

Application of Mercuric Iodide Detectors to the Monitoring and Evaluation of Stored Special Nuclear Materials

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Abstract

Mercuric iodide is a very promising material for the monitoring of stored nuclear materials that can be characterized by the energies and relative intensities of the gamma ray spectra that the materials produce. The high density and high average atomic number of mercuric iodide results in higher efficiency than other solid-state detectors such as CdTe, CZT or HPGe. In addition, the high resistivity of the material at room temperature makes it possible to produce relatively large detectors with very low noise levels.

Recent technological advances have made it possible to routinely fabricate detector structures of different dimensions tailored to the energy ranges to be investigated. Typical designs have a nominal energy operating range of 30 - 1300 keV with spectral resolution of 2% or better at 662 keV. This resolution can be improved by the use of advanced electronics and/or tailored detector design. These detectors operate over a temperature range of less than zero to greater than fifty degrees Celsius and have stable performance over long operating lifetimes. The stability, resolution, efficiency, and radiation hardness of these detectors make them ideally suited to unattended monitoring systems either as radiation counters or spectrometers.

The detectors are packaged together with a preamplifier in a small, lightweight and rugged Mercury ModuleTM that can be used individually or as part of an integrated array of detectors. Several modules can be connected to a common signal processing system so that several locations can be monitored using one spectrum analysis and monitoring system.

Spectral data of the detectors will be presented and the electronic layout of the Mercury ModuleTM will be shown. Performance measurements on several representative materials will be shown. Plans for an extended monitoring system to be used in large storage vaults will be presented and discussed.

Introduction

Mercuric iodide, on the basis of its physical and electronic properties, has been considered for use as an ambient temperature solid-state radiation detector for many years. 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. Approximately three years ago, as proof that a rugged but high-sensitivity mercuric iodide-based instrument was possible, a pocket-sized radiation counter was developed with a mercuric iodide detector volume of slightly larger than 3 cm³ (1). More recently it has become possible to fabricate efficient detectors with sufficient energy-resolution that

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together with suitable electronic pulse processing systems, can be applied to the intermittent or continuous monitoring of stored Special Nuclear Materials (SNM). This report describes the performance of these detectors and to a lesser degree the electronic systems that are being developed to optimize the spectral output of the monitoring system.

Properties of Mercuric Iodide

Mercuric iodide, in the single crystalline form suited for radiation detector 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 resistivity of 10^{14} Ohm-cm. As a result, the leakage current of large area detectors is in the nanoampere range, even at electric fields of 10^4 V/cm. Therefore, as is the case in other room-temperature detectors, the detector leakage current is not the overriding factor in the overall noise spectrum of the detector.

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 the 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 full-energy peak efficiency, especially at higher gamma ray energies. This property of mercuric iodide is illustrated in Figure 1, where the excitation processes in different detector materials are displayed as a function of photon energies (2).

Current detector fabrication techniques have resulted in the ability to produce detectors with long operating lifetimes. Detectors in the pocket gamma counter described above have been used in the field for three years with no evident degradation in performance. Several spectrometric detectors have been tested for stability of performance over several months and over a temperature range of -20 to +50 degrees Celsius. These detectors were stable during these tests with only minimal changes in the position of the full-energy peak. Details of these experiments will be reported in the near future (3,4).

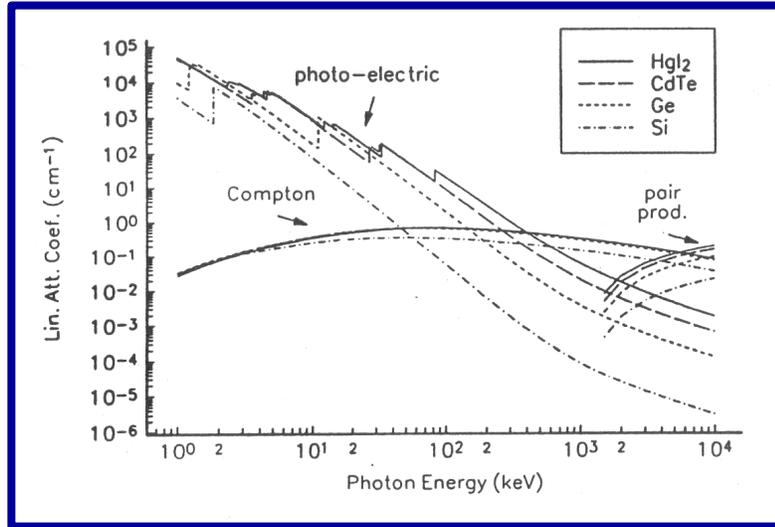


Figure 1. Ionization Processes in Different Detector Materials over a Range of Photon Energies (From Reference 2)

The stability and performance of mercuric iodide detectors have also been tested 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 (5). None of the experimental conditions used created observable changes in the properties of the detectors subjected to the particle beams, and one may well ask if any man-made facility is able to test the conditions under which damage can be created in mercuric iodide.

Detector Performance

The gamma ray detectors are usually tested at three different energies: 662 keV (^{137}Cs), 122 keV (^{57}Co) and 59 keV (^{241}Am). Standard NIM electronics are used for power supply and processing of the pulses. The shaping times used with the Gaussian shaping amplifier are 24 microseconds for the higher energies and less for the ^{57}Co and ^{241}Am sources. Both the detector and the preamplifier are kept at room temperature. A representative spectrum is shown in Figure 2.

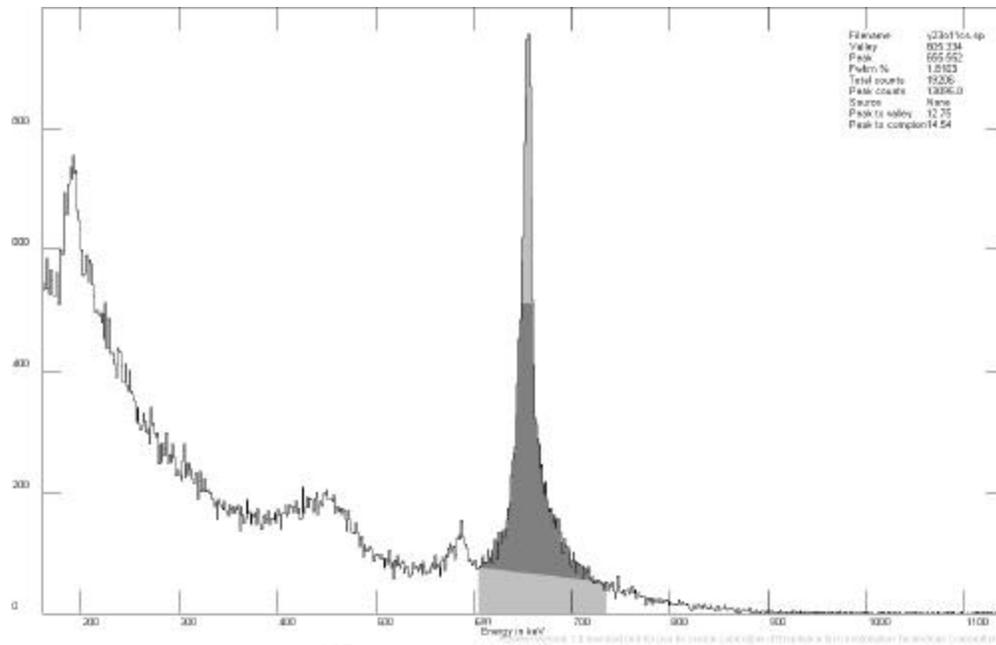


Figure 2. Spectrum of ^{137}Cs Measured With A Mercuric Iodide Detector

One can still observe some tailing at the low energy side of the full-energy peak. This is indicative of incomplete charge collection of the holes. This not only reduces the number of counts in the full-energy peak, but also increases the background between the full-energy peak and the mercury escape peak, so that the peak-to-valley ratio is reduced. The use of long shaping times to collect as many holes as possible presents two problems. The reset times required by the shaping amplifier restrict the count rates to a few thousand per second without having excessively large pulse pile-up. In addition, the noise originating from the preamplifier becomes a significant factor in the spectral resolution. Simplified evaluations of the contribution of this noise indicate that preamplifier noise can contribute as much as 3 keV to the FWHM of the spectra, which is a significant effect when lower energy pulses need to be measured. Electronic systems are being designed on the basis of gated integrator technology to alleviate these effects. In this way the reset time of the system can be reduced to less than 100 microseconds, and the count rate can be increased to several times 10,000 counts per second without excessive pile-up. This count rate is more in agreement with the radiation environment encountered in facilities where SNM is 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.

Systems Development

For actual applications, the detector is installed in a metal enclosure, together with the preamplifier and bias stabilizer. The Mercury ModuleTM 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 back of the module, so that it becomes

a forward-looking unit with only the front face of the detector exposed to the radiation. The current physical appearance of the module is shown in Figure 3.



Figure 3. Mercury Module.

To support the Mercury Module™, a small unit has been designed which provides power to the module and receives its output signal. This unit, called the MicroMax, also contains the gated integrator system and a multichannel analyzer. The MicroMax can be powered by batteries or from the line power, and can accommodate several Mercury Modules. The MicroMax can collect and store spectra for subsequent download to a PC of the PC can be used as the primary system controller. Figure 4 shows a photograph of the MicroMax.



Figure 4. MicroMax Unit.

This combination of the Mercury Module and MicroMax can be used with the flexibility required by the operator in semi-permanent situations or under portable conditions.

Applications to Safeguards

The mercuric iodide system was used to measure a spectrum of a weak ^{235}U source in the laboratory. The result is shown in Figure 5.

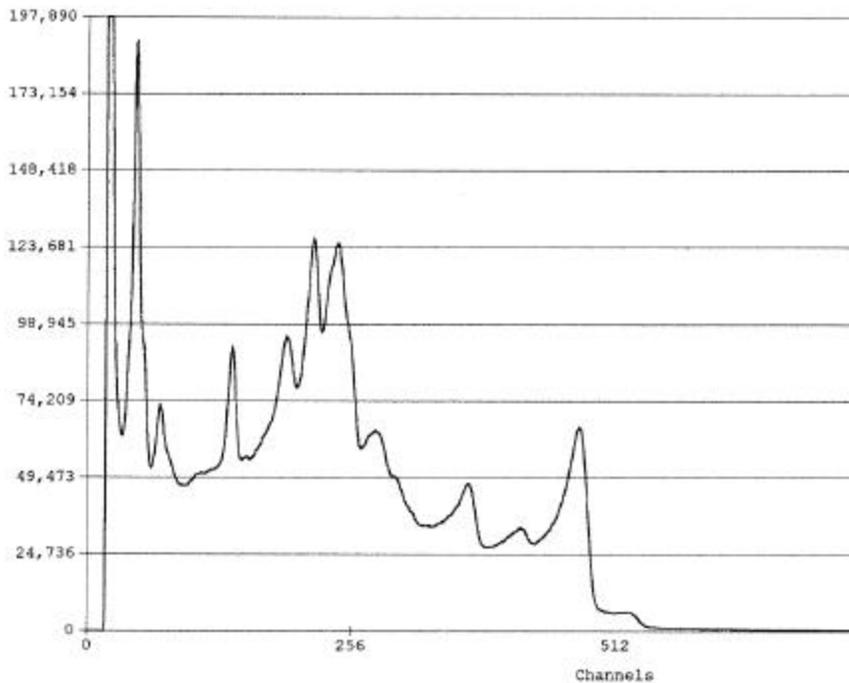


Figure 5. Spectrum of ^{235}U Measured With A Mercuric Iodide Detector

Although this spectrum of enriched uranium does not show the structure of a spectrum taken with an HPGe-detector, it does clearly show the significant lines in the spectrum, including the uranium and thorium fluorescence lines caused by self-absorption in the material. Furthermore, the spectrum is of sufficient quality to be amenable to a variety of software analysis systems.

Summary and Conclusions

The technology of fabrication of mercuric iodide detectors has progressed to the point that reliable detector systems can be manufactured. The high photoelectric efficiency of mercuric iodide, coupled with the low leakage current at room temperature, make these detectors prime candidates for applications in safeguards and other situations where spectral analysis of radioactive isotopes is required. The only drawback in the performance of the detectors is the long time needed to collect the holes. This due to a combination of hole trapping and of the low value of their mobility. Both these effects can be ameliorated by proper selection of the signal processing electronics. The relatively long shaping times needed to collect the holes can be reduced by the application of gated integrator technology. Compact electronic systems that incorporate

this feature, as well as a multi-channel analyzer and the power supplies for the detector and the preamplifier, are in production.

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