The Use of FRAM with a Portable, HPGe-Based Nuclide Identifier to Measure the Isotopic Composition of Plutonium and Uranium

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Abstract

The ORTEC Detective is an HPGe (High-Purity Germanium)-based portable nuclide identifier designed to search for and identify radionuclides to support the fight against illicit trafficking of nuclear materials. The Detective contains an HPGe crystal approximately 50 mm in diameter by 30 mm thick, cooled by a miniature mechanical cooler that operates from either battery or line power. The system weighs 10.6 kg and is easily man-portable for measurements where conventional liquid nitrogen (LN_2) cooling for the detector is not available.

The Detective operates with a fixed, unadjustable gain of 0.3662 keV/ch spanning 3 MeV in 8192 channels. The large, fixed keV/ch value coupled with relatively poor HPGe crystal resolution (excellent resolution is not required for the Detective's search-and-identify purpose and is not specified for the instrument) makes this a challenging system to use for high-resolution spectroscopy applications such as gamma-ray isotopic analysis. Nevertheless, we have been able to successfully use the FRAM (Fixed energy, Response function Analysis with Multiple efficiencies) gamma-ray isotopic analysis code to extract the isotopic composition of uranium and plutonium from Detective gamma-ray spectra.

We will present data from 21 well-characterized plutonium and uranium samples, including 12 certified reference materials, measured with three different Detectives and one conventional LN_2-cooled coaxial detector under the same measurement conditions. The Detective data are generally unbiased but exhibit poorer statistical precision and greater variability compared with the LN_2-cooled coaxial detector.

Introduction

The ORTEC Detective is a portable, mechanically cooled HPGe-based nuclide identifier designed for search-and-identify homeland security and nuclear safeguards problems. Given these applications, the system does not require as high a resolution and operates at a much larger conversion gain than the HPGe detector systems typically used in spectroscopy applications. Nevertheless, the Detective's portability and operating conditions that do not require LN_2 make it a very attractive system for portable, quantitative nuclear safeguards measurements. It is very desirable to determine if the Detective, with its poor resolution and fixed conversion gain of 0.3662 keV/ch, can be used with the FRAM gamma-ray isotopic analysis software for isotopic measurements on uranium and plutonium in environments where LN_2 is not available.

Comparison Measurements

We made measurements on 21 well-characterized uranium and plutonium samples with three Detectives and one coaxial LN_2-cooled HPGe detector. The characteristics of the four detectors are presented in Table I.

The operational resolution is extracted from the FRAM analysis of the data from a 6% ^{240}Pu sample reported in this paper. The Detective was used at its fixed gain of 0.3662 keV/ch with data collected in 8192 ch. The LN_2 detector was used with the standard FRAM gain for a coaxial detector, 0.125 keV/ch in 8192 ch.

Table I. Detector Specifications and Performance

ID	Mfg./Model	Size/Efficiency	Purchase Resolution keV @ 6 µs		Operational Resolution keV @ 129 keV
			122 keV	1332 keV	
LN_2	Ortec GEM30P4	58.6 mm dia 55.7 mm length 32% Rel. Eff	0.65	1.62	0.706
SN 240	Ortec Detective	Approx 50 mm dia Approx 30 mm length Rel Eff not specified	Not specified		1.48
SN 259	Ortec Detective	Approx 50 mm dia Approx 30 mm length Rel Eff not specified	Not specified		1.34
SN 252	Ortec Detective	Approx 50 mm dia Approx 30 mm length Rel Eff not specified	Not specified		1.61

The first three detectors in Table I (LN_2, SN 240, and SN 259) were used together with a single sample under identical measurement conditions. Detective SN 252 was characterized separately at a later date under essentially the same measurement conditions. Counting times varied from 1500–11000 sec for a single measurement. The detectors were not shielded to mimic the way the Detective is used in the field, but Cd filters were used to attenuate the 60-keV gamma ray from ^{241}Am. The lack of shielding affected the analysis of lower enriched uranium samples in a manner discussed later. For most of the samples, the first three detectors in Table I collected only a single spectrum with single repeats on a few samples. Detective SN 252 was characterized with more extensive measurements with single 500-, 1000-, and 2000-s measurements for the plutonium samples and single 2000-, 4000-, and 8000-s measurements for the uranium samples. In addition, for Detective SN 252, we carried out repeated measurements (8–20 repeats) with 2000-s count times on most of the larger mass standards.

Table II describes the standards used in the measurements and their characterization.

FRAM Analysis

FRAM version 4.2 was used for the analysis. This version is functionally identical to ORTEC version 4.3 and Canberra version 4.4.

We modified standard FRAM parameter files to accommodate the characteristics of the Detective. For plutonium, we first combined a standard parameter file for analysis from 120–450 keV with a parameter file for analysis from 200–800 keV to produce a single parameter file to analyze unshielded data in the 120–800 keV range. Next, we made extensive modifications to the peak and background regions of interest (ROIs) to accommodate the poor resolution and large keV/ch of the Detective. Finally, we changed the default fitting parameters to accommodate the high value of keV/ch for the Detective.

We made similar changes for uranium in a single parameter file analyzing from 120–1001 keV for all enrichments. The uranium parameter file used gamma rays from the Th decay chain (usually present in uranium enriched above ~10%) to help determine the relative efficiency curve. The unshielded detector allowed background Th daughters to contribute to the spectrum, and, for this reason, we subtracted a background spectrum before we analyzed the uranium data. The background subtraction was not necessary for plutonium spectra.

Table II. Description of Standards and Their Characterization

Iso Std or Set	Description	Characterization
CBNM Set	A set of four Certified Nuclear Reference Material PuO_2 standards. Each standard contains a nominal 6 g of Pu. The samples have nominal percentages of 6.3-, 14.2-, 18.3-, and 25.4-wt% ^{240}Pu.	Prepared and certified by the Institute of Reference Materials and Measurements in Geel, Belgium, under the auspices of the Commission of the European Communities.
Iso Std Set	A set of five PuO_2 working reference materials with nominal percentages of 3-, 6-, 7-, 12-, and 15-wt% ^{240}Pu. Sample masses vary from 8–20 g Pu.	Extensive characterization at Los Alamos National Laboratory and the Mound Laboratory. .
PIDIE Set	A set of seven samples, nominally 0.4 g Pu each, fabricated for the Plutonium Isotopic Determination Intercomparison Exercise. The samples have nominal percentages of 6.0-, 10.1-, 14.1, 19.8-, 21.3-, 24.7-, and 26.8-wt% ^{240}Pu.	The accepted values for the isotopic fractions of these samples come from a Los Alamos analysis of the published mass spectrometry results from six European laboratories.
NBL Uiso set CRM 146	A Certified Reference Material for gamma ray spectrometry measurements of uranium enrichment/isotopic composition. The set contains three containers with nominal enrichments of 20-, 52-, and 93-wt% ^{235}U. Each container is filled with approximately 230 g of U_3O_8	A CRM distributed under the auspices of the New Brunswick Laboratory.
NBS enrichment series	A set of reference materials for gamma ray spectrometry measurements of uranium enrichment/isotopic composition. The set contains five containers with nominal ^{235}U enrichments of 0.3-, 0.7-, 1.9-, 2.9- and 4.5 wt%. Each container is filled with approximately 200 g of U_3O_8	Certified Nuclear Reference Materials certified jointly by the National Bureau of Standards NBS (now National Institute of Standards and Technology—NIST) and the Central Bureau of Nuclear Measurements (now IRMM) in Geel, Belgium.

The quality of the spectra from the Detective is significantly degraded compared with what FRAM expects from a typical LN_2 detector. Figures 1 and 2 compare spectra from the Detective with spectra from the very good quality LN_2 detector used in this study. The comparisons are for the two regions, 160 keV and 640 keV, where ^{240}Pu gamma rays are found in the Detective's useful energy range. The region around 160 keV contains 4 peaks in the "hump" extending from 160–161.5 keV, while the region between 638 and 642.5 also contains 4 peaks and becomes a particularly difficult analysis problem for samples with high ^{241}Am content.

Measurement Results

An example of the measurement results is shown in tabular form in Table III for Detective SN 259. This tabular form for the measurement results was carried out for all three Detectives and the LN2 detector for both uranium and plutonium standards. The standard deviation (sd) in Table III is the standard deviation of a single measurement from the variability of the 18 measurements in the table. No corrections are made for the differing count times. The summary averaged results for each detector for both uranium and plutonium are shown in Table IV.

The summaries for plutonium shown in Table IV show variability (% relative standard deviation—RSD) in the range of 2–4 times greater for the Detective compared with the LN_2 detector. In contrast, the variability of the results for ^{235}U is essentially the same for the Detective(s) and the LN_2 detector. The simple nature of the uranium spectrum

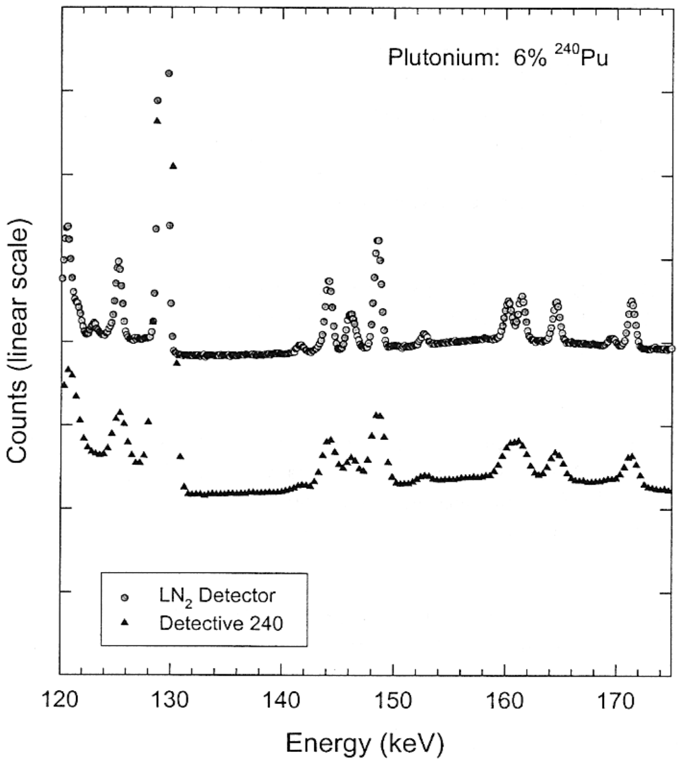

Fig. 1. Comparison of the spectra from the LN_2 detector (top) and the Detective (bottom) in the region from 120–175 keV.

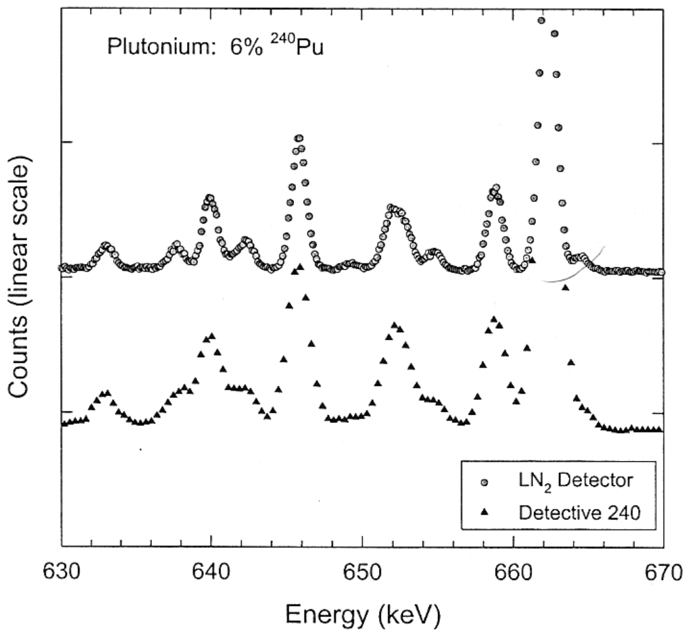

Fig. 2. Comparison of the spectra from the LN_2 detector (top) and the Detective (bottom) in the region from 630–670 keV.

does not place the demands on the peak fitting that the multiple overlapping and co-energetic peaks of a plutonium spectrum do. The poor resolution and large keV/ch values of the Detective spectra do not hinder the analysis of the uranium spectra, although there is some indication of increased variability for ^{234}U compared with the LN_2 spectra.

Repeated Measurements with Detective 252

Three of the larger plutonium samples were measured repeatedly with Detective 252, 20 measurements of 2000 s each. The measurement averages were unbiased for ^{239,240}Pu, and the measurement %RSD for these isotopes predicted in FRAM from counting statistics agreed very well with the %RSD observed from the distribution of the 20 measurements. This data is summarized for all the measured isotopes in Table V.

Summary

We have demonstrated that the FRAM gamma-ray isotopic analysis code can obtain very credible isotopic composition results for both plutonium and uranium when used with the ORTEC Detective, a mechanically cooled, portable, HPGe-based nuclide identifier. The measurement results are generally unbiased for the major isotopes, but the variability of the results for the plutonium isotopes is several times poorer than from a high-quality LN_2-cooled coaxial HPGe detector. The increased variability is attributed to the poorer resolution of the Detective compared with a LN_2-cooled detector coupled with the large fixed conversion gain (0.3662 keV/ch). This combination of attributes produces poorly resolved photopeaks in just a few channels of data, which lead to a decrement in the precision of individual measurements. The analysis of the less complex gamma-ray spectrum of uranium is affected much less than the analysis for plutonium.

Table III. Measured/Accepted Values for Plutonium Standards with Detective 259

Standard	wt % Pu240	Ct Time (s)	Pu-238	Pu-239	Pu-240	Pu-241	Am-241
STD ISO3	3.560	6800	0.4676	1.0010	0.9745	0.9033	1.1248
PIDIE6-1	5.965	3000	0.8737	1.0030	0.9528	0.9568	1.0684
PIDIE6-1	5.965	6000	0.8632	1.0041	0.9357	0.9861	1.0564
STD ISO6	6.131	11000	0.7078	0.9987	1.0205	0.9562	1.1083
CBNM Pu93	6.313	6000	0.8829	0.9998	1.0032	0.9452	1.0347
CBNM Pu93	6.313	2000	0.6944	1.0016	0.9766	0.9779	1.0167
STD ISO9	6.896	5000	0.7147	0.9998	1.0045	0.9621	1.0426
STD ISO12	11.861	5000	0.9446	0.9930	1.0524	0.9878	1.0165
PIDIE 6-3	14.160	3000	0.8593	1.0055	0.9673	1.0009	1.0130
PIDIE 6-3	14.160	6000	0.9512	1.0073	0.9559	1.0099	1.0157
CBNM Pu84	14.279	2800	0.9990	1.0010	0.9942	0.9992	0.9881
CBNM Pu84	14.279	1500	1.3539	1.0069	0.9577	0.9990	1.0083
CBNM Pu84	14.279	2000	0.8741	1.0048	0.9729	0.9719	1.0239
STD ISO15	15.541	5000	1.0139	0.9984	1.0094	0.9816	1.0104
CBNM Pu70	18.919	5400	1.0135	1.0193	0.9197	1.0190	0.9872
PIDIE6-5	21.389	10000	1.0040	1.0087	0.9688	1.0018	1.0040
CBNM Pu61	26.490	2000	1.0078	0.9958	1.0107	0.9946	0.9660
PIDIE6-7	26.540	6000	0.9812	1.0526	0.8669	1.0545	0.8944
		Avg.	0.9004	1.0056	0.9747	0.9838	1.0211
		sd	0.1848	0.0131	0.0421	0.0328	0.0512
		%RSD	20.52	1.30	4.32	3.33	5.01

Table IV. Summary and [Measured/Accepted] Averages for Plutonium and Uranium for Each Detector

		Pu-238	Pu-239	Pu-240	Pu-241	Am-241	U-234	U-235	U-238
LN2	Average	0.9544	1.0012	1.0008	0.9993	1.0002	0.9816	0.9924	0.9960
	% RSD	8.52	0.29	2.00	0.55	1.18	2.36	3.56	3.05
Detective 240	Average	0.9076	1.0043	0.9919	0.9731	1.0315	1.0778	1.0422	0.9886
	% RSD	16.88	0.90	4.33	4.53	4.84	9.81	2.60	1.76
Detective 259	Average	0.9004	1.0056	0.9747	0.9838	1.0211	0.9303	1.0039	1.0145
	% RSD	20.52	1.30	4.32	3.33	5.01	15.34	3.88	3.15
Detective 252	Average	0.9677	1.0046	0.9818	0.9672	1.0000	0.9921	1.0008	1.0045
	% RSD	8.97	0.65	3.39	3.71	3.66	6.64	3.38	3.48

%RSD is that of a single measurement of the 8 U or 18 Pu sample measurements considered in the average.

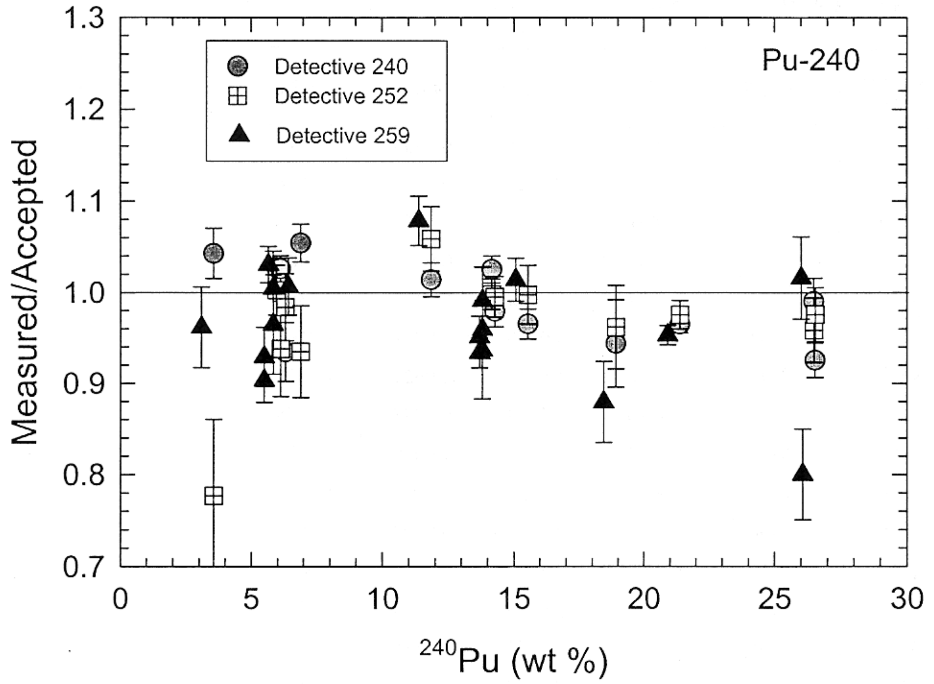

Fig. 3. Comparison of the individual measurements on three Detectives for the ^{240}Pu isotope.

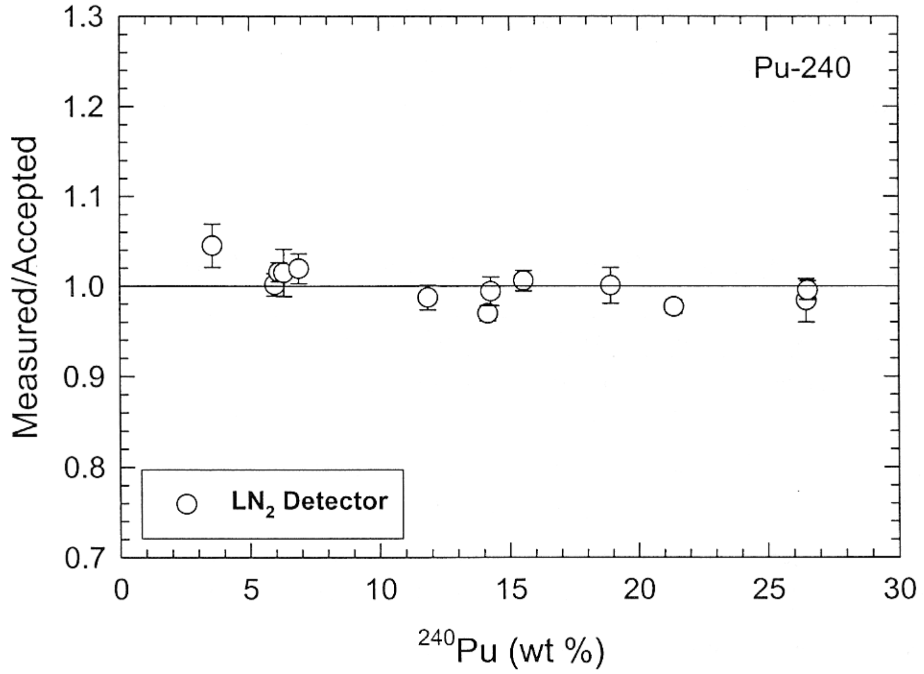

Fig. 4. Measurements with the LN_2 detector for the ^{240}Pu isotope.

Table V. Twenty 2000-s Measurements with Detective 252

Sample		Pu-238	Pu-239	Pu-240	Pu-241	Am-241
STD ISO6	Avg. Meas/Accept	0.9603	1.0009	0.9883	0.9085	1.0515
8.4 g Pu	% rsd, repeats	60	0.38	5.9	3.4	2.3
6.13% ^{240}Pu	%rsd, FRAM pred.	18	0.34	5.3	0.9	1.1
CBNM Pu93	Avg. Meas/Accept	0.7390	1.0005	0.9943	0.9453	0.9978
6 g Pu	% rsd, repeats	30.	0.28	4.1	1.6	0.7
6.31% ^{240}Pu	%rsd, FRAM pred.	21.	0.26	3.9	0.7	0.8
STD ISO9	Avg. Meas/Accept	0.9600	1.0009	0.9906	0.9202	1.0372
12 g Pu	% rsd, repeats	45	0.42	5.7	2.9	1.8
6.90% ^{240}Pu	%rsd, FRAM pred.	12	0.38	5.1	0.9	1.1