A New, Room-Temperature Gamma-Ray Detector for Improved Assay of Plutonium[*]

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Abstract

Gamma-ray spectroscopy for portable and unattended assay of nuclear materials requires rugged, reliable, room-temperature detectors that are stable in variable environments and detect gamma rays with good efficiency and energy resolution. For portable assays especially, compact detectors address needs for large numbers of measurements performed in rapid succession with heavy shielding and collimation by a user who must carry the spectroscopy equipment. Most measurements are made with compact NaI detectors. The assay of variable-burnup plutonium and other plutonium materials of variable isotopic composition challenges low-resolution gamma-ray spectroscopy in numerous safeguards applications including holdup measurements, safeguards inspections, monitoring, and safeguards close-out in decontamination and decommissioning. A new, commercial-prototype coplanar-grid CdZnTe detector has been evaluated using the assay of variable-burnup plutonium as a metric indicator to show the substantial benefit of its improved performance compared to results of the same measurements performed with the compact NaI detector. Detector performance, spectrum-quality, and assay results as well as gamma-ray spectra of reference sources are compared for the coplanar-grid CdZnTe and compact NaI detectors to illustrate the advantages of the new room-temperature gamma-ray detector. Isotope identification with the coplanar-grid CdZnTe detector is demonstrated. Preliminary calculations (Monte Carlo coupled to simulations of radiation transport and charge collection) of the spectral response of the detector to plutonium indicate promise for the use of the coplanar-grid CdZnTe detector for further improvements in the accuracy of assays and for analysis of gamma-ray isotopic distributions.

I.   Introduction

Room-temperature scintillator/photomultiplier detectors are used for gamma-ray spectroscopy in the quantitative nondestructive analysis of nuclear materials by gamma-ray spectroscopy in portable[1-8] and continuous, on-line[9-11] applications to nuclear safeguards. The disadvantages of NaI detectors are often outweighed by compactness and good gamma-ray detection efficiency, which are beneficial in

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portable and on-line applications where small and lightweight instruments are needed to provide spectral data in short count periods. Furthermore, these detectors are obtained from commercial suppliers with reliable performance in sizes and configurations to suit many needs for safeguards. However, their low energy resolution restricts applications and limits measurement sensitivity, and sensitivity is an issue for many portable measurements because signals are small in high backgrounds. Low resolution can also lead to bias in the quantitative analysis results for materials whose gamma-ray spectra are complex, whose isotopic composition varies, or whose chemical composition includes radionuclides that produce interfering gamma rays. Most plutonium-bearing materials for which portable and on-line measurements are required for safeguards, control and accountability of nuclear materials exhibit two and often all three of these characteristics. The magnitude of the bias that arises from the variable isotopic composition alone can approach 100% for the quantitative analysis of variable-burnup plutonium.[8, 12] Moreover, some of the newest miniature, self-contained gamma-ray spectroscopy systems are actually smaller and lighter than the shielded compact NaI detectors. Finally, instability in the electronic gain of the NaI spectrometer that arises primarily from changes in temperature is a serious drawback of gamma-ray spectroscopy performed with scintillators, particularly in portable and unattended applications for which extreme and variable measurement environments are common.

The most recent developments in room-temperature solid-state gamma-ray detectors indicate that the capabilities of compound semiconductor detectors such as CdZnTe with novel electrode designs[13-14] include significant improvements over the energy resolution provided by NaI detectors as well as very compact dimensions. These new detectors are fully compatible with the newest compact gamma-ray spectroscopy systems designed for portable and unattended applications. Their prospects for excellent stability (with changing temperature) and reliability are very good. Recent progress in development of high-quality materials in quantities that meet production needs and in sizes that satisfy expectations for comparable detection efficiency for some NDA applications is very promising.[15] The applications that involve portable gamma-ray spectroscopy are among those that will benefit the most from these new, improved room-temperature detectors. This is partly because relatively small detectors are needed and used currently to satisfy the criterion for hand-held portability. Compact NaI detectors employed for both uranium and plutonium holdup measurements[16], for example, use crystals that are 2.5 cm in diameter and 5.0 cm thick. This is also because demands for improved performance of room-temperature gamma-ray detectors are greatest in portable applications of gamma-ray spectroscopy of materials *in-situ*. These demands include the ability of the detector to address a

- variety of isotopic mixtures, chemical compositions, radiological properties and geometric distributions;
- wide range of assay quantities; and
- variety of environmental conditions including mechanical influences and constraints, radiological backgrounds and large changes in temperature that characterize *in-situ* measurement needs for large processing facilities.

We have recently obtained and tested a commercial prototype coplanar-grid CdZnTe detector.[17] The tests have included determination of performance characteristics, comparison of the performance with that of the compact NaI detectors, and an extended evaluation of performance and reliability in a three-month period. They have also included evaluation of the capability for quantitative analysis

of ^{239}Pu in isotopic mixtures of variable burnup and a direct comparison to the same but less accurate capability with the compact NaI detectors[16, 18] that are now used for such measurements. The capability for isotope identification with the new detector has been demonstrated. Finally, the potential for further significant improvements in accuracy as well as extended applications of these detectors to determine the isotopic composition of variable-burnup plutonium is discussed in terms of a new capability that uniquely exploits the improved performance of these new detectors.[19, 20]

II. Detectors
A. Coplanar-Grid CdZnTe Detector

The coplanar-grid CdZnTe detector that was used in these experiments is among the first, preproduction units to be manufactured commercially.[18] The dimensions of the rectangular crystal are 1 cm by 1 cm by 0.5 cm thick. The crystal, two preamplifier boards, and the analog summing circuit are mounted inside a rectangular aluminum box with a removable lid designed for ease of replacement of the crystal for evaluation purposes. Figure 1 is a photograph of the box with the lid removed showing the crystal and other internal components. The side view shown at the left in Fig. 2 is a sketch of Fig. 1 with the crystal, preamplifier boards and the analog summing circuit labeled.
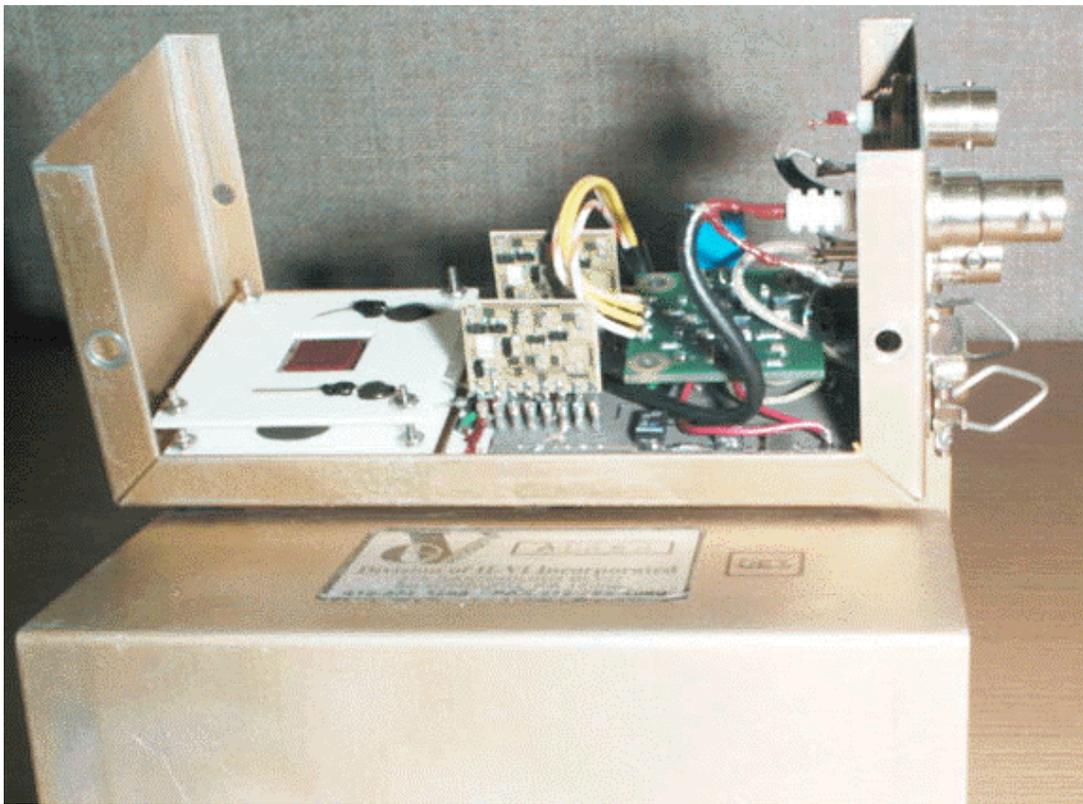
Fig. 1. Photograph of the rectangular box with the lid removed showing the crystal and other internal components of the prototype coplanar-grid CdZnTe detector. (Refer to Fig. 2 for labeling of the individual components.) The convenience of the rectangular box prototype design is in the ability to exchange components or modify the design, needs that can arise in an evaluation period, with relative ease.

SIDE VIEW

FRONT VIEW

1 OF 2
PREAMPLIFIERS

ANALOG
SUMMING CIRCUIT

COPLANAR GRID
CdZnTe
CRYSTAL

HV

DIFF

OUTPUT

LOW VOLTAGE

0.3cm
TIN

1.4cm

AMERICIUM-241
REFERENCE
SOURCE

PLUTONIUM
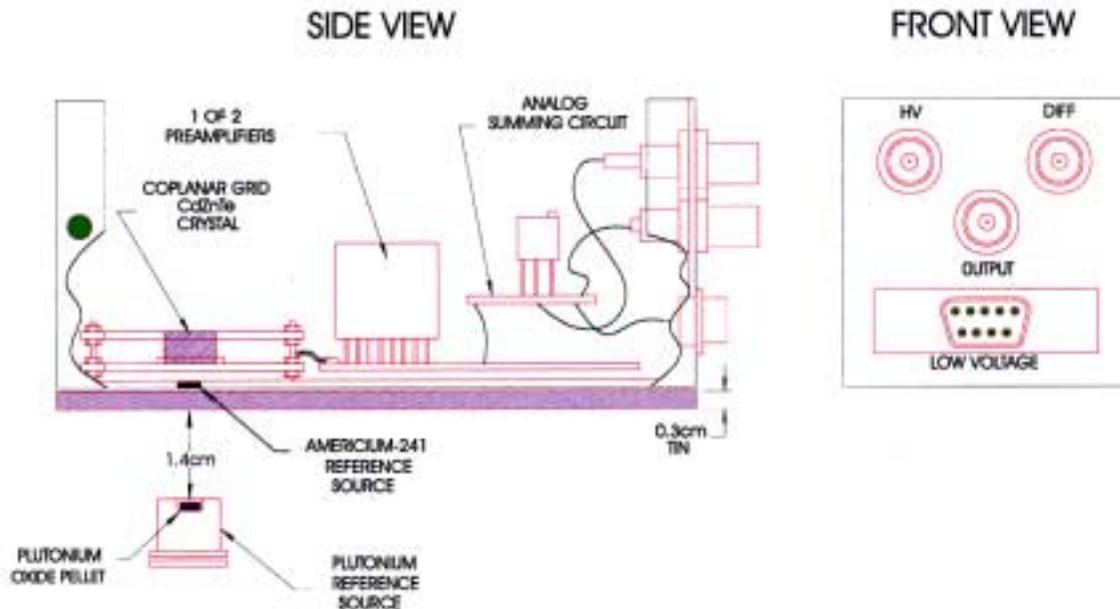OXIDE PELLET

PLUTONIUM
REFERENCE
SOURCE

Fig. 2. Labeled sketch of the prototype coplanar-grid CdZnTe detector. The side view is the same view shown in the Fig. 1 photograph. The experimental positions of the ^{241}Am reference source, the encapsulated plutonium reference sample and the tin absorber are also indicated.

Charged species (particles and holes) are produced in a solid-state crystal by interactions between photons and the electric fields of the solid-state lattice. Migration of the particles (electrons) and holes in an external electric field created by a potential difference across the crystal (detector bias) allows collection of charged species at the bias electrode to create an analog pulse that can be amplified and analyzed. The properties of the electronic states of the solids and characteristics of the production of electron-hole pairs in compound semiconductor materials (CdTe and CdZnTe, among others) have been candidates for intermediate resolution room-temperature gamma-ray spectroscopy for many years. However, because of effects that are dominated by the large differences in transport properties of electrons and holes in compound semiconductors, analog pulses collected with a simple planar electrode configuration have characteristics that vary widely, depending on the location of photon interaction within the crystal relative to the collection electrode. This translates to a degradation in the energy resolution of gamma-ray spectrometers that use simple integration and pulse height analysis, the conventional state of the art for field instrumentation. Because of the greater penetrability of higher energy photons whose interactions are more distributed throughout the crystal volume, the effect that appears as a plateau on the low-energy side of each peak in the digitized pulse height spectrum makes these simple planar-electrode detectors unusable for quantitative analysis. The coplanar grid[13, 14] is the breakthrough concept for the compound semiconductor detectors that addresses the need to produce a signal that reflects only the collection of electrons. The coplanar-grid electrode design includes two independent electrodes in an interwoven grid pattern on a common plane of the crystal. The detector is biased such that the electrons are collected on the coplanar-grid surface. Because of a small (relative to the bias voltage) potential difference between the two coplanar electrodes, the electrons are collected by the grid electrode that is at the higher potential, while the effects of hole migration (away from the surface of

the coplanar electrodes) induces charge on both grid electrodes. The output signal from the coplanar-grid CdZnTe detector is a simple linear combination of analog signals from the two preamplifiers of the coplanar electrodes that eliminates the induced effect of migration of holes by subtracting it from the signal that includes the effects of both holes (induced) and electrons (collected). This output signal is processed with conventional gamma-ray spectroscopy electronics, including the newest and most compact, self-contained systems designed for portable applications.

This prototype coplanar-grid CdZnTe detector uses a grid design that consists of sixteen strips (eight per electrode), each with a width and spacing of 0.025 cm and length of 0.8 cm, in a square array. The detector operates with a negative bias (nominally, -600 V) applied to the 1-cm^2 surface opposite the grid electrodes (bottom surface of the crystal shown in Fig. 2) with +30 V applied to the collecting grid electrode relative to ground on the non-collecting electrode (differential bias). A variable low-voltage power supply was used for the differential bias to observe the performance as a function of the differential bias. Otherwise, a small battery would serve this purpose. Aside from the need for the differential bias, the electronics requirements for operation of the coplanar-grid CdZnTe detector are the same as those for the compact NaI detector.

B. Compact NaI Detector

The integral compact NaI detector[16, 18] includes a cylindrical gamma-ray collimator and shield, which also serves to keep light from reaching the photomultiplier tube. The crystal that is used for portable measurements of both uranium and plutonium is 2.5 cm in diameter and 5.0 cm thick. The dimensions of the photomultiplier tube are comparable to the crystal dimensions. The overall length and diameter of the collimated, shielded package are 22 and 5 cm, respectively, with a weight of 2.5 kg. The compact NaI detector has been a reliable component for portable gamma-ray spectroscopy needs for its ready commercial availability (and maintainability) and consistent performance. This detector, along with commercial, self-contained, portable gamma-ray spectroscopy systems, has been used for over ten years for quantitative measurements of uranium and plutonium *in-situ* as holdup and in-process inventory in domestic and international facilities.[2-11] Figure 3 is a photograph that includes the compact NaI detector, the prototype coplanar-grid CdZnTe detector and a self-contained, miniature, portable gamma-ray spectroscopy system.

III. Experimental Procedures and Materials
A. Experimental Setup

The same standard commercial electronics were used to obtain gamma-ray spectra with the coplanar-grid CdZnTe and the compact NaI detectors. The electronics included a commercial combination linear amplifier, analog-to-digital converter and bias supply (Canberra model 1510) and a personal-computer-based multichannel analyzer (Canberra System 100). The optimum amplifier shaping time of 1 μs for the coplanar-grid CdZnTe detector and a bias of -600 V was used to obtain all of the data presented. (The NaI data were obtained with a shaping time and bias of 1-μs and 500 V, respectively.) For both detectors, The gamma-ray spectra were digitized into 1024 channels with the gain set for 1 keV per channel. All gamma-ray spectra were stored for subsequent analysis.

Fig. 3. Photograph of the prototype coplanar-grid CdZnTe detectors in the rectangular box (left) and the cylindrical package (right, foreground), the shielded compact NaI detector (right, background) and a self-contained, miniature, portable gamma-ray spectroscopy system (center, background).

The experimental setup used to acquire plutonium gamma-ray spectra with the coplanar-grid CdZnTe detector (a comparable setup was used for the NaI detector as well) is illustrated in Fig. 2. The drawing shows the plutonium sample positioned 3 cm beneath the crystal. This constant configuration was maintained by apparatus (not illustrated) that held both the detector and the holder for the plutonium sample in a fixed relative geometry. A 3-mm thick layer of tin was positioned between the plutonium sample and the detector as shown in Fig. 2 to eliminate the variable and dominant effects of the ^{241}Am 60-keV gamma-rays from the spectra. The presence of this filter also enables the use of an ^{241}Am reference source as illustrated in Fig. 2. This experimental configuration described above was maintained throughout the two-month period in which the plutonium samples were measured. The activity of the ^{241}Am reference source is a very small fraction of the ^{241}Am activity in the plutonium reference samples.

B. Quality Assurance and Quality Control Procedures

For assurance and control of the quality of gamma-ray spectra and of the quantitative results obtained from spectral analysis, reference peaks are commonly created or identified and monitored. A reference peak is a peak in the gamma-ray spectrum whose characteristics are known. Comparison of each known characteristic with the result obtained experimentally from each unique gamma-ray spectrum that is acquired is the basis for assurance and control of performance (gain and resolution for each spectrum, for example), reliability (accuracy of the quantitative analysis), and stability (precision). The performance is tested by determining the full-width at one-half and one-tenth maximum (FWHM and FWTM, respectively) and the centroid of a peak that appears in all spectra with sufficient intensity that it can be characterized adequately, independent of other activities, for even the shortest count periods.

The 60-keV gamma-ray of the ^{241}Am reference source was used as an internal (present in all gamma-ray spectra) low-energy reference peak whose net count rate, centroid and FWHM were used to monitor reliability and performance as well as stability on a spectrum-by-spectrum basis for the extent of the experimental period. The activity of the internal reference source of ^{241}Am was

2 µCi. The centroid and FWHM of the prominent 208-keV gamma ray of ^{237}U (daughter of ^{241}Pu) were used to monitor performance and stability internally at the higher energy. The external reference source was ^{137}Cs, whose net count rate, centroid, and FWHM were used to monitor performance and stability at 662 keV before and after each set of plutonium gamma-ray spectra were acquired (externally) in a day. The activity of the external reference source of ^{137}Cs was 7 µCi.

There was no experimental knowledge of the stability of the prototype coplanar-grid CdZnTe at the start of these measurements. The use of a digital stabilizer can degrade the performance for stable operation, and there was interest in characterizing and quantifying any instabilities in performance. Therefore, the gamma-ray spectra used to evaluate the detector for quantitative analysis of variable-burnup plutonium were obtained in many short count periods so that performance and reliability could be monitored for stability over the extended time and under the variable conditions of the data acquisition period, and so that digital compensation for gain drift could be employed if necessary to optimize the quality of the combined data.

C. Plutonium Reference Samples

Gamma-ray spectra were acquired for each of seven small reference samples of plutonium oxide of the same chemical composition, plutonium mass (0.4 g), geometric configuration, and encapsulation but with differing isotopic composition from low- to high-burnup. Table I gives the isotopic composition of the reference samples when the measurements were performed. These are aged (since chemical separation) samples, as indicated by the ^{241}Am fractions at the higher burnups. The oxide in pellet form is packed within a small cavity of the machined stainless steel capsules of precise, cylindrical dimensions such that the measurement geometry was the same for all samples and easily reproduced. Because of the uniformity of the encapsulation as well as the small size, uniform dimension, and constant plutonium mass of each oxide sample, these variable-burnup reference materials are well-suited for testing quantitative nondestructive gamma-ray analysis methods because the gamma-ray attenuation effects can be assumed to be the same for the seven samples.

Table I. Isotopic Composition of Plutonium Reference Samples 1-7

Isotope	Isotope Weight Percent* (7-8-96)						
	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6	Sample 7
^{238}Pu	0.01	0.02	0.04	0.10	0.12	0.89	1.21
^{239}Pu	93.86	89.52	84.93	78.30	76.52	67.80	63.69
^{240}Pu	5.97	10.07	14.14	19.82	21.30	24.29	26.18
^{241}Pu	0.13	0.29	0.66	1.22	1.37	3.55	4.39
^{242}Pu	0.03	0.09	0.23	0.56	0.70	3.47	4.53
^{241}Am	0.29	0.41	0.94	0.62	0.69	1.79	5.69

The coplanar-grid CdZnTe detector was used to obtain gamma-ray spectra for all seven plutonium samples individually in a measurement series, with a count time of 600 s for each sample. Each series of seven measurements took place in a single day. Each was preceded and followed by counting of the external reference source of ^{137}Cs and was accompanied by a measurement of the spectrum of the room background. A measurement series took place either in the morning or in the

afternoon during the summer. The room temperatures were typically 20°C in the morning and above 32°C in the afternoon. Ten series of measurements were obtained in this way in a period of two months (with several intervening periods without power) for a total count time of 6000 s per sample. No adjustments were made in the electronics settings for the duration of the data acquisition period except to turn the power off and on.

The compact NaI detector was used to obtain gamma-ray spectra for the same seven plutonium samples individually in a measurement series, with a count time of 600 s for each sample, and a counting geometry approximately equivalent to that used with the coplanar-grid CdZnTe detector. Only one series of seven measurements was used in this case because the systematic behavior of this detector is understood. The order-of-magnitude shorter total count time (600 s as opposed to 6000 s) is justified by a greater (by more than an order of magnitude) detection efficiency at 414 keV for the compact NaI detector compared to the coplanar-grid CdZnTe detector, as indicated in Table II.

Table II. Detector Comparison Data

Detector		Compact NaI	Coplanar-Grid CdZnTe	
			Current	Proposed
122 keV	FWHM %	14.5	8.5	NA
	FWTM %	27.5	19.9	NA
662 keV	FWHM %	8.0	3.4	NA
	FWTM %	14.4	13.0	NA
Relative efficiency*	122 keV	100	19	44
100 **x** e / e$_{NaI}$	414 keV	100	7	23
	662 keV	100	6	21
Crystal: shape		cylindrical	rectangular	rectangular
cross-sectional area (cm^2)		5.1	1.0	2.3
depth (cm)		5.1	0.5	1.0

* including solid angle

IV. Experimental Measurements
A. Settings and Performance of the Coplanar-Grid CdZnTe Detector

The signal from the coplanar-grid CdZnTe detector is a linear combination of analog signals from the preamplifiers on the two coplanar electrodes. A potential difference between these two electrodes distinguishes one of them as the collecting electrode for particles (electrons). The magnitude of the potential difference required for optimum performance is illustrated in Fig. 4a and b, which shows gamma-ray spectra of ^{137}Cs and [57]Co, respectively, obtained at four differential bias settings up to +30 V. The 30-V data for these two sources are shown in Fig. 5.

Gamma-ray spectra of ^{137}Cs and [57]Co obtained with the coplanar-grid CdZnTe detector above the 30-V setting are very similar to the 30-V spectra. Figure 6a shows the centroid of the 662-keV gamma-ray peak of ^{137}Cs plotted vs differential bias settings up to 50 V. Figure 6b and c are the FWHM and FWTM of the 662-keV peak, also plotted vs differential bias settings up to 50 V.

Above +30 V differential bias, the centroid shows little dependence on differential bias, and the FWHM and FWTM remain constant. All subsequent data were acquired with a differential bias of +30 V and -600 V bias.
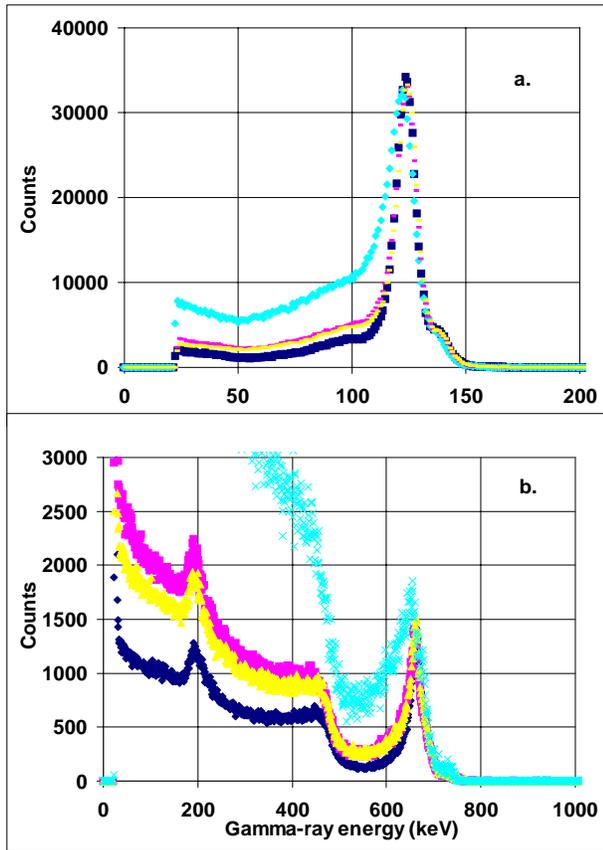

Fig. 4a and b. Overlaid plots of four (a)^{137}Cs and (b) ^{57}Co gamma-ray spectra with 10-volt increments in the differential bias voltages across the coplanar electrodes. The ^{137}Cs and ^{57}Co spectra with the least-resolved peaks were obtained with a near-zero differential bias. Those with the best resolved peaks, obtained with a differential bias of 30 V, are also shown with an adjusted vertical scale in Fig. 5.

Table II gives details on energy resolution, detection efficiency, and crystal dimensions of the prototype coplanar-grid CdZnTe detector and the compact NaI detector. The energy resolution of the coplanar-grid CdZnTe detector at 662 keV is 3.4%, FWHM is improved by more than a factor of 2 compared to the NaI detector. However the FWTM is comparable to that of NaI, a detail that is qualitatively noticeable from the peak shape of the 662-keV gamma ray in Fig. 5. The detection efficiency (including solid angle) relative to the NaI detector, which was measured at 662 and 22 kev, is 6% and 19%, respectively. Therefore, this CdZnTe detector will not be a practical replacement for the compact NaI detector in portable quantitative measurements of the 414-keV gamma ray of ^{239}Pu where its relative detection efficiency is calculated to be 7% of that for the compact NaI detector. The estimated relative efficiency data for a proposed coplanar-grid CdZnTe detector with dimensions of 1.5 cm on each side of the cube are also given in Table II. This detector is a candidate for measurements of ^{239}Pu if its performance with the larger crystal proves to be sufficient.
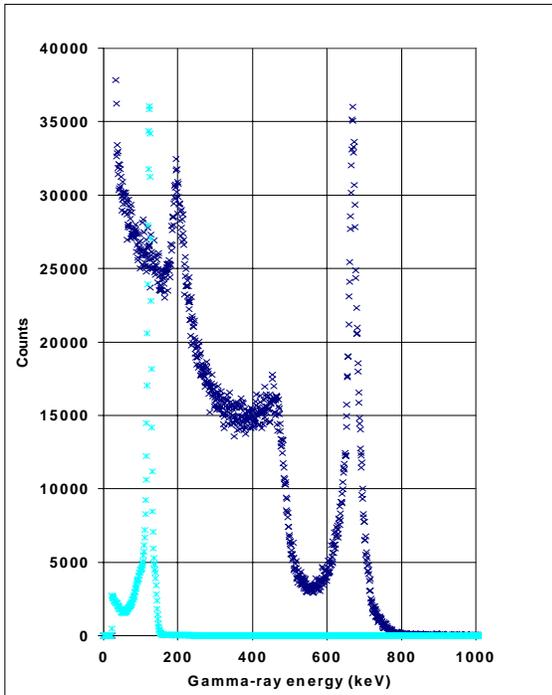
Fig. 5. Overlaid gamma-ray spectra of (a) ^{137}Cs (peak at 662 keV) and (b) ^{57}Co (peak at 122 keV) obtained with the prototype coplanar-grid CdZnTe detector, demonstrating the resolution (FWHM of 8.5% at 122-keV and 3.4% at 662 keV) in the gamma-ray energy range frequently used for portable quantitative analysis of plutonium.

Fig. 6. Performance characteristics of the prototype coplanar-grid CdZnTe detector vs differential bias voltage across the coplanar electrodes. The (a) centroid, (b) FWHM and (c) FWTM are shown for the ^{137}Cs 662-keV gamma-ray peak.
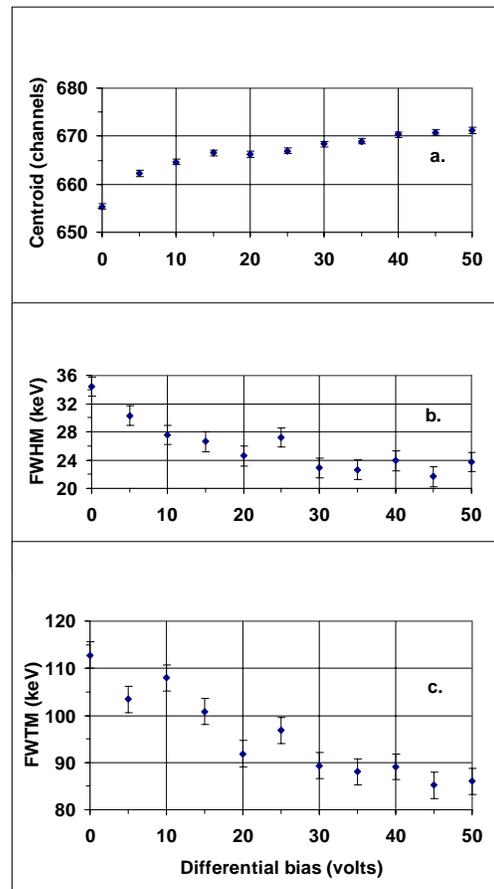
B. Performance with Variable Count Rates

The count rates for measurements of the plutonium reference samples varied from 1700 to 3000 s^{-1} for the lowest to highest burnup sample, respectively, where only 200 to 1500 s^{-1} corresponds to count rates from the plutonium reference samples. The net count rate of the 60-keV gamma-ray from the ^{241}Am internal reference source was used to measure effects such as rate-dependent losses in counts from the plutonium reference samples at the low count rates described above. However, some preliminary measurements were also performed at higher detector count rates to determine if an influence on the stability of the performance could be detected. These measurements used the peak from the 662-keV gamma-ray, with the ^{137}Cs counting geometry fixed, as the reference peak for monitoring the centroid and FWHM with increasing detector count rate. A ^{57}Co source was used to introduce a random source of counts in an energy region below that of the 662-keV reference peak. Figures 7a and b show the centroid and FWHM of the 662-keV peak plotted vs detector count rate, up to 50,000 s^{-1}. There is no evidence of systematic effects that exceed the statistical limits of these measurements in the range of these tests.

C. Energy Calibration

A linear gamma-ray energy calibration was determined for the coplanar-grid CdZnTe detector with the three peaks at 60, 208 and 662 keV, using the ^{241}Am internal reference source, the plutonium reference sample number 5 (21.3 % ^{240}Pu), and the ^{137}Cs external reference source. The plot of gamma-ray energy vs peak centroid and the linear fit to these data are given in Fig. 8. The limited gain setting and sources used for the energy calibration were sufficient for the relatively small energy range of less than 400 keV of these plutonium measurements, but the linearity of the energy calibration over the wider energy range from 60 to 1836 keV is documented separately.[22]

D. Spectrum Quality: Long-Term Performance and Stability

The performance and stability of the coplanar-grid CdZnTe detector gamma-ray spectrometer over the two-month duration of the acquisition of plutonium data was determined using the 662-keV gamma-ray from the external ^{137}Cs reference source. A 300-s count of the ^{137}Cs reference source was made after each of the ten plutonium measurement series. Figure 9a is a plot of the centroid of the 662-keV external reference gamma ray vs plutonium measurement series number. Figure 9b plots the percent FWHM and FWTM of the 662-keV external reference gamma ray vs plutonium measurement series number. The long-term performance data indicate excellent stability, despite the large range in daytime temperature and several extended power shutdowns, with no evidence of systematic effects exceeding short-term random uncertainties during the two-month period of acquisition of the plutonium data.
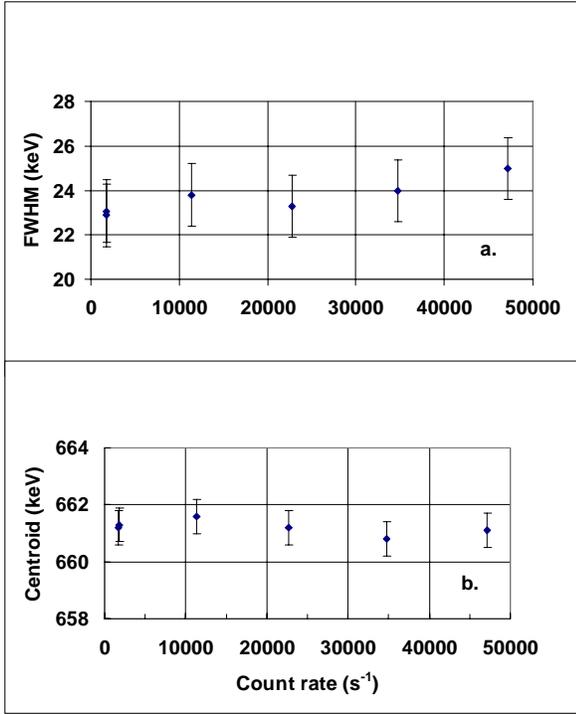
Fig. 7. Performance characteristics of the prototype coplanar-grid CdZnTe detector vs total detector count rate. The (a) relative FWHM and (b) centroid are shown for the 662-keV gamma-ray peak from ^{137}Cs. The ^{137}Cs source rate remained constant for these measurements. The gamma-ray count rate was adjusted using a ^{57}Co source.
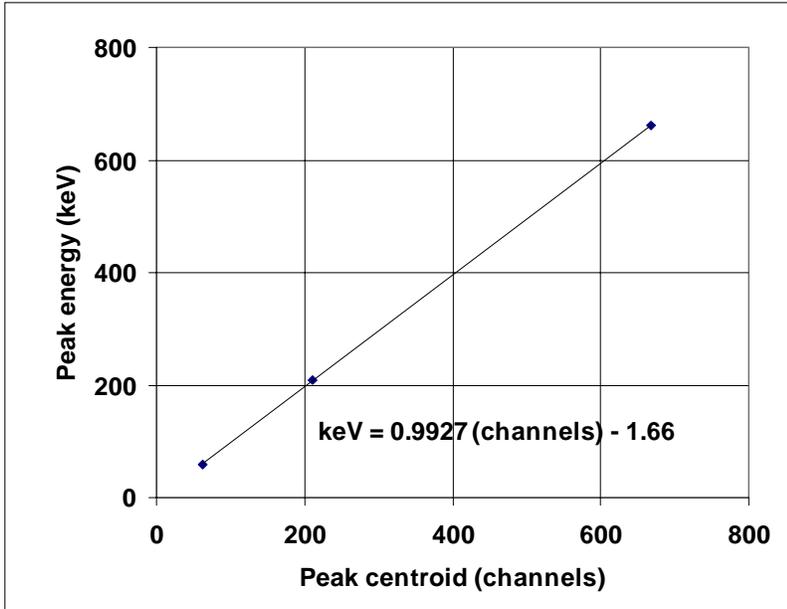

keV = 0.9927 (channels) - 1.66

Fig. 8. Gamma-ray energy for three gamma-ray peaks (at 60, 122 and 662 keV) vs the measured peak centroid channel. The solid line is the linear fit to the data, with the fit equation indicated on the graph.

Fig. 9. The long-term stability of the perform-ance is indicated by the centroid (upper graph) and relative FWHM and FWTM (lower graph) of the 662 keV peak of the ^{137}Cs external reference source plotted vs time (trial #) over the two-month period in which the plutonium data were acquired.
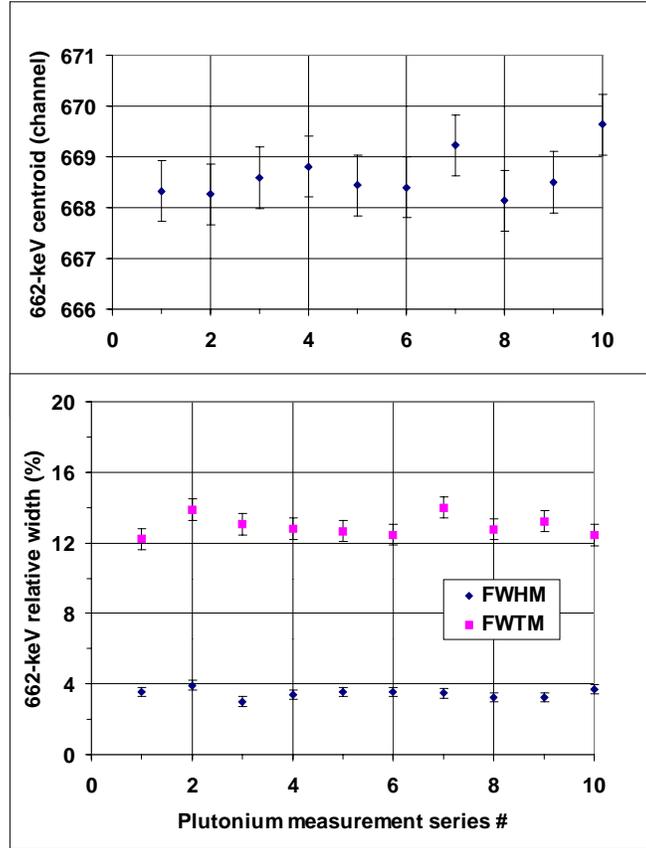
E. Spectrum Quality: Performance, Reliability, and Stability vs Burnup

The immediate spectrum-by-spectrum readout of performance, reliability, and stability of the gamma-ray spectrometer based on the coplanar-grid CdZnTe detector is evaluated using the 60-keV gamma-ray peak from the internal ^{241}Am reference source. The use of the 208-keV gamma-ray from the ^{237}U daughter of ^{241}Pu was also tested as an internal indicator of performance of the gamma-ray spectrometer across the range of burnup of the plutonium reference samples. The internal performance data obtained at 60 and 208 keV for each plutonium reference sample (from the ten 600-s counts on each source performed in the two-month period) also demonstrates this stability of the performance in this time period. Therefore, the ten 600-s gamma-ray spectra for each plutonium reference sample were combined to give one 6000-s gamma-ray spectrum for each plutonium sample. These internal performance data with improved counting statistics were then examined across the range of burnup of the plutonium reference samples.

Figure 10a is a plot of the centroid of the 60-keV gamma ray vs percent of ^{240}Pu (burnup) of the plutonium reference samples. The standard deviation of the data in Fig. 10a for the full range of burnup is consistent within the random uncertainty (~1/3 of the error bar) in each point. Figure10b is a plot of the centroid of the 208-keV gamma ray vs percent of ^{240}Pu (burnup) of the plutonium reference samples. The systematic increase in the centroid with increasing burnup is the result of the influence of the 203-keV gamma-ray of ^{239}Pu whose peak is not resolved from that of the 208-keV gamma-ray. Because the intensity of the 208-keV peak varies from approximately twice that of the 203-keV peak for the lowest burnup plutonium sample to more that 100 times the 208-keV intensity

for the highest burnup sample, the influence of the lower energy gamma-ray on the 208-keV centroid diminishes rapidly with burnup. Therefore, above 14% [240]Pu (where the ratio of intensity of the 208- to the 203-kev peak exceeds 10), the systematic effect on the measured centroid is below 0.25%, 1-σ, and the 208-keV peak centroid is a useful internal reference peak for monitoring performance in plutonium applications. However, even if the lowest-burnup (6% [240]Pu) materials are included, the systematic effect over the full burnup range is below 0.5%,1-σ. Therefore, the 208-keV centroid is a useful reference in many applications in the entire burnup range. Figure 11a shows the centroid data for the two internal reference peaks (the data from Fig. 10) plotted on the same scale.

Figure 11b is a plot of the percent FWHM of the 60- and 208-keV internal reference gamma rays vs percent of [240]Pu (burnup) for the combined (6000-s) gamma-ray spectra of plutonium reference samples. The relative standard deviation (1-σ) of the data in Fig. 11b is <0.1% and 3%, respectively, at the two gamma-ray energies.
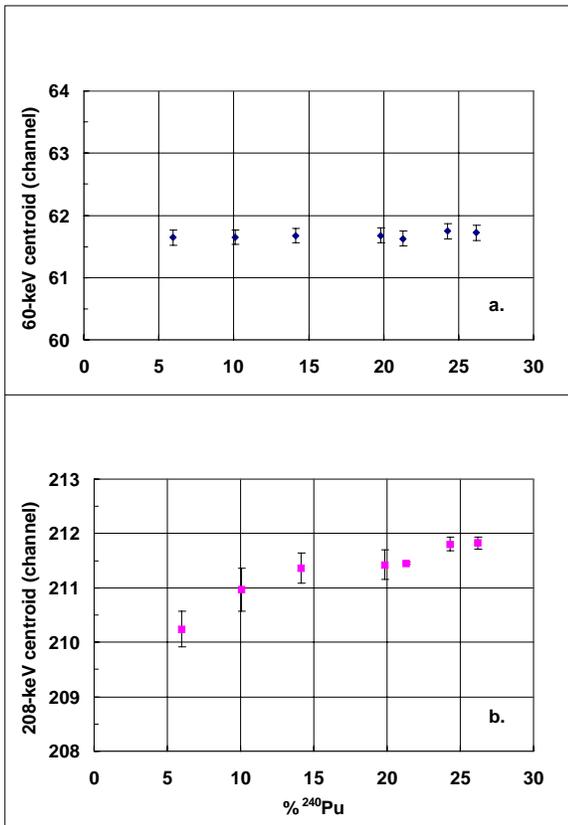

Fig. 10. The reliability of the performance vs burnup (percent of the [240]Pu isotope) is indicated by the mean centroid measured in the ten 600-s counts for the (a) [241]Am 60-keV and (b) [241]Pu/[237]U 208-keV internal reference peaks. The standard deviation of the ten results for each sample, indicated by the error bars, is consistent with the magnitude of the statistical uncertainty for each 600-s count. The systematic effect observed in (b) is caused by the influence of the [239]Pu 203-keV gamma-ray, which decreases with increasing burnup, on the 208-keV centroid. These data are also plotted in Fig. 11a with a different vertical scale.

Figure 12 is a plot of the ratio of the normalization count rate of each plutonium sample to that of the lowest-burnup sample vs burnup. The normalization count rate in this case is the net count rate of the 60-keV gamma ray from the internal reference source. Aside from count-rate losses and radioactive decay of the reference source (for which corrections can be made in both cases), a net rate for the 60-keV peak that is constant with burnup assures the reliability of the spectroscopic data over the range of burnup. The data plotted in Fig. 12 were obtained from the 6000-s (combined)

spectrum for each plutonium reference sample. The magnitude of this ratio decreases monotonically from unity at the lowest burnup to 0.99 at the highest burnup. The ~1% effect is consistent with the expected rate loss in this range of detector count rate (1700 to 3000 s^{-1}) with an amplifier shaping
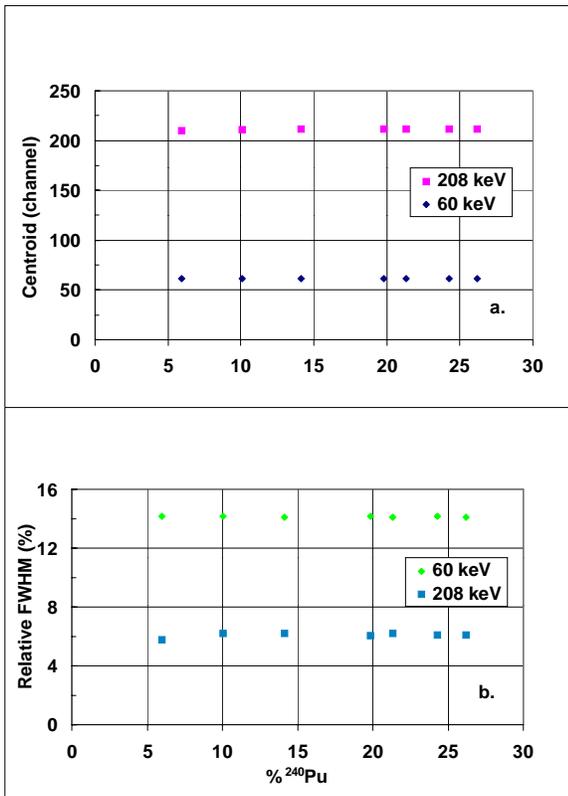

Fig. 11. The reliability of the performance in the range of burnup is indicated by the mean (a) centroid channel and (b) percent FWHM for the internal reference peaks at 60 and 208 keV for the ten 600-s counts of each plutonium reference sample plotted vs burnup (percent of the ^{240}Pu isotope) of the plutonium reference sample.

Fig. 12. The reliability of the performance in the range of burnup is indicated by the mean normalization count rate (count rate of the internal reference peak at 60 keV) for the ten 600-s counts of each plutonium reference sample plotted vs burnup (percent of the ^{240}Pu isotope) of the plutonium reference sample. Each count rate plotted has been divided by the rate for the lowest-burnup plutonium reference sample.
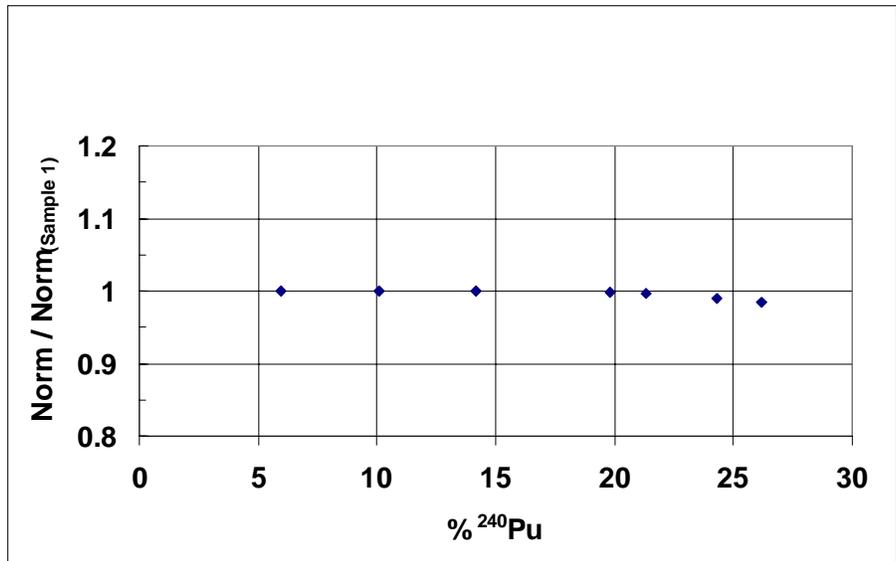
time constant of 1 µs. The absence of systematic effects other than small rate loss effects is confirmed by this ratio, establishes the reliability of the data obtained for the quantitative analysis of plutonium, and illustrates the usefulness of the net area of the 60-keV reference peak for normalization. The simplicity of this approach is appealing for applications of low- and intermediate-resolution detectors to gamma-ray spectroscopy in the portable and unattended mode.

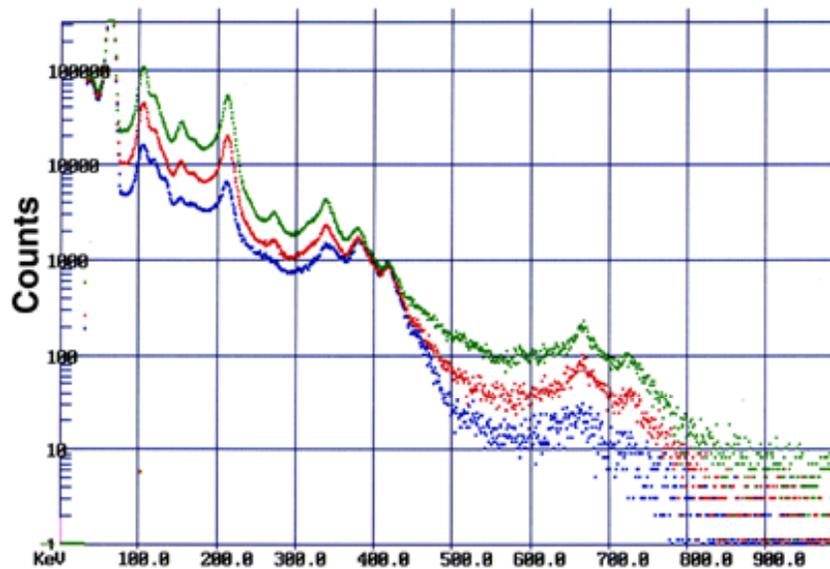## F. Gamma-Ray Spectra of Variable-Burnup Plutonium

No systematic effects over time on performance, reliability and stability of the coplanar-grid CdZnTe detector gamma-ray spectra of plutonium and ^{137}Cs were indicated by the quality tests. Therefore, the ten gamma-ray spectra were combined into a single gamma-ray spectrum of each plutonium reference sample for analysis of the coplanar-grid CdZnTe detector data. Logarithmic plots of these combined (6000-s) spectra from zero to 1000 keV are shown for plutonium reference samples 1, 3, 5 and 7 (Fig.13a) and 2, 4 and 6 (Fig. 13b). The analogous spectra obtained (in 600-s counts) with the compact NaI detector are plotted in Fig. 14 a and b, respectively. The enhancement of spectral detail from the improved resolution of the coplanar-grid CdZnTe detector is apparent from a visual comparison of the spectra obtained with the NaI and CdZnTe detectors. The 60-keV peak from [241]Am is absent from the NaI spectrum because this reference source was not used during the acquisition of NaI data because only one 600-s count was made on each plutonium reference source. The very low activity in this low-energy portion of the spectrum is the result of the 3-mm-thick layer of tin absorber in all measurements. For both the CdZnTe and NaI gamma-ray spectra, the 208-keV gamma-ray used for internal quality assurance is prominent. The apparent absolute intensity of the 414-keV peak that is readily identified in the CdZnTe spectra varies little with burnup, consistent with the fact that the [239]Pu fraction of these samples of equal plutonium mass decreases by only one third from the lowest to the highest burnup. Other important isotopic differences among the spectra for the seven samples are visually apparent in the energy region between 200 and 400 keV. For the lowest-burnup samples, most of the activity between 300 and 450 keV comes from the decay of [239]Pu. As burnup and [240]Pu content increase, a prominent spectral feature at ~333 keV dominates the spectrum above 200 keV. For equal isotope fractions, [241]Pu (in equilibrium with its short-lived [237]U daughter, with a gamma ray at 332 keV) and, to a lesser extent, [241]Am (with three gamma rays at 323, 332, and 335 keV) both contribute to this peak. Furthermore, two gamma rays from [241]Am at 662 keV and 722 keV increase in relative intensity with increasing burnup. Table I shows that the fractions of [241]Pu and [241]Am increase by factors of 34 and 20, respectively, from the lowest to the highest burnup reference sample of plutonium. The effect of these rapidly increasing activities (from [241]Pu and [241]Am) with increasing burnup and age/percent [241]Am is a bias in the 414-keV activity measured with NaI and other low-resolution gamma-ray spectrometers.[8, 12] The bias impacts low-resolution gamma-ray measurements of variable-burnup plutonium in facilities that use or process plutonium fuel, numerous research and test facilities that use higher-burnup, enriched or blended materials, and production facilities in which chemical extraction processes for concentrating plutonium create americium-laden residues.

G. Quantitative Analysis of [239]Pu

The energy region of the 414-keV gamma-ray was used to evaluate the mass of [239]Pu for the seven variable-burnup plutonium reference samples. Figure 15 consists of linear plots of the 200- to 600-keV portions of the overlaid CdZnTe and NaI gamma-ray spectra of samples 1-7. The analysis of NaI gamma-ray spectra for [239]Pu mass has used the energy region from 401 to 450 keV[23] with
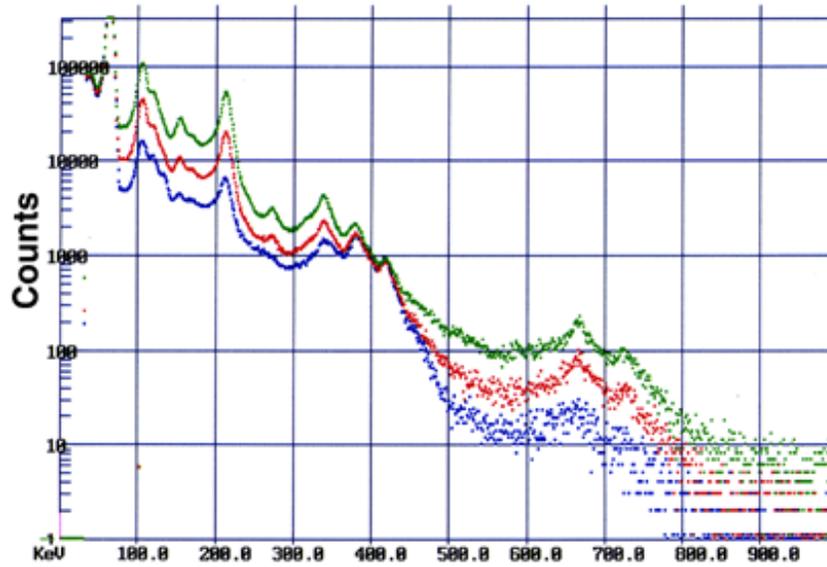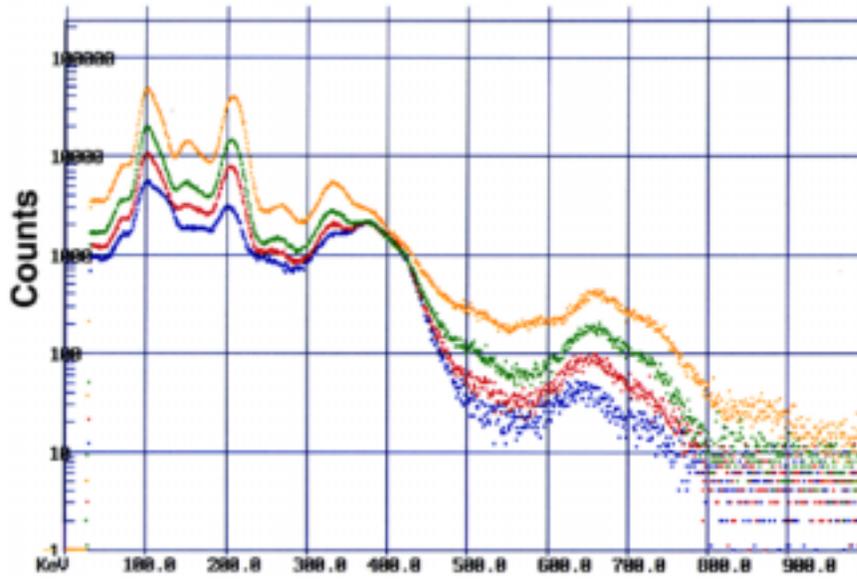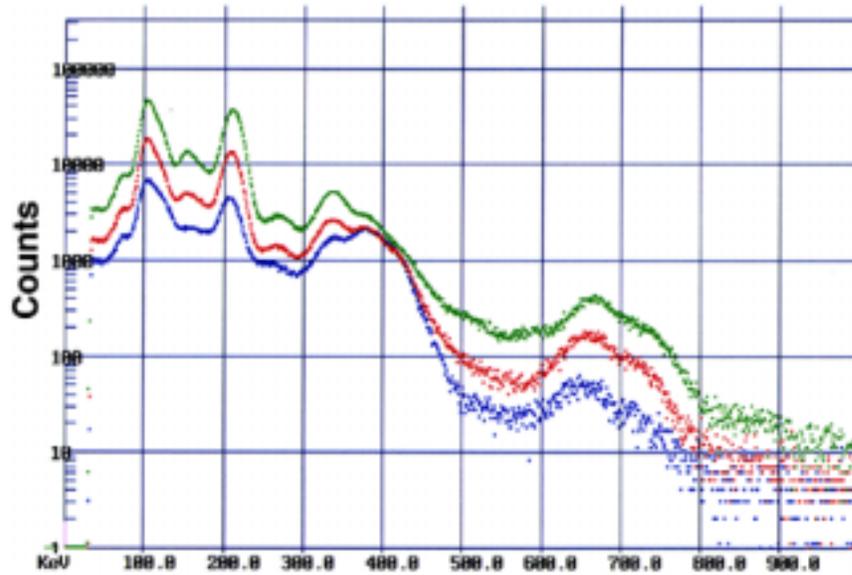
(a)

(b)

Fig. 13. *Logarithmic plot of gamma-ray spectra (counts vs gamma-ray energy) from 0 to 1000 keV obtained with the prototype coplanar-grid CdZnTe detector for the seven variable-burnup plutonium reference samples described in Table I. From bottom to top (increasing burnup), plutonium reference samples 1, 3, 5 and 7 (6%, 14%, 21%, and 26% ^{240}Pu) are plotted in (a) and 2, 4, and 6 (10%, 20%, and 24% ^{240}Pu) in (b). Each 100-min spectrum is the sum of ten spectra obtained in 10-min counts.*

(a)

(b)

Fig. 14. Logarithmic plot of gamma-ray spectra (counts vs gamma-ray energy) from 0 to 1000 keV obtained with the compact NaI detector for the seven variable-burnup plutonium reference samples described in Table 1. From bottom to top (increasing burnup), plutonium reference samples 1,3,5 and 7 (6%, 14%, 21%, and 26% ^{240}Pu) are plotted in (a) and 2, 4, and 6 (10%, 20%, and 24% ^{240}Pu) in (b). Each 100-min spectrum is the sum of ten spectra obtained in 10-min counts.
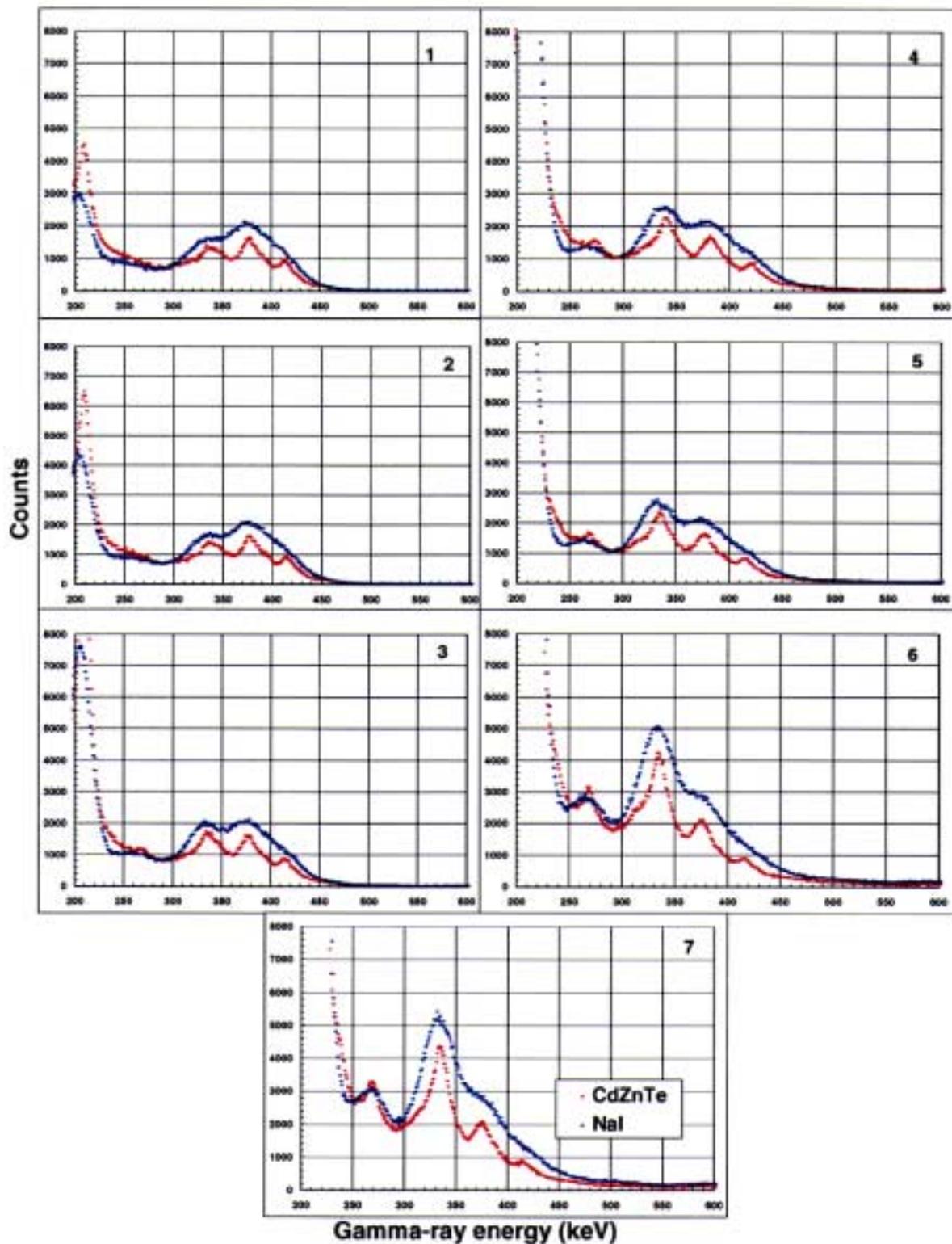
Fig. 15. Linear plots of counts vs gamma-ray energy (in the 200- to 600-keV energy region used for the quantitative analysis) of the CdZnTe gamma-ray spectra (lower plot) overlaid with the analogous NaI spectra (upper plot) that were obtained under identical circumstances. The frames numbered 1 through 7 (in order of increasing burnup or ^{240}Pu fraction) correspond to plutonium reference samples 1 through 7 in Table I.

continuum background obtained from 501 to 550 keV. The current analysis uses these energy regions for both the CdZnTe and NaI gamma-ray spectra, but also uses new regions of interest for the peak (401 to 425 keV) and background (428 to 477 keV) to take advantage of the improved resolution of the CdZnTe detector and to illustrate the magnitude of reduction in bias with improved energy resolution. For the NaI gamma-ray spectra of plutonium, this energy region sits on the high-energy tail of the broad peaked region from 300 to 450 keV. For CdZnTe, it brackets the highest peak of the same broad energy region. (Refer to Fig. 15.). It also sits on the Compton continuum from [241]Am gamma rays at 662 and 722 keV.

The specific response for each plutonium sample is normalized to the result for the lowest-burnup sample. In Fig. 16, the normalized specific response for the (a) NaI and (b) CdZnTe detector obtained using the old (401 to 450 keV) peak energy region is plotted vs percent [240]Pu. Figure 17a and b shows the same results obtained using the new (401 to 425 keV) peak energy region. All results show a bias that increases with percent [240]Pu, with the steepest increase occurring after sample 5 and corresponding to a tripling of the [241]Pu content. For common analysis regions, the range of the bias in the NaI assay of [239]Pu is nearly twice that for the CdZnTe detector (66% vs 40% for the old energy regions and 32% vs 17% for the new regions). Evidence that the differences are partly from contributions of unresolved low-energy contaminants that increase with worse resolution comes from the increase in the bias range of the assay of [239]Pu if the assay region is extended to lower energies. Evidence of the contribution of the 662-keV Compton continuum to the bias comes from the 50% reduction in bias between the old (501 to 550 keV) and new (428 to 477 keV) background regions.
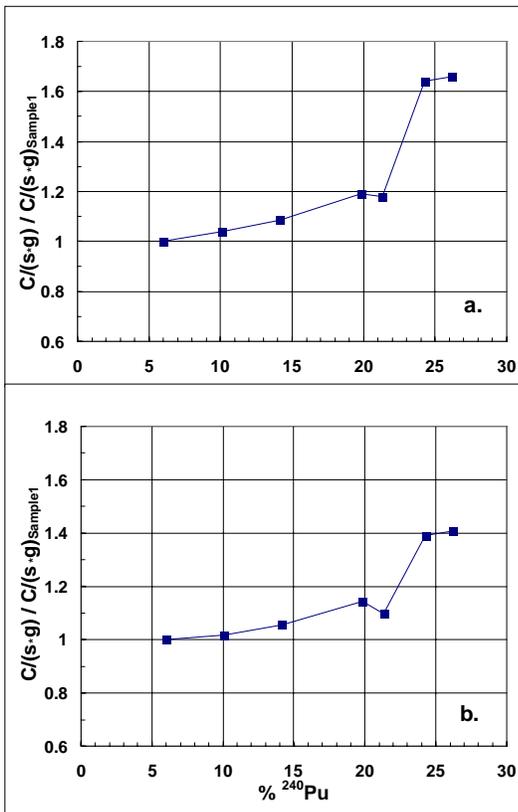

Fig. 16. Net specific response (counts•[s⁻¹g⁻¹]) normalized to reference sample number 1 containing the lowest [240]Pu content in the [239]Pu assay region of interest for the (a) NaI and (b) coplanar-grid CdZnTe gamma-ray spectra of the seven variable-burnup plutonium reference samples, vs burnup (percent [240]Pu). The [239]Pu assay region of interest is 375 to 450 keV.

Fig. 17. Net specific response (counts•[$s^{-1}g^{-1}$]) normalized to reference sample number 1 containing the lowest ^{240}Pu content in the ^{239}Pu assay region of interest for the (a) NaI and (b) coplanar-grid CdZnTe gamma-ray spectra of the seven, variable-burnup plutonium reference samples vs burnup (percent ^{240}Pu). The ^{239}Pu assay region of interest is 400 to 450 keV.

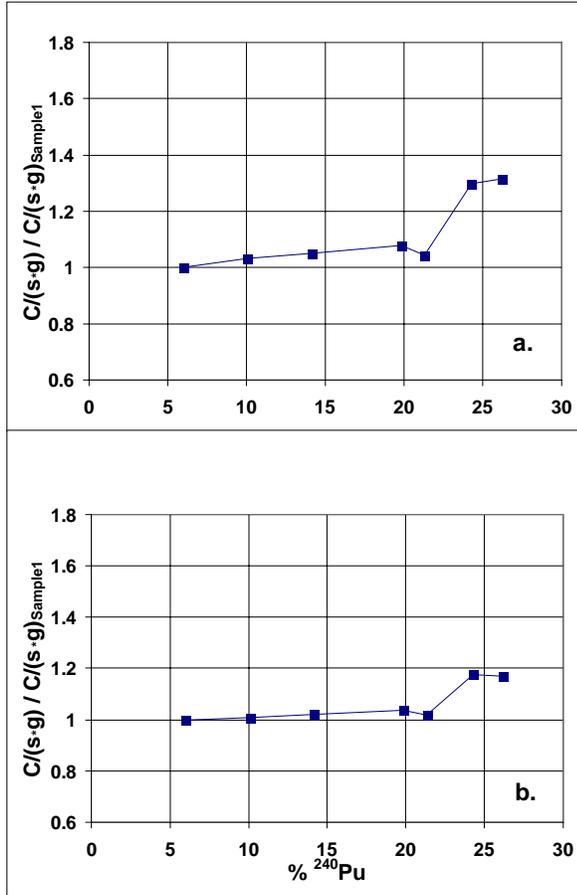## V. Discussion and Conclusions
A. Performance, Reliability/Stability and Efficiency

The measured performance of the commercial, coplanar-grid CdZnTe detector is summarized in Table II. The energy resolution (FWHM at 662 keV) is 2.4 times better than that of the compact NaI detector to which it is compared in this experimental study. This performance, as well as the net count rates of events in the gamma-ray peaks, was shown to be constant with count rate, temperature, and time, and unaffected by power shutdowns between measurement periods. The response of the detector is a linear function of gamma-ray energy. Each of these characteristics exceeds the capability of the compact NaI detector, and each contributes directly to improvements in quantitative analysis by gamma-ray spectroscopy, with particular benefits to portable and unattended applications. The FWTM at 662 keV is comparable to that of the NaI detector which limits the benefits of the improved performance to some extent, as discussed below.

The experimental gamma-ray detection efficiency (including solid angle) of the commercial, coplanar-grid CdZnTe detector is also summarized in Table II relative to that of the of the compact NaI detector. At 122 keV, it is 19%, which is approximately equal to the ratio of the solid angles (1:5, respectively) of the two detectors. At 662 keV, the lower intrinsic efficiency of the CdZnTe detector causes the detection efficiency to drop to 6% of that of the NaI detector. The relative value at the assay energy of 414 keV is 7%. Despite resolution that is superior to that of NaI, this detector

is not a sufficient substitute for the compact NaI detector in the portable applications to quantitatively assay plutonium. A larger CdZnTe detector with comparable energy resolution (all within expectations of the relatively near-term commercial capability) that is proposed in Table II has a sufficient detection efficiency at 414 keV, relative to the compact NaI detector, to replace the compact NaI detector in the portable applications to quantitatively assay plutonium and provide the significant advantages of increased stability and reliability, smaller size and greater assay accuracy (as discussed next).

B. Quantitative Analysis of ^{239}Pu with Coplanar-Grid CdZnTe Detector

The quantitative analysis of ^{239}Pu in variable-burnup plutonium oxide has been performed using room-temperature gamma-ray spectroscopy with the commercial prototype coplanar-grid CdZnTe detector and the compact NaI detector. The exercise was designed to evaluate the assay bias that is caused by finite resolution and interference effects that increase with burnup. The results demonstrate a factor of 2 reduction in the assay bias observed with CdZnTe compared to NaI using identical spectral energy regions, indicating the magnitude of the benefits of improved resolution. Furthermore, analysis regions of 401 to 425 keV (peak) and 428 to 478 keV (background) for the CdZnTe detector gives a factor of 4 reduction in the range of the bias (66% compared to 17%) compared to results obtained historically with analysis regions of 401 to 450 keV(peak) and 501 to 550 keV (background) for the NaI detector. However, improvements in accuracy are still desired to reduce the residual 0% to 17% bias over the range of 6% to 26% for the ^{240}Pu fraction. Most of the 17% bias determined experimentally is incurred between 16% and 26% ^{240}Pu, and a realistic range for fabrication of fuel processed from high-burnup material might be 16% to 36%. Furthermore, residues from chemical extraction processes that concentrate plutonium in production facilities can be burdened with ^{241}Am at the 50% level, an order-of-magnitude increase over the ^{241}Am content of the highest-burnup plutonium sample used in these experiments. Therefore, a larger bias range than 17% might be observed in equivalent field applications of a coplanar-grid CdZnTe detector with these performance characteristics.

Improved energy resolution will reduce the bias further. Compensation for the burnup-dependent contributions of the Compton continuum in the assay region, which will be simplified with improved resolution, will also contribute to a reduction in the bias. However, given that the applications also require larger crystals, the benefits of the improved commercial technology that has recently achieved a resolution of 2% with a 1-cm^3 coplanar-grid CdZnTe detector[24] may be offset somewhat by the need to compromise performance for higher detection efficiency. Another realistic focus for improved assay (reduced bias) with larger crystals is an improved ratio of the FWTM to FWHM at 662 keV (2 to 1 is now available compared to 4 to 1 for the detector used in these experiments; refer to Table II).

C. Extension of Capabilities with Coplanar-Grid CdZnTe Detectors

1. Isotope identification and verification of isotopic distribution. Additional benefits of the improved resolution that is provided by the coplanar-grid CdZnTe detector are demonstrated by applying a robust peak-search routine to the gamma-ray spectra shown in Fig. 13-14. The program entitled Spectool uses a first-difference procedure[25] to instantaneously and automatically identify gamma-ray peaks without fitting the data. The application of such a program to NaI gamma-ray spectra of plutonium does not give a useful result. However, the results for the 6000-s (100-min)

CdZnTe spectra given in Table III show that 18 expected peaks (13 corresponding to ^{239}Pu and ^{241}Pu, as well as ^{241}Am and ^{237}U gamma-rays, three representing plutonium and americium x-rays) are identified in these spectra in patterns that are consistent with the burnup of the plutonium material. Only two anomalous identifications are made. Similar results are obtained with the 600-s (10-min) gamma-ray spectra as well. This capability suggests several useful enhancements for unattended and portable applications of room-temperature gamma-ray spectroscopy including (1) isotope identification during the survey phase of holdup measurements, (2) diagnosis of interferences that give rise to bias in quantitative nondestructive analysis and cause verifications to fail, (3) reduction of the sampling requirements for destructive analysis by increasing the number of items verifiable by nondestructive analysis during inspections, and (4) determination of the relative ^{239}Pu and ^{241}Pu fractions to verify stream-value assumptions on isotopic distribution that are frequently made in the real-time quantitative interpretation of the results of hundreds or thousands of gamma-ray spectra obtained in the portable or unattended mode.

2. Portable or unattended gamma-ray spectroscopy. The stability of the coplanar-grid CdZnTe detector that has been observed experimentally is very important for applications to portable or unattended gamma-ray spectroscopy. In both applications, the real-time readout of quantitative results obtained from gamma-ray spectra acquired in short count periods is required continuously for extended periods of time. The user/operator in portable applications cannot be simultaneously responsible for performing the measurements and assuring the quality of the data. In unattended applications, no user is present. Room-temperature gamma-ray detectors such as CdZnTe that display minimal temperature drifts in the gain are being coupled with automated, rapid, robust procedures for gain-drift compensation[21] that are implemented in real time within the miniature gamma-ray spectroscopy system (see Fig. 3) on a spectrum-by-spectrum basis for extended time periods without need for operator intervention. The improved stability and performance (energy resolution) as well as the reduced size and increased ruggedness will also simplify measurements, improve the results obtained, and extend the information that the measurement provides in portable and unattended monitoring applications. The compact (3.2-cm diameter, 12.5-cm length) cylindrical package in which the recent prototype coplanar-grid CdZnTe detector has been provided (Fig. 3) is easily shielded, and the shielded detector is easily handled or placed in remote locations for both applications. Detectors similar to this design will be used in unattended and selected portable applications of gamma-ray spectroscopy.

Despite the advantages of its performance, implementation of the coplanar-grid CdZnTe detector will not be practical for plutonium assay in the majority of portable applications (including measurements of holdup) until larger detectors of comparable performance are available. The efficiency for detecting 400-keV gamma rays with the prototype coplanar-grid CdZnTe detector used to perform the measurements described in this paper is only 7% of that with the compact NaI detector that is currently used for holdup measurements of plutonium. The "proposed" detector of comparable performance described in Table II, with dimensions sufficient to achieve a detection efficiency that is 31% of that with the compact NaI detector at 400 keV, meets the minimum requirement for consideration as a replacement for the compact NaI detector for portable measurements of plutonium holdup. Portable pplication to uranium assay are in progress with current detectors.

Table III. Second-Difference Peak-Search Results (keV) for the 100-min Plutonium Gamma-Ray Spectra (Coplanar-Grid CdZnTe Detector)

Gamma-Ray Energy (keV)	Isotope or Element	Plutonium Reference Sample Number						
		1	2	3	4	5	6	7
59.5	^{241}Am	60	60	60	60	60	60	60
100-104	Pu K$_a$	101	101			102	101	101
105-107	Am K$_a$						108	108
116-117	Pu K$_B$	117	117	117	118	118	118	118
129.3	^{239}Pu	130	130	130				
148.6	^{241}Pu	148	149	149	149	149	149	149
								157*
164.6,166.0,169.6	^{241}Am			165	166	166	167	167
203.5	^{239}Pu							
208.0	^{237}U	208	208	209	209	209	209	209
267.5	^{237}U							
332.3	^{237}U	334	334	333	334	334	334	334
375.0	^{239}Pu	379	380	377	376	377	375	375
					380		381	381
413.7	^{239}Pu	413	413			414		
		417	418	418	419	420		
511.0	e$^+$ annihil.					518	520	516
582.9	^{22}Na						585	583
		615*						
662.4	^{241}Am	664	659	662	662	662	663	662
721.9	^{241}Am	724	721	725	726	725	725	726
769.4	^{239}Pu	764	763					

*peaks of unknown identity found in the search.

3. Response functions applied CdZnTe spectra for plutonium assay. Two of the origins of bias in nondestructive assay of plutonium with room-temperature gamma-ray detectors are variable burnup and other variations in isotopic composition. The effects impact domestic production facilities, facilities that use or process plutonium fuel, and numerous research and test facilities, as discussed in Section IV.G earlier. However, bias in the gamma-ray spectroscopy assays of plutonium (uranium, etc.) that arises from gamma-ray interference is not limited to effects of variable plutonium isotopics and burnup. Examples from experience with room-temperature

detectors in holdup measurements of ^{239}Pu using the 414-keV assay region include several direct interferences with the assay region. One is caused by the Compton continuum from the 662-keV gamma ray of ^{137}Cs, a fission product that is often found in recycled materials (similar to the effects of the 662- and 722-keV gamma rays of ^{241}Am). Another direct interference with the 414-keV assay region encountered in holdup measurements of plutonium is caused by gamma rays from ^{237}Np[8], which is extracted with plutonium. Common indirect interferences with other useful portions of the spectrum include ^{235}U gamma-ray peaks, predominantly at 186 keV, in the analysis region of the 208-keV reference peak used for internal assurance of spectrum quality in quantitative measurements of plutonium. Similar examples exist for uranium assays with room-temperature gamma-ray detectors. Thus, all facilities and needs in which room-temperature gamma-ray detectors are or could be used for quantitative assay of uranium or plutonium will benefit from the improved performance/resolution of the coplanar-grid CdZnTe detectors. However, while improved resolution can contribute significantly to reduction of bias from these effects in room-temperature applications of gamma-ray spectroscopy, a residual bias remains in many cases, including those examined experimentally. For most cases involving room-temperature, finite- (low- or intermediate-) resolution gamma-ray detectors, elimination of the bias requires extraction of the assay component of the signal from an assay region that includes a significant interference component. Because the resolution is finite and the influence of the continuum on the signals measured with low- or intermediate-resolution gamma-ray detectors is large, the only consistent extraction involves the use of gamma-ray response functions.

An empirical approach to the development of response functions for compound semiconductor detectors with novel electrode designs is not practical. The crystal shape and dimensions alone give rise to visible qualitative differences in the shapes of monoenergetic gamma-ray peaks and the continuum from interactions of these gamma rays. The electrode design and how bias is applied to the electrodes are two independent variables that impact the peak shape significantly. The design of the analog circuitry that combines the multiple preamplifier inputs (such as those for the pair of coplanar-grid electrodes of the prototype CdZnTe detector) also independently influences the peak shape and continuum in a visible way. The application of response-functions to analysis of gamma-ray spectra obtained with the compound semiconductor detectors requires a new, computational approach that incorporates the variable characteristics of the detector to determine the response functions specific to the design and setup of each detector.[19, 20] This computational approach requires detailed input on the intrinsic properties and the design of the detector (including electrodes) in order to model the electric field and the production, transport, and collection of charge. Some preliminary calculations have been performed for proof of the principle of such an approach. The gamma-ray spectra that have been calculated for the prototype, coplanar-grid CdZnTe detector include some monoenergetic gamma-ray sources and four (numbers 1, 3, 5, and 7) of the seven plutonium reference samples.[19, 20] The isotopic fractions for these samples given in Table I and the gamma-ray energies and branching ratios for the isotopes of plutonium and their ^{241}Am and ^{237}U daughters were the only sample-specific inputs to these calculations.

Figure 18 shows the four composite gamma-ray spectra that were measured with these samples (shown previously in Fig. 13a) plotted on the same scale as the corresponding calculated gamma-ray spectra for these samples. The preliminary calculations are a good visual match to the measured data and demonstrate the promise of this approach in obtaining improved, quantitative assays of

plutonium using room-temperature gamma-ray detectors such as the coplanar-grid CdZnTe detector with enhanced capability provided by calculated response functions. They also support enhanced functionality of the room-temperature gamma-ray detectors in determination of plutonium isotopic composition and in evaluation of effects of self-attenuation in portable applications. Future field instruments that use the new, robust, room-temperature coplanar-grid CdZnTe detectors with enhancements provided by calculated response functions to

- simultaneously determine the quantitative gamma-ray assay and the isotopic composition of plutonium and uranium,
- supplement neutron assays by measuring the isotopic composition, or
- provide detailed spectroscopic information to improve the quantitative gamma-ray assay will be substantial additions to the tools for nondestructive analysis in safeguards applications.
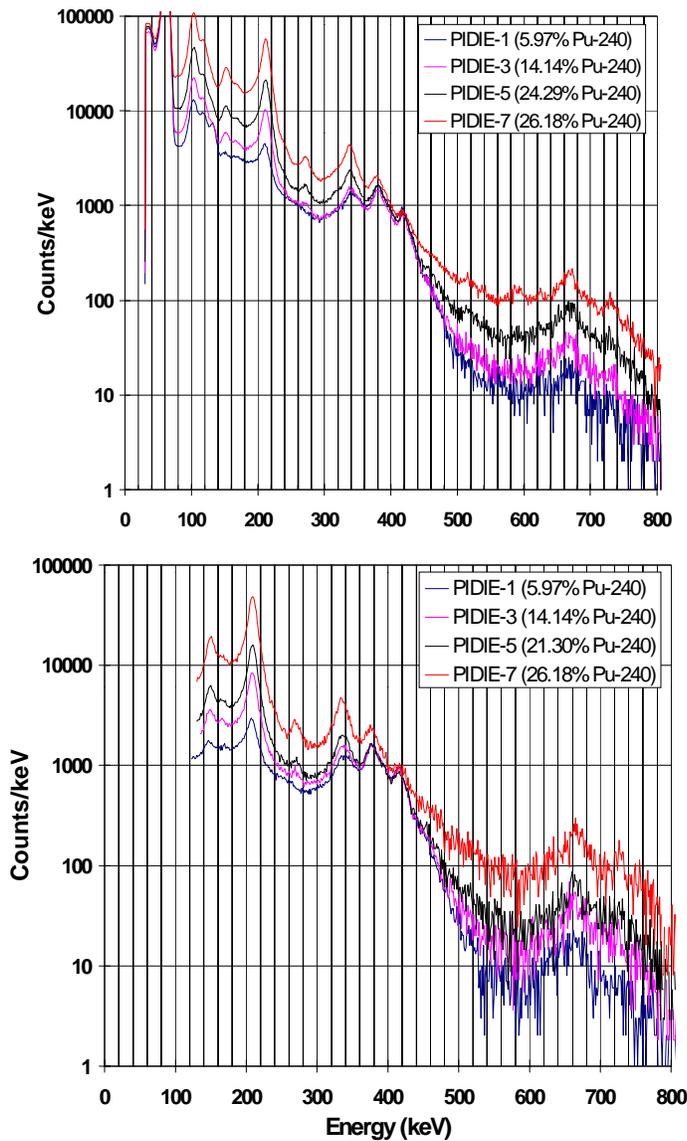

Fig. 18. Logarithmic plot of the (a) experimental and (b) calculated gamma-ray spectra from 0 to 800 keV of plutonium reference samples 1, 3, 5, and 7 (as described in Table I) for the prototype coplanar-grid CdZnTe detector.

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