec DETECTIVE® or a MicroTransSpec instrument. Concerning the categorization, from LLNL documentation and discussions it was determined that the ^{235}U enrichment range evaluation made by the RADID or Ortec DETECTIVE® embedded software is a gross evaluation based on the net area ratio at 185 and 1001 keV, specific gamma emission rates at those energies, detection efficiencies from 1-m distance measurements with 20% up to 170% relative efficiency detectors, and U metal auto-absorption coefficients at these energies. In the case of Pu, when the gamma lines in the 600 keV region are properly resolved, the ratio between the number of counts in the peaks of ^{239}Pu and ^{240}Pu in this region gives an approximate value of ^{240}Pu content and, consequently, information on the Pu category. All experts present at the April 2017 meeting agreed that, for U and/or Pu categorization, dedicated U and Pu isotopic composition determination codes should be applied on higher-statistics spectra instead of using RADID, which was not developed for this primary purpose.

8. Conclusion

Newer ECHRGS systems offer numerous advantages over traditional LNCHRGS instruments. ECHRGS systems are portable and do not require liquid nitrogen for operation, increasing the efficiency of safeguards implementation by saving inspector time. However, the reduced energy resolution of ECHRGS systems limits the applicability of the Pu and U isotopic analysis codes commonly used with LNCHRGS. This has presented a challenge for safeguards inspectors. Action Sheet 4 was initiated to attempt to address this issue. IRSN, LANL, and LLNL first examined the inaccuracies found in the isotopic analysis performed in FRAM v5.1, MGA++ v1.06, and U235HI v0.9 codes when used with ECHRGS systems. LANL and LLNL then made improvements to FRAM v5.2 and U235HI v1.1 codes to address these inaccuracies. IRSN tested the new beta versions of these codes. The results of the tests showed that the modified codes could now assess the isotopic concentration of U and Pu samples with a comparable level of accuracy when used with ECHRGS systems as they could previously when used with LNCHRGS systems. These modified codes provide new capabilities for safeguards inspectors to use ECHRGS, which will improve flexibility during inspections and increase the efficiency and accuracy of facility inspections.

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