

Evaluation of Mercuric Iodide Detectors for SNM Monitoring

L. van den Berg^a, K.R.Pohl^a, A.E.Proctor^a, I.Hartley^b and Z.W.Bell^c

^aConstellation Technology Corporation, 7887 Bryan Dairy Road, Largo Fl 33777,U.S.A.

^bIAEA, Wagramer Strasse 5, P.O. Box 100, A-1400, Vienna, Austria

^cBWXT Y-12, P.O. Box 2009, Oak Ridge, TN 37831, U.S.A.

Abstract

Detectors made from single crystals of mercuric iodide are used in increasing numbers for the detection and analysis of gamma radiation. The large density and high atomic numbers of the constituent elements of mercuric iodide result in a high efficiency compared with other solid state detectors. The material has a very high threshold for damage by neutrons and charged particles, and its high resistivity makes it possible to fabricate large area detectors with low noise levels at ambient temperatures. The technology to fabricate these detectors makes it possible to manufacture large volume counters and sensitive spectrometers with a resolution of 3% FWHM or less at the gamma ray energy of 662 keV. They have been operated between -20 to 60 degrees Celsius, and the performance is stable over periods of years. These properties make the mercuric iodide detectors ideally suited for the routine monitoring of stored materials, hold-up measurements, and in unattended systems.

The spectra of several concentrations of Plutonium 239, Highly Enriched Uranium, and Depleted Uranium have been measured at facilities where samples of these materials were available. To perform these measurements the detectors were installed in a light-tight module, together with a preamplifier and a voltage stabilizing system. Modified NIM power and signal processing electronics were used. The detailed features of the spectra will be shown and discussed.

Introduction

Mercuric iodide has been considered for use as an ambient temperature solid-state radiation detector for many years because of its physical and electronic properties. It is only during the last few years, however, that the technologies of material preparation, crystal growth, and detector fabrication have progressed to the point that high quality detectors with large active volumes can be fabricated on a routine basis. These detectors, combined with suitable electronic pulse processing systems, can be applied to the intermittent or continuous monitoring of stored Special Nuclear Materials (SNM).

The single crystalline form of mercuric iodide suited for radiation detection applications is a semiconductor with an electronic bandgap of 2.13 eV at room temperature. Because of this wide bandgap, the resistivity of the material approaches a value of 10^{14} Ohm.cm. Consequently, the leakage current of large area detectors is in the nano-ampere range, even at electric fields of 10^4 V/cm. Therefore, as is often the case in other semiconductor detectors at room temperature, the detector leakage current is not a significant factor in the overall noise spectrum of the detector, except at lower energies.

The crystalline material has a density of 6.3 g/cm^3 , which gives it a high absorption coefficient for x-rays and gamma rays. The most distinguishing feature of mercuric iodide, however, is the high atomic numbers of the constituent elements (80 and 53), which results in a very large photoelectric effect and a high gull-energy peak efficiency, as discussed in a previous publication.⁽¹⁾

Several spectrometer grade detectors have been tested for stability of performance over several months,⁽²⁾ and over a temperature range of -20 to $+50$ degrees Celsius.⁽³⁾ The performance of these detectors remained stable, with only minimal changes in the position of the full-energy peak. The stability and performance of mercuric iodide detectors have also been evaluated while they were subjected to different types and intensities of beams of charged particles and neutrons. A comprehensive report has been published recently reviewing the results of these experiments.⁽⁴⁾ None of the experimental conditions created observable changes in the properties of the detectors subjected to the particle beams.

This manuscript describes the performance of these detectors when exposed to a variety of samples of typically stored and monitored special nuclear materials, and to a lesser degree the electronic systems that have been developed to optimize the spectral output of the detecting system.

Equipment

Since Constellation Technology does not have access to the sources used in these experiments, it was necessary to perform the measurements at different locations where the proper samples were available. The detectors used in these measurements varied in area and thickness. For the measurements performed at Los Alamos National Laboratory, the detector size was $10 \text{ mm} \times 10 \text{ mm}$ contact area and 2.8 mm thick. The detector used at the IAEA measured $25 \text{ mm} \times 25 \text{ mm}$ contact area and 1.7 mm thick, and the data from the Y-12 facility in Oak Ridge were collected with a $25 \text{ mm} \times 25 \text{ mm} \times 2.6 \text{ mm}$ detector. For this reason, and because of the differences in the sources and in the physical arrangements of sources and detectors, it is not possible to make any deductions from the intensities of the spectra. Only the significant spectral features of the measured results can be related to the performance of mercuric iodide detectors for the purpose of monitoring and analyzing the properties of the different samples of customarily stored materials.

To perform the tests described in this paper, the detectors were installed in a light-tight metal enclosure, called a Mercury ModuleTM, together with the preamplifier and the voltage stabilizer. This module can be used as a hand-held mobile detector unit, or be permanently installed on top of or to the side of a storage container. Shielding can be added to the sides and the back of the module, so that it becomes a forward-looking unit with only the front face of the detector exposed to the source of the radiation. The back of the module contains the appropriate connections for power supplies and the signal from the preamplifier. A photograph of the module is shown in Figure 1.



Figure 1: Photograph of Mercury Module

The first part of the signal processing unit used in these experiments consisted of a NIM shaping amplifier modified by incorporating a gated integrator in the module. The collection of the charges generated in the mercuric iodide by the incident radiation can take a relatively long time, mainly because of the low transport properties of the holes. Using a standard shaping amplifier requires preset shaping times of up to 24 microseconds, which restricts the count rate to a few thousand per second. Using a gated integrator with a preset shaping time of 2 microseconds and an integrating time of 12 microseconds reduces the noise in the signal and increases the count rate of the detector to at least 10,000 counts per second without causing excessive pile-up. This count rate is more in agreement with the radiation environment encountered in facilities where SNM's are stored. In addition, when the detector/preamplifier is coupled to certain gated integrator-based pulse processing systems, the noise effect of the preamplifier is reduced significantly, improving the spectral resolution of the detectors over the whole energy range.

Experimental Results

1. ^{239}Pu samples

Spectra of ^{239}Pu were measured at LANL and the IAEA. The results are shown in Figure 2. The spectra show clearly the significant lines in the plutonium spectrum. An interesting feature of the LANL spectrum is the relatively intense americium peak at 60 keV, which indicates that the material has aged to some degree.

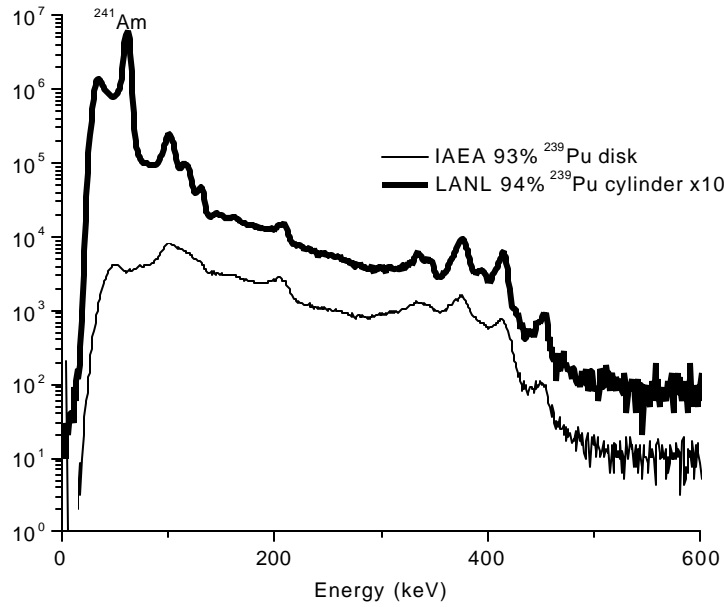


Figure 2: Spectra of ^{239}Pu

2. Spectra of samples with varying concentrations of ^{239}Pu

Spectra of samples with concentrations of ^{239}Pu decreasing from 93% to 61% measured at the IAEA are shown in Figure 3.

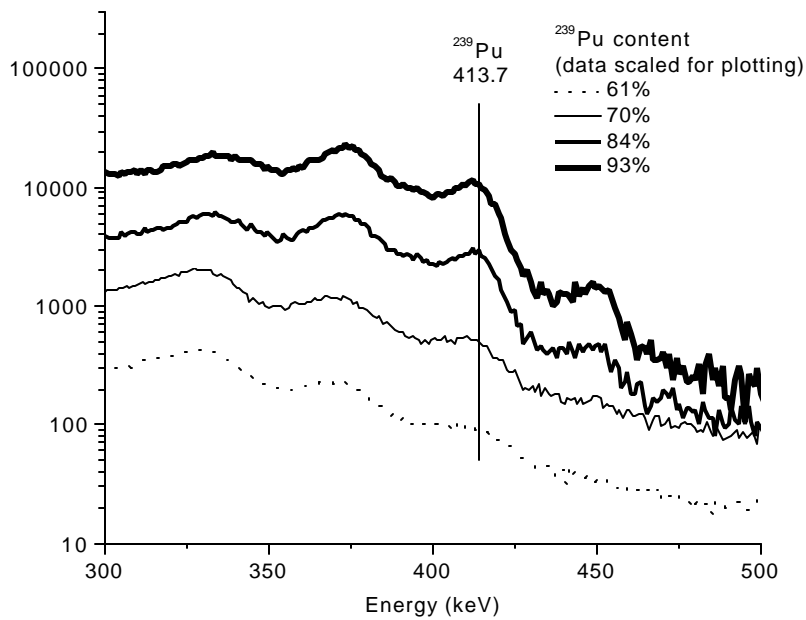


Figure 3: Spectra of Samples with Decreasing Concentrations of ^{239}Pu

The spectral line at 413.7 keV can easily be observed for all samples, except for the lowest concentration of 61%. At this level of activity, it is difficult to distinguish the spectral peak above the noise in the system.

3. Spectra of HEU Samples

Two samples of highly enriched ^{235}U were tested at LANL and at the IAEA. The results are shown in Figure 4.

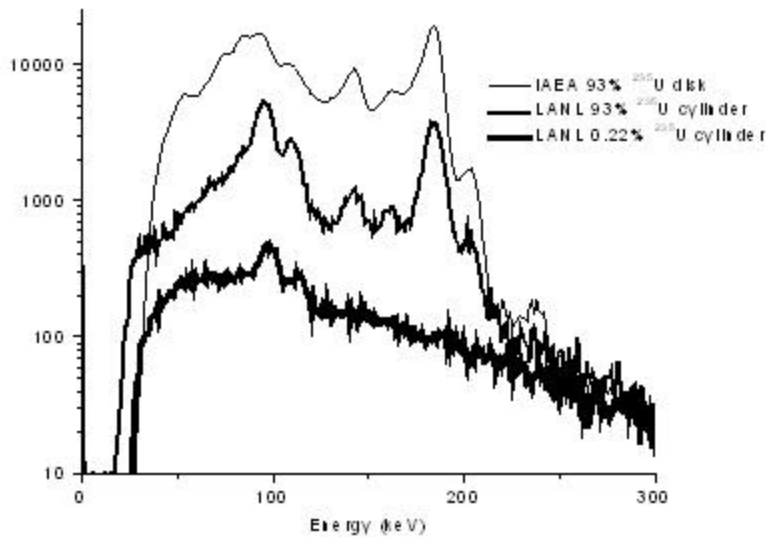


Figure 4: Spectra of two HEU Samples

The identifying line of ^{235}U at 185 keV can clearly be identified in the spectra, in addition to many of the lower energy features. This line is not visible in the spectrum of depleted uranium, which has been added for comparison.

4. High Energy Spectrum of Depleted Uranium

The high energy spectral line at 1001 keV of depleted uranium was measured at the Y-12 facility in Oak Ridge, TN. The spectrum is shown in Figure 5.

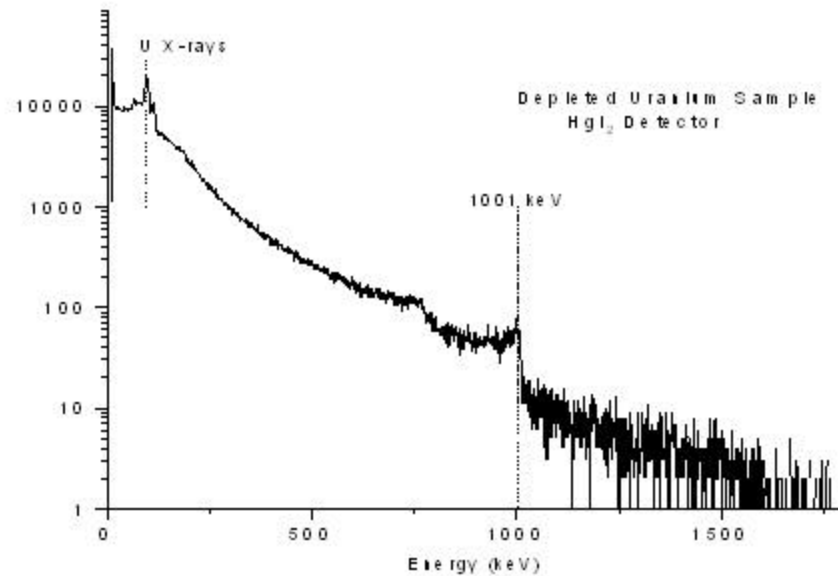


Figure 5: High Energy Spectrum of Depleted Uranium

The use of this high-energy spectral feature makes it possible to detect and monitor the presence of depleted uranium, even when it is shielded by heavy-duty storage containers or in containers with mixed wastes at long-term disposal sites. Monitoring systems based on mercuric iodide technology could be permanently installed at these sites and be remotely controlled and interrogated.

Discussion and Conclusions

The experimental results presented show that spectra with good resolution can be obtained with different samples of SNM using a variety of mercuric iodide detectors. The quality of the spectra may make them amenable for isotopic abundance determinations, using the software developed for this purpose. Typical analysis codes, for example, the MGA code used for analysis of plutonium isotopic composition, require an analytical peak shape function. To determine if such a function could be developed for mercuric iodide, spectra of ^{133}Ba have been measured using a mercuric iodide detector (Figure 6).

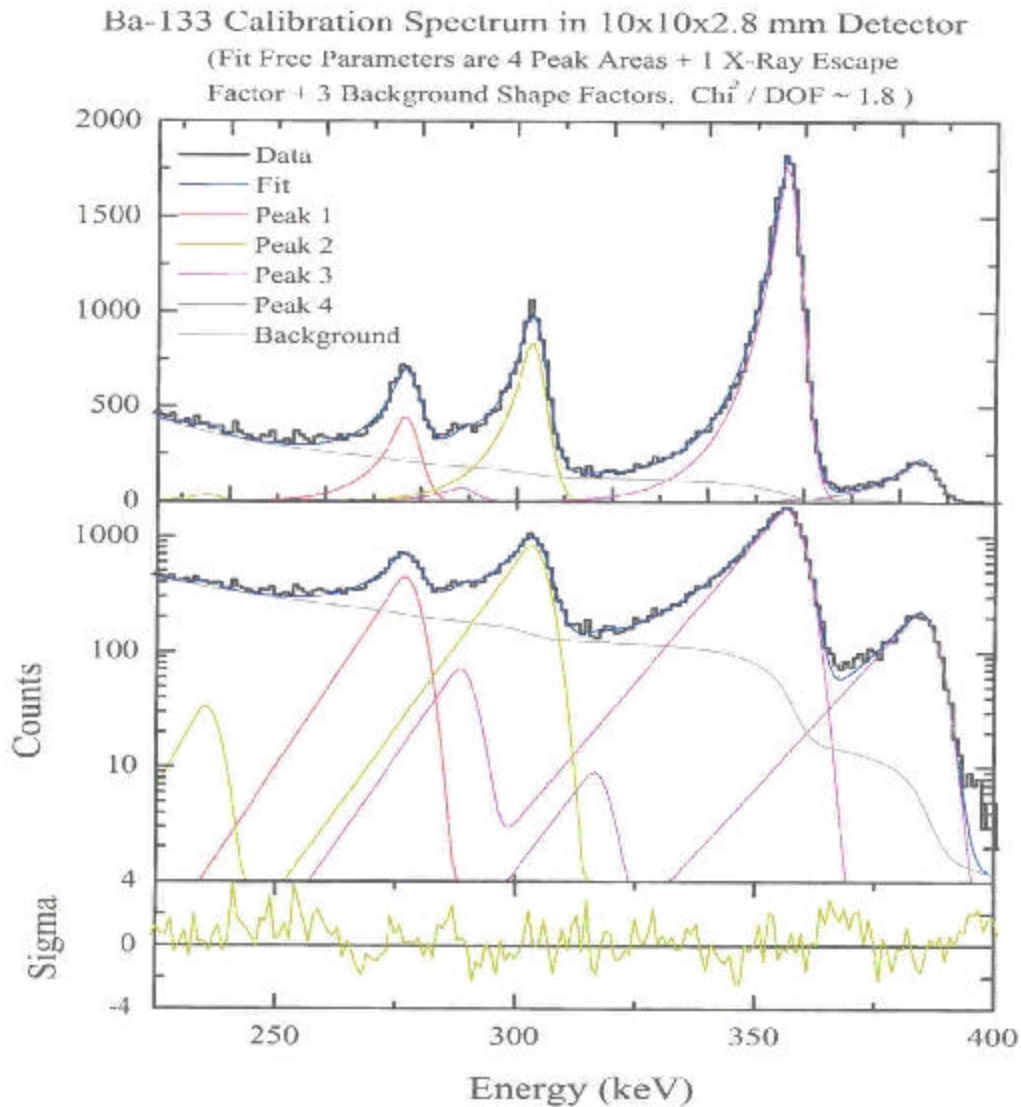


Figure 6: Spectral Peak Fitting of Mercuric Iodide Spectra

The fitting function consists of a Gaussian with tail functions and a second smaller ‘escape’ peak with shape similar to the larger ‘full energy’ peak. Quality of fit, as shown in Figure 6, is excellent. Having determined the analytical function, it should be possible to modify an analysis code, such as MGA⁽⁵⁾, to perform plutonium isotopic analysis. This possibility was discussed briefly with developer of MGA⁽⁶⁾, but no work has been completed to date.

It is clear that improvements in individual detector performance are highly desirable in order to realize the full potential of mercuric iodide, and efforts in this area are continuing. Combined with advanced electronic systems, it will be possible to fabricate detection systems that show higher

spectral resolution and reduced background noise, so that versatile measurement units geared to specific applications can be realized.

References

1. L. van den Berg, A.E. Proctor and K.R. Pohl, Proceedings INNM 42nd Annual Meeting (2001).
2. F.P. Vaccaro et al., SPIE Proceedings Vol.4507, 108-118 (2001).
3. L. van den Berg et al., submitted to SPIE 2003 Annual Meeting.
4. B.A. Brunett et al., SPIE Proceedings 3768, 433 (1999).
5. R. Gunnink, "MGA: A gamma Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances", Report # UCRL-LR-103220 Vol. 1&2, (ISPO No. 317, Task No. A.161), (1990).
6. R.Gunnink, private communication.