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Approaching cryogenic Ge performance with Peltier cooled CdTe

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ABSTRACT

A new class of hand-held, portable spectrometers based on large area (1cm^2) CdTe detectors of thickness up to 3mm has been demonstrated to produce energy resolution of between 0.3 and 0.5% FWHM at 662 keV. The system uses a charge loss correction circuit for improved efficiency, and detector temperature stabilization to ensure consistent operation of the detector during field measurements over a wide range of ambient temperature. The system can operate continuously for up to 8hrs on rechargeable batteries. The signal output from the charge loss corrector is compatible with most analog and digital spectroscopy amplifiers and multi channel analyzers. Using a detector measuring 11.2 by 9.1 by 2.13 mm^3, we have recently been able to obtain the first wide-range plutonium gamma-ray isotopic analysis with other than a cryogenically cooled germanium spectrometer. The CdTe spectrometer is capable of measuring small plutonium reference samples in about one hour, covering the range from low to high burnup. The isotopic analysis software used to obtain these results was FRAM Version 4 from LANL. The new spectrometer is expected to be useful for low-grade assay, as well as for some *in-situ* plutonium gamma-ray isotopics in lieu of cryogenically cooled Ge.

Keywords: Cadmium Telluride, CdTe, p-i-n detector, energy resolution, plutonium, isotopics, gamma ray spectroscopy

1. INTRODUCTION

Our earlier work shows that the technology of CdTe crystals growth and the fabrication technology of p-i-n structures with high breakdown voltage allows for creation of high performance detectors with sensitive volume larger than 0.3 cm^3. The detection efficiency for γ-rays of such detectors is equivalent to that of Ge detectors with sensitive volume of about 2 cm^3 in the energy range from 200 keV to 1500 keV. Near room temperature operation, excellent energy resolution and reasonable detection efficiency of CdTe detectors can significantly impact the measurement capabilities in the area of nuclear material inspection particularly by use of battery operated portable spectrometric systems. However, in order to examine nuclear materials with low intensity of emission of γ-rays detectors with higher detection efficiency are required. Experimental measurements and theoretical calculations conducted by many researchers [1-4] show that the main restriction of spectrometric characteristics for CdTe detectors is the incomplete charge collection, which limits the useful active thickness of the detector. On the other hand the active area of the detectors is limited by the size of the single crystal grains available. With the increase of the detector thickness the drift time of charge carriers increases proportionally, and charge loss increases due to the trapping effect. It is apparent that even for the maximum value of $\mu\tau$ (mobility-trapping time) product for electrons and holes achieved now in CdTe, the thickness of the sensitive zone of spectrometric detectors (approaching the energy resolution of cryogenic Ge detectors) will be limited to about 3-4 mm. Further increase in volume can be achieved by stacking CdTe detectors.

In this paper we report the latest spectrometric results obtained with CdTe p-i-n structures. We also describe the implementation of a portable gamma spectrometric system based on a high-resolution CdTe p-i-n detector. The portable system has been used at Los Alamos National Laboratory (LANL) for inspection of a low-grade plutonium assay in lieu of cryogenically cooled Ge.

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2. CdTe P-I-N STRUCTURES

Recent remarkable progress has been achieved with CdZnTe (conduction counter) and CdTe (p-i-n) structures in the x-ray and gamma ray spectroscopy area. CdZnTe can provide resistivity as high as 10^{11} Ω-cm. Despite the use of a conduction counter detector configuration, at -40°C with $5x10^3$ V/cm electric field, the leakage current can be suppressed to the degree that 240 eV FWHM at 5.9 keV is possible [5]. CdTe compensated with Cl can achieve a resistivity of about 10^9 Ω-cm. The use of p-i-n junction structures helps to control the leakage current and allows the use of higher electric fields. Optimally, detectors should operate at electric field values assuring the saturation of the drift velocity of charge carriers. For CdTe crystals this value is around 15 - 20 kV/cm, thus for a p-i-n structure with a 3-mm thick i-region, the p-i-n diode should not exhibit a breakdown under a reverse bias condition at voltages below 3kV (for planar geometry). We have developed fabrication technology for p-i-n detectors that satisfy these requirements. Also, we have incorporated detector surface passivation providing long-term stability of the leakage current.

The CdTe p-i-n structures presented in this paper can tolerate bias voltages of 2.5 - 3.0 kV with leakage currents much lower than 10^{-11}A at – 30 ^{0} to – 40 ^{0}C. This feature, in combination with the good charge collection for both electrons and holes leads to construction of high-energy resolution spectrometers. In addition to good energy resolution there are a number of other detector characteristics that are of particular high importance for commercial devices, including: a) stability of detector characteristics over a long operating time b) good peak-to-background ratio and c) symmetry of peaks in the collected spectra.

3. MEASUREMENTS OF MOBILITY-TRAPPING TIME PRODUCTS

The selection of a starting material for fabrication detectors includes measurements of μτ □products and their non-uniformities. The measurements are performed with alpha particles separately for electron and holes. The results of measurements are fitted to the Hecht relation [6] from which values of $(\mu\tau)_e$ and $(\mu\tau)\square_h$ are derived. The non-uniformity of the charge collection parameters are measured by scanning of the detector with a collimated alpha source. Since the current method for measurement of mobility-lifetime products for electrons and holes in CdTe and their distribution in the crystals give reliable results, it is possible to predict the spectral characteristics of a detector produced from each crystal with a sufficient accuracy.

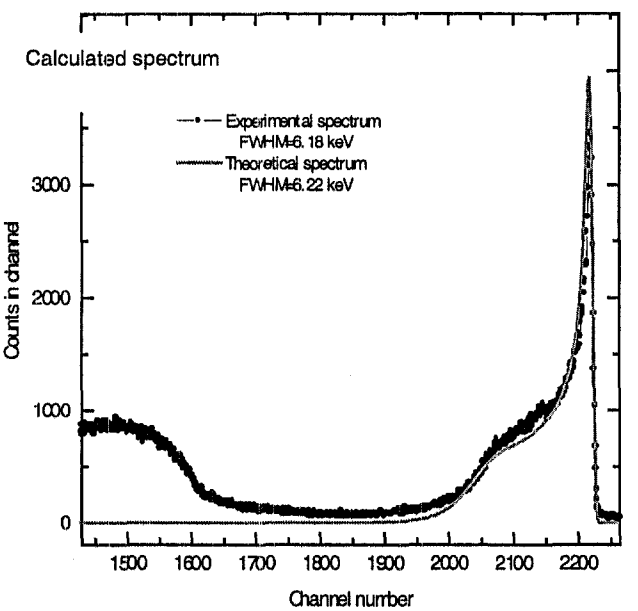


Figure 1: Calculated vs. experimental spectrum obtained from CdTe detector as a response to 662 keV γ-rays.

For example, models of amplitude spectrum calculations in response to gamma radiation developed by Iwanczyk [7,8] and Trammel and Walter [9] are useful to predict the CdTe detector spectral characteristics. The measured and calculated spectra of ^{137}Cs are shown in Figure 1. The dotted curve represents the measured spectrum whereas and the narrow solid line is the spectrum calculated from the theoretical model [7,8,9]. It can be seen from Figure 1 that the calculated full energy peak fits the experimental data quite well. The detector parameters used in the calculation are the following: sensitive region thickness - 2.8 mm; sensitive region area - 100 mm^2 (planar geometry); detector operation voltage - 3000 V; $(\mu\tau)_e = 2.45 \times 10^{-3}$ cm^2 / V; $(\mu\tau\square)_h = 1.4 \times 10^{-4}$ cm^2 / V, non-uniformity of $(\mu\tau E)_e$ FWHM = 80% of average value, non-uniformity of $(\mu\tau E)\square_h$ FWHM =45%, and E is electric field. We have assumed that the drift length of electrons and holes $\lambda = (\mu\tau E)$ has normal distribution over the detector area.

It can be seen from the model that for the described values of the $\mu\tau$ products for electrons and holes, if their non-uniformities are kept low (FWHM < 100%) it is possible to produce p-i-n detectors with energy resolution of better than 0.5 % for gamma radiation above 200 keV.

4. MEASUREMENTS OF NON-COMPENSATED IMPURITIES

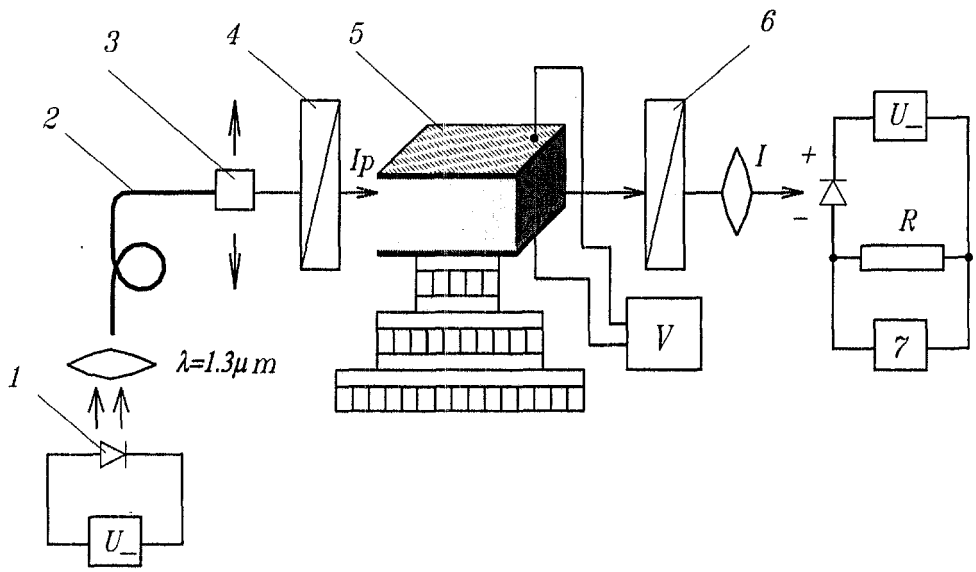


Figure 2: Experimental setup used to measure electric field intensity across the Peltier-cooled crystal. 1 – semiconductor laser $\lambda = 1.3$ μm, 2 – multimode optical fiber, 3 – lens, 4 – IR polarizer, 5 – detector, 6 – IR analyzer, 7 – voltmeter/oscilloscope

Another important characteristic of p-i-n diodes constructed for spectrometric detector applications is the long-term stability of the electric field distribution in the intrinsic region of the structure. For measuring the distribution of the electric field intensity in the p-i-n structure we use a well-known technique, which is based on measurement of electro-optic effects in crystals. The technique utilizes a scan over the intrinsic region of the p-i-n diode by a narrow beam of infrared light passing through the sensitive region and providing information on the electric field intensity along the illuminated path. Our implementation of this technique is based on rotation of polarized IR light by the Pockels effect. A block diagram of the instrumentation used in the measurements is shown in Fig.2. Our experimental setup is similar to that used by Kasherininov [10] and Hagi Ali [11] with the only difference that we make measurements of the electric field distribution as the crystal is cooled to – 40°C with the use of a Peltier cooler.

After the reverse voltage is applied to the detector some polarization occurs in the intrinsic region due to the presence of deep trapping levels (non-compensated impurities) and the field intensity distribution assumes a shape characteristic of the space charge region of the p-n junction. The concentration of charge centers producing the particular electric field intensity distribution E(x) can be easily obtained from the width of space charge region and the E(x) dependence slope. The concentration of the charge centers in our best material is measured to be less than $N_t = 6 \times 10^9$ cm^{-3}. In addition, under high

working voltage conditions the non-uniformity of electric field intensity is reduced due to increase of the average field intensity in the intrinsic region. Nevertheless, charging of the centers over time creates changes in the distribution of the spatial charge region leading to changes in the electric field and corresponding changes in the pulse shapes generated in the response to gamma radiation. As a result of this effect we observe changes in energy resolution of about 0.2 – 0.5 keV (FWHM) at 662 keV over 8 hours of continuous detector operation. These changes can be ameliorated to a certain degree by external electronics such as an adaptive charge loss corrector or by periodic cycling of the detector bias voltage (bringing the charge centers to their original state).

5. PORTABLE SPECTROMETRIC SYSTEM "POLARIS".

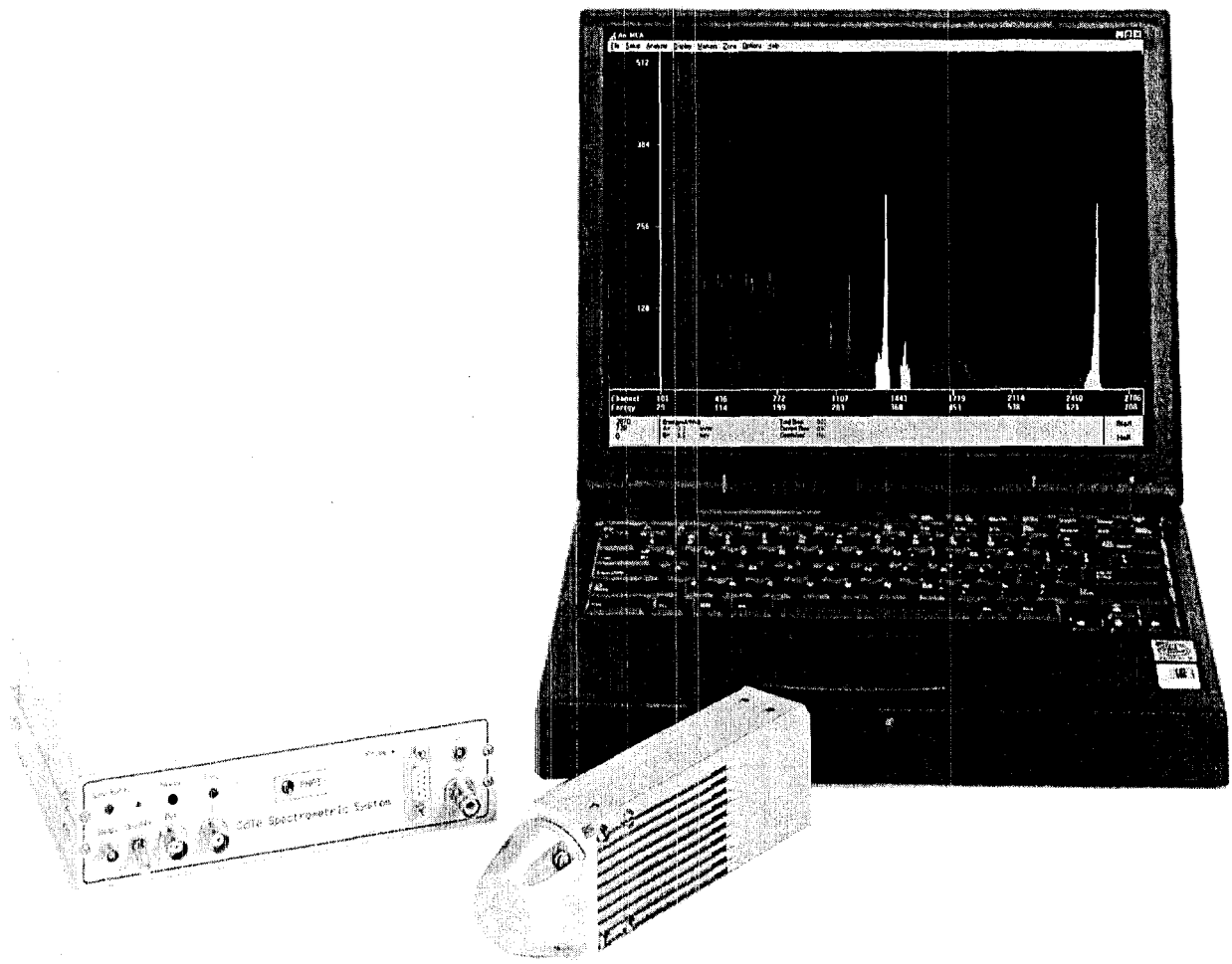


Figure 3. Portable spectrometric system "Polaris".

A portable spectrometric system "Polaris" was developed based on the high resolution p-i-n CdTe detectors. "Polaris" is a portable, battery operated system intended for detection and spectroscopy of x-rays and γ-rays. The system was designed to work with standard analog spectroscopy amplification and processing electronics such as MCA-166 or with digital processors such as the Canberra Inspector 2000™ or the Perkin Elmer-Ortec Digidart™. The entire detector system consists of two main parts connected by a cable, namely a detector unit and a power supply/processing module.

By coupling the Polaris device with one of the portable battery operated MCA's the device is autonomous. Three Li rechargeable batteries supply power for the whole detector system for up to 8-hours of continuous operation. Besides batteries, an external power source, battery pack or standard charger can be used. The total weight of the system is 2.6 kg and maximum power consumption is lower than 7W. Figure 3 shows a photograph of the Polaris system including detector unit, power supply, and laptop computer for displaying the measured data.

5.1 Detector unit.

The detector unit (Fig. 3) consists of detector head, charge-sensitive preamplifier and low noise fan. The detector head is situated on the top surface of the unit and includes detector and front-end electronics, both mounted on the Peltier cooler. Such arrangement together with the conical shape of radiator allows measurements in hard-to-reach spots.

The detector is produced from high quality single crystal CdTe. The typical detector parameters are as follows: dimensions - 10x10x3.0 mm, working temperature -35°C, bias voltage - 3.0 kV. These parameters slightly vary from one to another unit.

5.2 Power Supply module

The module is used to supply power for and control the detector unit. It consists of the battery pack, low and high voltage supply circuits, charge loss corrector, timer, temperature control and regulation circuit (thermocontroller), and inhibit module.

All necessary connectors and switches are situated on the front panel shown in Fig. 3. Three LEDs (also on the front panel) indicate the device operating status. Inside the module there are three lithium batteries, 21.6 W-Hr and 7.2 V each. The battery is deep discharge protected. When the battery pack voltage drops to 5.5 V the yellow LED "Low power" is lit signaling that the battery pack must be recharged; an external power source may be used to go on with measurements in this case. A green "Ready" LED indicates the mode of operation (flashing LED indicates that the device is in set-up mode, constantly on LED indicates that the device is ready). The electronics fit on one main system board, containing the logic circuits, the thermocontroller and timer, four low voltage supply modules and a single high voltage DC-DC converter module.

The battery pack voltage of 7.2 V is transformed to ±12V and +5V. Voltage transformers, based on MAXIM integrated circuits, produce +12V and –12V needed for the amplifier, CLC and thermo-controller supply, DC-DC high voltage converter module, and low noise fan. The Peltier cooler is fed directly from the battery voltage through the controller.

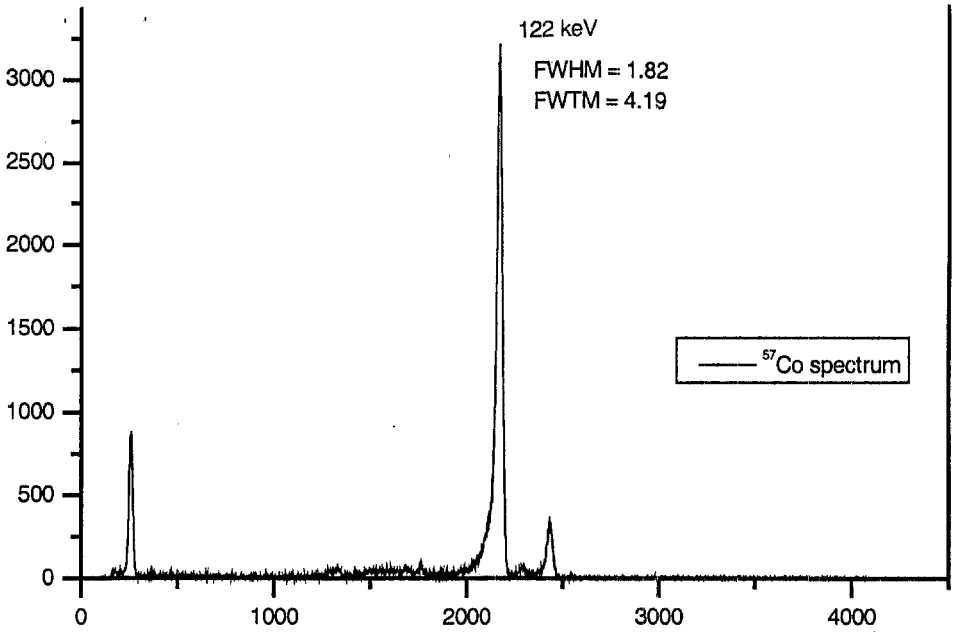
Special stabilization circuitry has been added to Polaris for long term measurements (7 to 30days) such as unattended control of technological processes. This circuit is necessary to mitigate drift in the energy resolution caused by polarization. The polarization is effectively "cleared" from time to time (as necessary based on the stability requirements) by cycling the detector bias voltage. A timer circuit is employed to generate gating signals that periodically shut the high voltage down during very long term operation or intermediate length operations that require ultimate resolution stability. The timer frequency is individually selected for each detector and type of measurements. Under the tightest constraints voltage cycling is required each two to four hours, and lasts for a few tens of seconds. Jumpers on the system board are used to set the timer values. An inhibit circuit is used to prevent measurement during the above-mentioned HV shutdown. The circuit produces a TTL gating signal that can be used with compatible MCA's to gate off the acquisition during the HV cycling. In addition a Red "Inh" LED is lit when the HV is cycled off.

5.3 Temperature measurements

To estimate the influence of environmental conditions a thermal test was performed. During the test the Peltier cooler current, radiator temperature, and detector temperature were monitored during several hours. Two series of tests were performed with ambient temperatures of +20°C and +30°C. Both current and heat sink temperature increased slowly but did not change significantly during the 6-hour test.

5.4 Spectral measurements

Figure 4 shows spectra of a ^{57}Co source and combined ^{57}Co and ^{137}Cs sources taken with a "Polaris" system. The detector used in the system had a sensitive volume of 308 mm^3 (S=100 mm^2 and W=3.08 mm). The operating temperature of the detector was –35°C. The energy resolution of the system for the 662 keV line was 3.54 keV (FWHM) and 1.82 keV (FWHM) for 122 keV line. Energy resolution, peak to Compton ratio and peak symmetry are similar to that of small cryogenically cooled germanium detectors.


122 keV
FWHM = 1.82
FWTM = 4.19
^{57}Co spectrum
3000
2500
2000
1500
1000
500
0
0
1000
2000
3000
4000

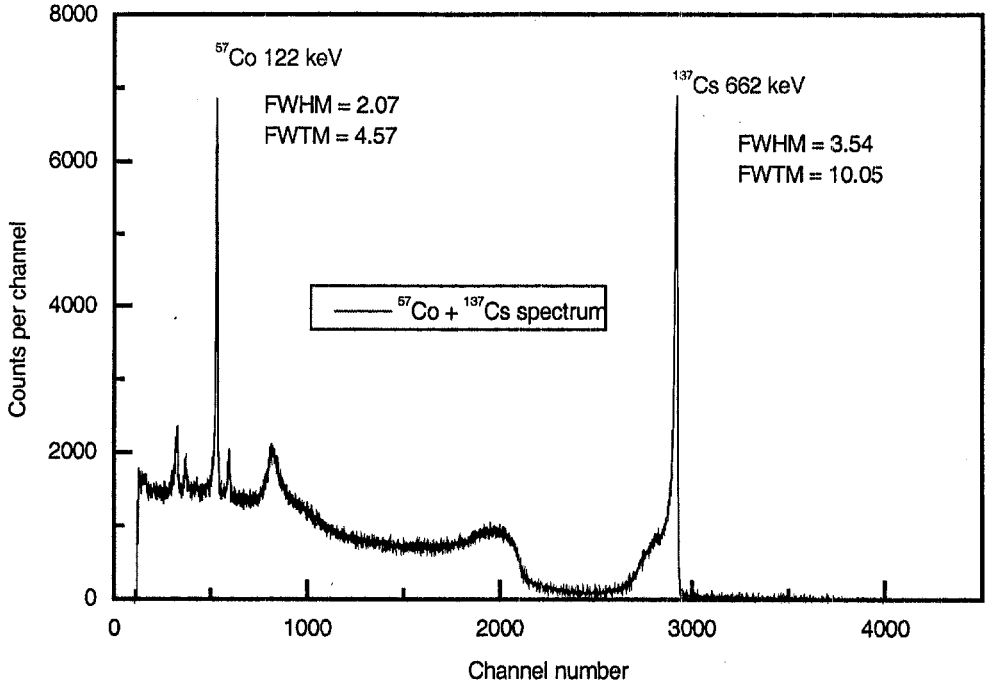

^{57}Co 122 keV
FWHM = 2.07
FWTM = 4.57
^{137}Cs 662 keV
FWHM = 3.54
FWTM = 10.05
^{57}Co + ^{137}Cs spectrum
Counts per channel
Channel number
8000
6000
4000
2000
0
0
1000
2000
3000
4000

Figure 4. Spectra of ^{57}Co and combined ^{57}Co + ^{137}Cs taken with a typical "Polaris" system.

Slight tailing noticeable in the low energy part of the full energy peaks are most probably the result of charge collection non-uniformities associated with the edges of the sensitive volume and can be removed with an improvement of a charge loss corrector.

6. FIELD APPLICATIONS

Nondestructive analysis (NDA) techniques applied to bulk nuclear materials (NM) are important for nuclear safeguards and material control because of timeliness, cost-effectiveness and containment integrity. The use of NDA is vital for quantitative measurements of heterogeneous bulk materials that are difficult or impossible to sample.

Progress with CdTe detectors in the last decade has advanced portable NDA capabilities previously addressed with scintillators. Measurements of uranium and plutonium performed previously with NaI detectors are now feasible with detectors that use relatively large (up to ~0.3 cm^3), near room-temperature operating CdTe detectors with p-i-n structures. Among other advantages in their applications to holdup measurements, the gamma-ray energy resolution of the CdTe detectors is more than an order of magnitude better than NaI in the range from 122 to 662 keV. This can reduce or eliminate bias from interferences that affect the low-resolution NaI measurements. However, detection efficiency with germanium is several times better than that of CdTe due to large volumes of Ge detectors available. Adapting techniques developed for germanium detectors, such as isotopics analysis based on the complex gamma-ray spectra of plutonium, are now becoming possible with CdTe. Los Alamos National Laboratory (LANL) was recently able to obtain the first wide-range plutonium gamma-ray isotopics analysis with a Polaris system. This is important because it is the first time that this type of measurement has ever been made successfully with any detector other than a cryogenically cooled germanium detector. The CdTe spectrometric system "Polaris" is capable of measuring small plutonium reference samples in about one hour, covering the range from low to high burnup. The isotopic analysis software used to obtain these results was FRAM Version 4 from LANL [12]. The new spectrometer is expected to be useful for low-grade assay, as well as for some *in-situ* plutonium gamma-ray isotopics in lieu of cryogenically cooled Ge detectors.

CONCLUSIONS

The attainable gamma ray performance with CdTe (p-i-n) based spectroscopy systems approaches that of cryogenically cooled Ge systems without the need for the liquid nitrogen. The parallel development of miniaturized, low noise, low power processing electronics allows one to construct very convenient instruments for a variety of applications. The reduced weight, power, and size of the spectrometers makes them extremely attractive as portable battery operated instruments for nondestructive analysis (NDA) techniques applied to bulk nuclear materials (NM) that are important for nuclear safeguards and material control.

Future work will concentrate on construction of detectors with larger volume to enhance the detection efficiency of CdTe systems and further improvements of the spectrometric performance.

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