A New, Portable High-Resolution Gamma-Ray Instrument For Use In On-Site Safeguards Inspection

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Portable, high-resolution gamma spectroscopy has long been a vital technique in routine safeguards accountancy. The high resolution enables the spectrum analysis to accurately determine the isotopic content of the sample. All currently deployed systems for this application are based on liquid-nitrogen cooled, high-purity germanium (HPGe) detectors, with the spectrum analysis performed on portable PCs. These systems require the ready availability of liquid nitrogen. Detector cool down times on the order of 4 hours add to the difficulty of performing inspections, especially unannounced inspections. Such systems can be demanding of operator skill.

Recently, a simple-to–use new, portable high-resolution gamma spectroscopy system based on HPGe has become available. Liquid nitrogen cooling drawbacks have been eliminated by the use of a long-life, miniature Stirling-cycle cooler. This instrument has been developed as a hand-held radionuclide identifier (HHRIID), and is being widely deployed in prevention of illicit nuclear trafficking. It was designed to be easy to use by non-spectroscopists, such as first responders. The ID algorithm uses the high resolution abilities of HPGe, but the resolution requirements for trafficking applications are not as demanding as for accountancy applications.

Work by the Los Alamos National Laboratory has shown that a development version of PC/FRAM is capable of producing results with acceptably low levels of bias with HPGe detectors with resolution larger than previously used.

A new instrument is proposed which will combine the detection and identification abilities of the DETECTIVE for undeclared nuclear materials with the isotopic ratio abilities of PC/FRAM for providing accurate estimates of isotopic content of the located material.

The proposed instrument will be described and performance data measured with the current instrument and analyzed with PC/FRAM will be presented.

Introduction

In a recent paper[1], the IAEA called for instrumentation which could improve the investigation of undeclared nuclear facilities and materials.

The ORTEC DETECTIVE™ family members are the only commercially available radioisotope identification (RIID) devices which use High Purity Germanium (HPGe) technology. The DETECTIVE (see Figure 1), introduced in 2003, positively identifies the radionuclides present in the source and classifies them as "industrial" "medical", "natural" or "nuclear" in nearly any type of measurement situation.

Figure 1. DETECTIVE Portable Radioisotope Identification System (RIID).

While the energy resolution of the HPGe detector is somewhat degraded by the electromechanical cooler in comparison to LN2-cooled systems, it is perfectly acceptable for the purposes of a nuclide identifier, even for LEU, HEU and WG plutonium. Here it has been tested with PC/FRAM, the Los Alamos Isotopic Analysis code, and it has been shown that isotopic ratio estimates are easily achievable with accuracies useful in the interdiction of illicit trafficking. A proposal for the form of such an instrument is made.

The ORTEC DETECTIVE Family

The ORTEC DETECTIVE is a complete radioisotope identification system consisting of an electromechanically cooled HPGe detector, cryostat, cooler, signal electronics, analysis process, and user display. The detector is 50mm diameter x 30mm deep and is housed in a, ruggedized cryostat specially designed for portable use. The robust, long life cryostat is all-metal sealed and is cooled by a long life Stirling-cycle cooler. There are more than one hundred in continuous field operation. More than 50,000 hrs of operational life is expected. A major benefit of the design is that the detector may be "short-cycled"; that is, it can be turned off or restarted at any part of its warm-up-cool-down cycle without difficulties. The design has been shown to survive dropping onto concrete from more than 1 meter while operating.

A digital MCA with proprietary anti-microphonic rejection[2] circuitry is included. The instrument is controlled from a simple user interface with 1-button operation for search, dose rate, and identification functions. The nuclide identification software is licensed from LLNL[3]. The internal batteries are able to support the instrument for up to 3 hours, and external 12 volt DC power from a variety of sources can be used. In particular, it can operate or recharge from mains power or from a car battery making it useful for both unattended and unannounced safeguards activities.

The overall weight is less than 10 kg and maximum overall dimensions (including handle, HPGe detector end cap and shock absorbers) are 37 cm L x 32 cm H x 16 cm W. A high speed USB port allows connection to laptop computers.

The DETECTIVE-EX variant incorporates a personal digital assistant (PDA) providing nuclide identification and detection software, external communications capability, and expansion slots for cellular modems, additional data storage, or GPS. It also includes moderated ^{3}He detectors for neutron detection (see Figure 2).

A third variant, the trans-SPEC is a general purpose HPGe gamma-spectrometer without the ID software.

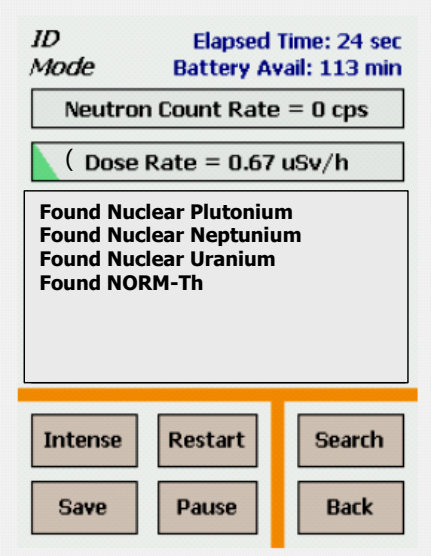

Figure 2 DETECTIVE EX screen

Performance as a RIID

The DETECTIVE has been extensively tested and has been shown to be a high performance identifier. See, for example, refs 4, 5, 6. The key to its performance is

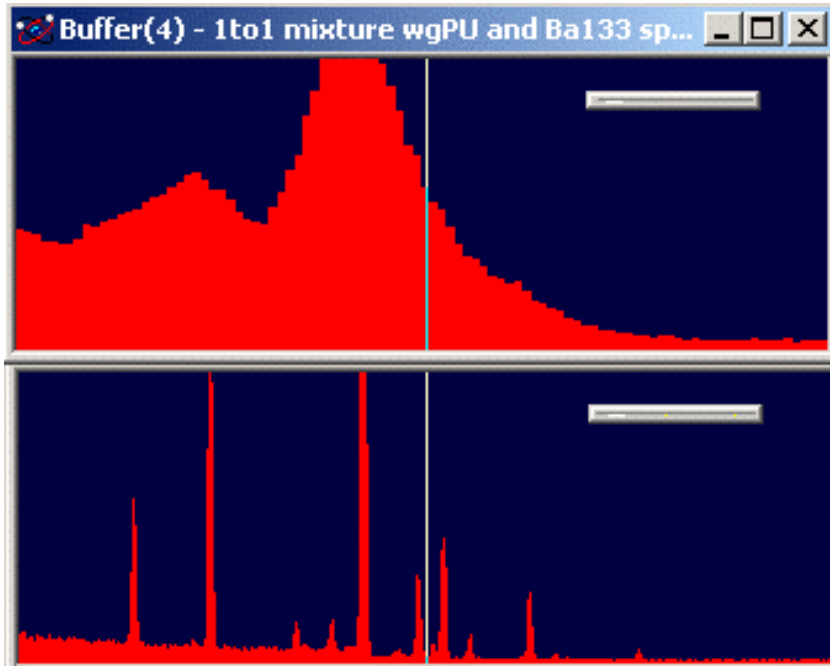

Figure 3 Comparison of Barium and Plutonium Mixture for (upper) NaI Detector, and (lower) DETECTIVE HPGe Detector.

energy resolution, illustrated by figure 3, which clearly shows that HPGe can "see" nuclides which are not seen by NaI systems. It is also up to 100 times faster to ID than comparable NaI identifiers.

Performance with PC/FRAM

PC/FRAM[*, 7, 8], the Isotopic ratio code from Los Alamos, has recently[9] been shown to be capable of obtaining good results using HPGe detectors with a range of resolutions. Even with a resolution of ~1.8 keV at 122 keV, good results may be obtained. In order to assess the possibilities of use of the DETECTIVE with PC/FRAM, DETECTIVE serial number 258 was tested at Los Alamos.

Resolution

Energy resolution was tested using gamma rays from three sources: 122 keV peak from ^{57}Co, 1,332 keV peak from ^{60}Co, and 2,615 keV peak from ^{208}Tl. Table 1 shows the FWHM and FWTM/FWHM results. These compare favorably with the manufacturer's warranted trans-SPEC resolution values.

Table 1. Measured resolutions of the DETECTIVE system.

Energy (keV)	Input rate (kHz)	FWHM (keV)	FWTM/ FWHM
122	1	1.29	1.88
	30	1.30	1.92
1,332	1	2.05	1.85
	30	2.07	1.86
2,615	20	2.72	1.90

Isotopic Analysis

FRAM normally analyzes plutonium and uranium data taken with a coaxial germanium detector that has been energy calibrated at 0.125 keV/Ch and has a resolution of about 1 keV or better at 122 keV. In this case the energy resolution was somewhat greater than this, and the energy per channel was about three times larger than is usually employed, at 0.3664 keV/Ch. An important task of this evaluation was, therefore, to see what quality of isotopic information could be reasonably obtained from plutonium and uranium measurements with this system.

The system was used to measure four PIDIE sources with a wide range of fractional plutonium content from 64.2 to 93.9% ^{239}Pu, five LEU samples of the NBS-SRM 969 set and three HEU samples of the NBL-CRM 146 set with a wide range of fractional uranium content from 0.3 to 93.2% ^{235}U. A 1.2 mm thick cadmium absorber was used to reduce the intensity of the low-energy gamma rays for all the measurements. The acquisition time for each spectrum was 15 minutes of live time. Sixteen spectra were obtained for each data set.

The FRAM software analysis settings are controlled by "parameter sets" which can be easily modified for use with different Ge detectors (COAX, LOAX or others). The parameter set normally used for uranium and plutonium analysis of COAX spectra was modified to account for the broader resolution and larger energy calibration prior to use.

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[*] **F**ixed-energy **R**esponse-function **A**nalysis with **M**ultiple Efficiency

Table 2 shows the uranium results. The bias values are the ratios of the average measured values to those of the accepted values. The %Err is the average percent error of the 16 runs. The %RSD is the relative standard deviation of the results of the 16 runs.

Table 2. Uranium results.

%^{235}U		^{234}U	^{235}U	^{238}U	%^{235}U		^{234}U	^{235}U	^{238}U
0.3	Bias	1.482	1.027	1.000	4.5	Bias	0.934	0.968	1.001
	%Err	74.5	3.4	0.0		%Err	13.3	1.5	0.1
	%RSD	92.7	7.1	0.0		%RSD	8.1	4.3	0.2
0.7	Bias	0.942	1.019	1.000	20.1	Bias	1.016	1.025	0.995
	%Err	63.2	2.2	0.0		%Err	5.8	1.4	0.4
	%RSD	53.3	4.9	0.0		%RSD	5.0	2.1	0.6
1.9	Bias	0.990	0.980	1.000	52.6	Bias	1.017	1.040	0.955
	%Err	23.9	1.7	0.0		%Err	6.2	2.0	2.4
	%RSD	20.6	6.5	0.1		%RSD	7.6	5.0	6.2
2.9	Bias	0.955	0.993	1.000	93.2	Bias	0.969	0.994	1.093
	%Err	15.8	1.6	0.0		%Err	4.1	0.4	6.6
	%RSD	18.4	5.6	0.2		%RSD	3.5	0.4	6.9
Calc % Bias		**1.51**	**0.90**	**Var**	**Calc % Bias**		**1.51**	**0.90**	**Var**

Most of the bias results for the ^{234}U, ^{235}U, and ^{238}U samples appear to be reasonable. The result for ^{234}U at 0.3% 235U enrichment is about 50% off due to the very weak and only peak of ^{234}U at 121 keV which has an abundance of only 0.03%. The %RSD values for ^{235}U and ^{238}U are about twice the values of the %Err values. This means that FRAM underestimates the random errors of these two isotopes. This is to be expected, because there is no overlap of the peaks of ^{235}U and ^{238}U used for the determination of the relative efficiency curve. Also, a large gap exists between the two sets of peaks from the two isotopes. These conditions tend to increase the magnitude of the random error beyond the part corresponding to statistics of the peaks.

The last row in Table 2 shows the estimated percent biases for the data. These percent biases for ^{234}U and ^{235}U were calculated using the formulas in Reference 9 with the 186 keV peak, a FWHM of 1.4 keV and a tail of 2%. The estimated bias for ^{238}U is determined from the estimated bias of ^{235}U through the relationship (^{238}U bias) = (^{235}U bias) * (^{235}U/^{238}U). These estimated biases represent the systematic errors of the measurements.

The total biases of the measurements agree reasonably well when both the random errors and systematic errors are taken into account. (The random error for each result is 0.25 of the %RSD shown in Table 2).

Table 3 shows plutonium results analyzed with a parameter set for the peaks in the 120 to 460 keV region. The bias values are the ratios of the average measured values to those of the accepted values. The %Err is the average percent error of the 16 runs. The %RSD is

the relative standard deviation of the results of the 16 runs. The measured values for ^{242}Pu are from the correlation calculations.

Table 3. Results of plutonium spectra analyzed in the 120–460-keV region.

%^{239}Pu		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
93.9	Bias	0.917	0.996	1.060	1.005	1.069	0.976
	%Err	14.4	0.2	3.2	0.6	7.6	0.9
	%RSD	14.2	0.3	3.8	1.3	7.9	1.4
85.1	Bias	0.984	0.995	1.030	1.010	1.030	0.974
	%Err	4.5	0.3	1.9	0.5	2.8	0.6
	%RSD	4.2	0.5	2.6	0.7	3.1	0.9
76.8	Bias	0.986	0.989	1.033	1.025	1.136	0.973
	%Err	2.6	0.5	1.7	0.6	2.1	0.7
	%RSD	2.3	0.6	2.0	0.7	2.9	0.8
64.5	Bias	1.019	1.026	0.949	1.020	0.918	0.952
	%Err	1.6	1.1	3.3	1.2	3.5	1.3
	%RSD	1.5	1.4	2.9	1.4	4.2	1.3
Calc % Bias		**5.67**	**0.49**	**2.54**	**3.03**	**NA**	**3.26**

The ^{242}Pu results in Table 3 were obtained by correlation. The %RSD values appear to be about the same as the %Err values for most isotopes.

Most of the bias results, including those for ^{242}Pu, are reasonable. The results for ^{240}Pu are slightly outside the combined random and systematic errors. This is due mainly to the energy calibration of 0.3664 keV/Ch of the DETECTIVE. This coarse energy calibration does not significantly affect the results other than for ^{240}Pu, because they all have good peaks that can easily be measured regardless of the energy calibration. However, ^{240}Pu has only one weak peak at 160 keV. There are several other peaks nearby and they interfere with the determination of the peak area of the 160 keV peak of ^{240}Pu. The coarse energy calibration of the DETECTIVE makes it harder to accurately extract the 160 keV peak area, which then makes the bias larger than should be.

Table 4 shows plutonium results analyzed with a parameter set for the peaks in the 200 to 1,010 keV region. The bias values are the ratios of the average measured values to those of the accepted values. The %Err is the average percent error of the 16 runs. The %RSD is the relative standard deviation of the results of the 16 runs. The measured values for ^{242}Pu are from the correlation calculations.

Table 4. Results of plutonium spectra analyzed in the 200–1,010-keV region.

%^{239}Pu		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
93.9	Bias	0.693	1.005	0.932	0.979	0.814	0.984
	%Err	42.7	0.6	10.5	1.9	28.9	1.5
	%RSD	45.2	0.6	10.4	9.5	29.5	1.5
85.1	Bias	0.888	1.008	0.956	1.010	0.901	0.985
	%Err	13.9	1.0	6.2	1.1	8.9	1.1
	%RSD	20.9	1.5	9.7	1.5	12.1	1.7
76.8	Bias	0.930	0.998	1.005	1.006	1.065	0.970
	%Err	10.1	1.7	6.1	1.7	7.7	1.8
	%RSD	12.0	2.1	7.2	2.1	11.1	2.2
64.5	Bias	1.011	0.996	1.023	0.908	0.984	0.844
	%Err	7.5	4.2	11.4	4.3	12.2	4.3
	%RSD	9.5	5.1	10.8	6.7	12.0	5.5
Calc % Bias		**5.41**	**0.39**	**2.66**	**2.07**	**NA**	**1.57**

The ^{242}Pu results in the Table were obtained by correlation. . The %RSD values appear to be about the same as the %Err values for most isotopes. Most of the bias results appear to be reasonable.

Conclusion and Possible future instrument Development

It is clear from previous results and those presented here that the DETECTIVE or a derivative may be configured as a combined instrument which will identify the nuclides present and, where SNM is detected, will provide isotopic ratios in a short time which are accurate to a few percent. A variant of the FRAM analysis Engine could be made to run on the PDA used by the DETECTIVE-EX to provide a self-contained instrument with the following performance benefits:

- Continuously available, RUGGED and portable high resolution, high efficiency gamma ray spectrometer.
- Long operational lifetime (50,000 continuous operation design life)
- “short-cycling” of Ge detector
- Battery operated. No liquid nitrogen required.
- Neutron detection optionally available.
- Advanced in situ analysis:

  Simple user interface providing sophisticated nuclide identification via LLNL algorithms.

  INTEGRATED functionality of PC/FRAM allowing found fissile material to be rapidly and accurately analyzed for isotopic composition.

- Removable data storage
- Wired or wireless data transfer and remote control if required.

This instrument could expand the situations in which high purity germanium detectors may be employed in the detection of undeclared activities. An instrument can be continuously available in a vehicle or building. The only "supply" needed is electricity.

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