Lead Iodide Optical Detectors For Gamma Ray Spectroscopy

K.S. Shah, P. Bennett, M. Klugerman, L. Moy, L. Cirignano, Y. Dmitriyev, M.R. Squillante, F. Olschner and W.W. Moses*

RMD, Inc., 44 Hunt Street, Watertown, MA 02172

*Lawrence Berkeley National Laboratory, Berkeley, CA

Abstract[1]

This paper describes the research performed in developing low noise, high quantum efficiency lead iodide photodetectors for use with scintillators. These photodetectors operate with very low leakage current and show high quantum efficiency (>60%) in 350 to 500 nm region. Successful scintillation studies have been performed at room temperature as well as elevated temperatures (100 °C) using PbI_2 photodetectors with LSO and CsI(Na) scintillators. Detailed analysis of noise has also been performed and potential applications are discussed.

I. INTRODUCTION

PbI_2 is a wide bandgap semiconductor (E_g = 2.3 eV) and has been investigated previously as a direct X-ray and γ-ray detector [1-5]. It has a hexagonal layered crystal structure with a relatively low melting point of 408 °C. PbI_2 is a promising detector material due to its high resistivity, good charge transport, and good stability. At RMD, we have been conducting research towards development of high resolution, room temperature PbI_2 X-ray detectors for several years. This has involved systematic evaluation of material purification, crystal growth and detector fabrication procedures. As a result, we have successfully grown large volume (2.54 cm diameter, 3.5 cm long) single crystals of zone refined PbI_2. These crystals have shown extremely high resistivity and good electron charge transport. Table I lists important physical and electronic properties of lead iodide. X-ray detectors are generally fabricated from PbI_2 crystals using ohmic contacts such as Pd or graphite. These detectors exhibit very good energy resolution at room temperature. Figure 1 shows an ^{55}Fe spectrum (5.9 keV X-rays) recorded with a 1 mm^2 PbI_2 detector and the energy resolution of the 5.9 keV peak is 370 eV (FWHM).

Since many requirements such as high resistivity, low noise, and good electron transport are also necessary for optical detectors, PbI_2 is a potential candidate as a photodetector in scintillation studies. Fabrication of PbI_2 photodetectors requires an optically transparent front electrode, and we have experimented with thin evaporated films of palladium (<10 nm). Some photodetectors have also been fabricated with liquid (phosphoric acid solution) front electrodes for comparison with the metal electrodes. PbI_2 photodetectors have been evaluated by measuring their quantum efficiency, noise, and scintillation studies have been performed using LSO. Some high temperature studies have also been conducted using CsI(Na) scintillators.

Table I. Properties of Lead Iodide

Property	Value
Bandgap	2.3 eV
Crystal Structure	Hexagonal
Melting Point	408°C
Vapor Pressure at 200°C	10^{-5} torr
Dielectric Constant	21
Density	6.2 g/cm^3
Resistivity (ρ)	>10^{13} Ω-cm
electron (μτ) product	10^{-5} cm^2/V
hole (μτ) product	2×10^{-6} cm^2/V

II. QUANTUM EFFICIENCY AND DARK CURRENT:

PbI_2 optical detectors have been characterized by measuring their optical quantum efficiency as a function of the incident wavelength. The experiment involved recording the photocurrent from a PbI_2 detector as a function of the wavelength of incident light (using a monochromator) and then normalizing the detector response to that of a calibrated silicon photodiode to generate the quantum efficiency data. This experiment was performed using PbI_2 detectors with Pd as well as liquid front contacts and the results are shown in Figure 2. Also shown in Figure 2 is the quantum efficiency of a standard silicon p-i-n photodiode (Hamamatsu S-2506). As seen in the figure, the quantum efficiency of PbI_2 photodetectors is quite high in the blue region where scintillators such as LSO, GSO, LuAP, NaI(Tl), and CsI(Na) emit. As expected the liquid contacts due to their lower intrinsic absorption provide higher quantum efficiency, but even Pd contacts provide reasonably good results (>60% in the 350 to 530 nm region). The drop in the quantum efficiency of PbI_2 photodetectors at 530 nm is due to the band-edge of 2.3 eV in PbI_2. It is worth noting that the efficiency with PbI_2 is higher than that with conventional silicon p-i-n photodiodes (for λ≤450 nm). While the liquid contacts are not practical, they provide information about the

[1] Support for this work was provided by a grant from the National Institute of Health (#9R44NS35698-02)

0018-9499/97$10.00

intrinsic capability of PbI_2 crystals and we plan to experiment with more practical transparent electrodes such as evaporated indium tin oxide (ITO).

The dark current of the photodetectors was measured as a function of the applied voltage. As expected, the photodetectors showed very low dark current at room temperature (< 1 pA/mm^2 at an operating bias needed for good charge collection). Hence the detectors operate with very low shot or parallel thermal noise.

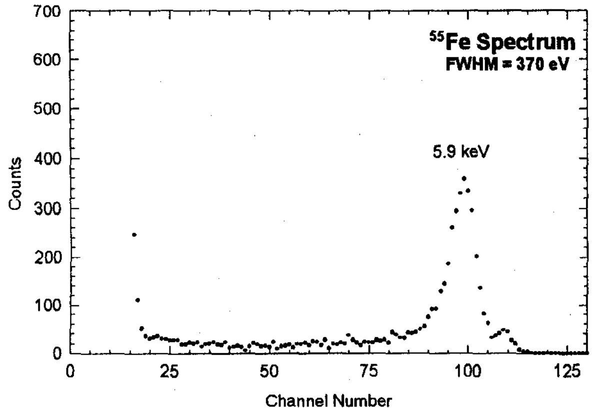

Figure 1. ^{55}Fe spectrum with a PbI_2 detector (1 mm^2 area, 100 μm thick) at room temperature.

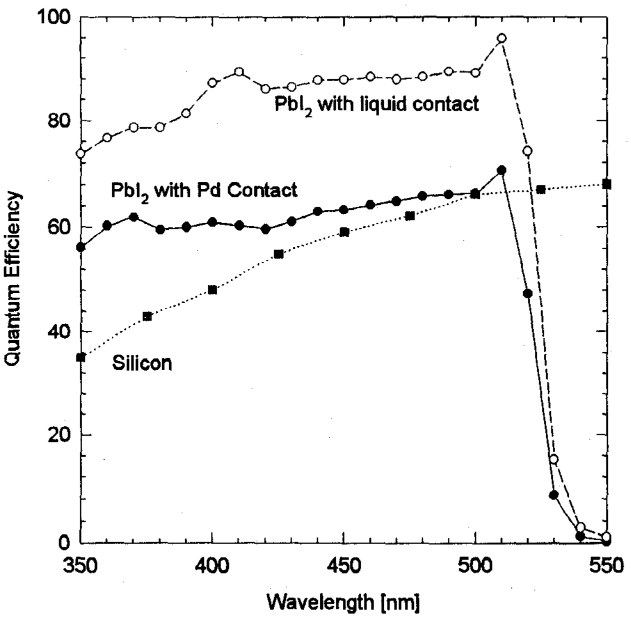

Figure 2. Quantum efficiency versus wavelength behavior for lead iodide photodetectors with two different front electrodes and for a standard silicon p-i-n photodiode.

III. SCINTILLATION STUDIES

PbI_2 optical detectors were tested as scintillation spectrometers by coupling them to LSO crystals. The detector was 3.5 x 3.5 mm^2 in area with evaporated Pd front electrode and the scintillator was 3 x 3 x 3 mm^3. Standard nuclear instrumentation was used and the detector was irradiated with isotopic sources such as ^{137}Cs and ^{22}Na and the resulting pulse height spectra are shown in Figures 3 and 4. The resolution of 662 keV peak was measured to be 12% (FWHM), for 511 keV the resolution was 16% (FWHM). As shown in the figure, the signal magnitude corresponding to 662 keV γ-ray interaction was estimated to be 3500 electrons (by calibrating the energy scale with direct X-ray detection in PbI_2). The electronic noise as measured by test pulser was 9% (FWHM) which corresponds to 300 electrons (FWHM) or 130 electrons (rms). The electronic noise in absence of any scintillator was measured to be 120 electrons (rms). Previous analysis has shown that 1/f noise is the dominant noise source in PbI_2 detectors [4]. Due to the high resistivity of lead iodide crystals, the shot noise (or the parallel thermal noise) resulting from the detector dark current was found to be less than 45 electrons (rms) and the series thermal noise was measured to be 75 electrons (rms). Thus the 1/f noise in the detector is about 100 electrons (rms). These measurements were at an integration time of 4 μs.

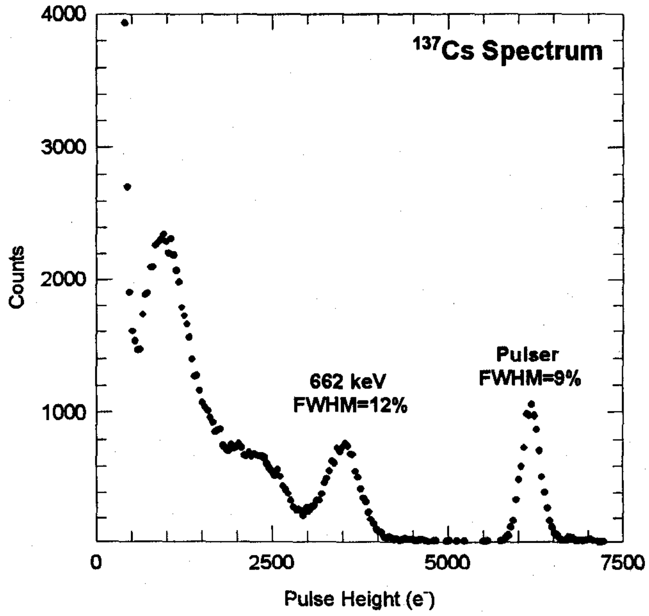

Figure 3. ^{137}Cs spectrum with a PbI_2 photodetector coupled to LSO scintillator at room temperature. The detector was operated at bias of 75 V.

IV. HIGH TEMPERATURE STUDIES:

In view of the wide bandgap, low vapor pressure, and high resistivity, PbI_2 detectors are capable of low noise operation at elevated temperatures. Successful operation of lead iodide X-ray detectors at 100 °C has been reported earlier [3]. In this paper we report the early results on the operation of PbI_2 optical detectors at elevated temperatures. PbI_2 photodetectors with palladium front contacts were investigated first by measuring their dark current as a function of temperature as shown in Figure 5. The dark current was very low at temperatures over 100 °C, indicating that the photodetectors should operate with low noise at elevated temperature. Figure 6 shows an α-particle (5.5 MeV, ^{241}Am source) spectrum recorded with a PbI_2 photodetector (36 mm^2) coupled to CsI(Na) scintillator at 100 °C.

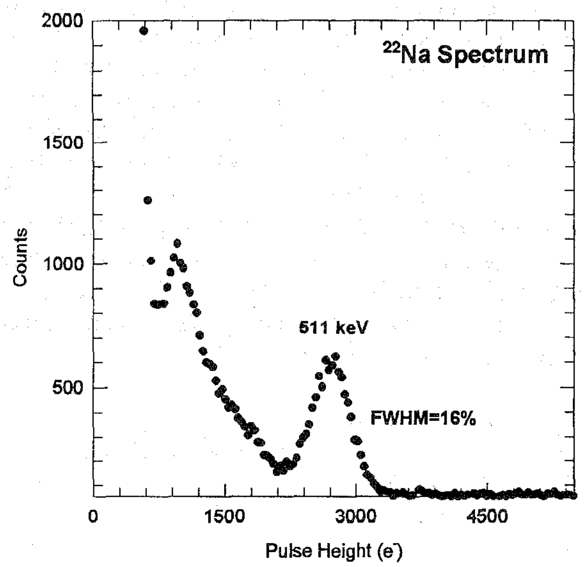

Figure 4. ^{22}Na spectrum with a PbI_2 photodetector coupled to LSO scintillator.

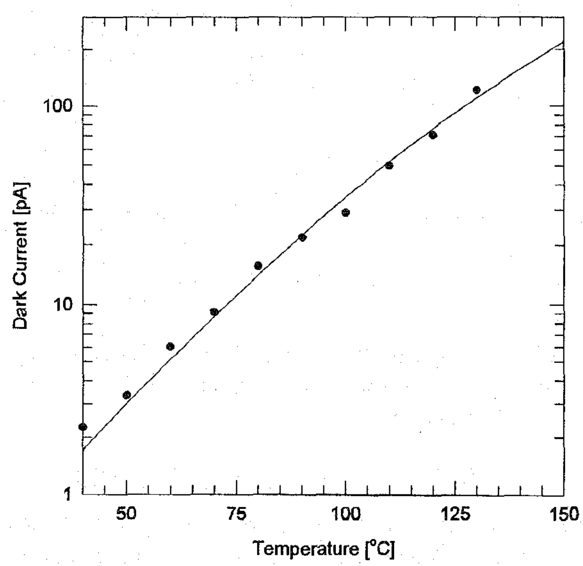

Figure 5. Dark current versus temperature behavior for PbI_2 detector (4 mm^2 in area). The solid line represents regression fit to the data set.

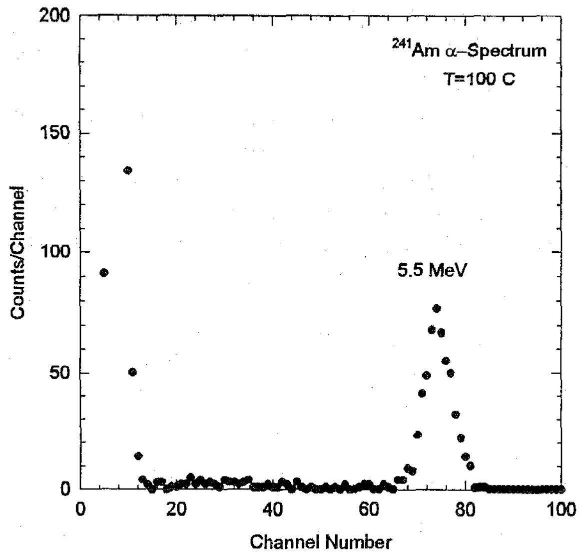

Figure 6. ^{241}Am spectrum (5.5 MeV α-particle) recorded with a PbI_2 detector (36 mm^2) coupled to CsI(Na) (5 mm diameter, 5 mm long) at 100 °C.

V. SUMMARY

PbI_2 optical detectors have been developed which are capable of providing high quantum efficiency and low noise operation at room temperature as well as elevated temperatures. Successful detection of scintillation photons has been accomplished using LSO and CsI(Na) crystals. In view of their high blue response, PbI_2 detectors are well suited for medical imaging applications such as PET where blue emitting scintillators such as LSO and LuAP are attractive. The high temperature capability of PbI_2 makes it promising for applications such as bore-hole logging and environmental monitoring.

VI. REFERENCES

[1] S. Roth and W.R. Willig, Lead Iodide Nuclear Particle Detectors, Appl. Phys. Lett., 18, p. 328, (1971).

[2] C. Manfredotti, R. Murri, A. Quirini, and L. Vasanelli, PbI_2 Nuclear Detectors, IEEE Trans. Nuc. Sci., NS-24, p.126, (1977).

[3] J.C. Lund, K.S. Shah, M.R. Squillante, and F. Sinclair, Properties of Lead Iodide Semiconductor Detectors, Nucl. Inst. and Meth, A283, p. 299, (1989).

[4] K.S. Shah, J.C. Lund, F. Olschner, P. Bennett, J. Zhang, L.P.Moy, and M.R. Squillante, Electronic Noise in Lead Iodide X-ray Detectors, Nucl. Inst. and Meth., A353, p.85, (1994).

[5] K.S. Shah, F. Olschner, P. Bennett, M.R. Squillante, and J.C. Lund, Lead Iodide X-ray Detection Systems, presented at the 9th Intl. Workshop on Room Temperature Semiconductor Detectors at Grenoble, France (1995), accepted for publication in Nuc. Inst. and Meth.