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Performance of lead iodide nuclear radiation detectors with the introduction of rare earth elements

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Abstract: Lead iodide has been recognized as a promising material for room temperature radiation detectors. It has a wide band-gap (∼ 2.3 eV), high atomic numbers (82, 53) and it is environmentally very stable compared to mercuric iodide. Electrical and optical properties of lead iodide grown crystals purified under the influence of selected rare earth elements have been investigated. Photo-luminescence and capacitance-voltage measurements have been performed using different rare earth elements.

Keywords: Lead iodide, rare earth elements, capacitance measurements, wide band-gap, room temperature radiation detectors
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1 Introduction

Lead iodide (PbI_2) is a high atomic numbers (82, 53) semi-conducting material with a wide band-gap of around 2.3 eV at room temperature and has potential as a room-temperature radiation detector. Lead iodide detectors have more favorable properties when compared to mercuric iodide detectors. Lead iodide has lower vapor pressure, better chemical stability and the capability to work at elevated temperatures up to 90 °C

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without deterioration.

The operation and the desirable features of room temperature nuclear radiation detectors can be summarized as follows:

Incident radiations (gamma or x-rays) interact with the detector volume and generate electron – hole pairs. An appropriate electric field can generate a current that can be detected by a low noise preamplifier followed by a shaping amplifier and possibly by a multi-channel analyzer for further processing.

High atomic number materials (such as lead iodide and mercuric iodide) are very sensitive to gamma and x-rays compared to low atomic number materials (such as silicon and germanium).

Both, lead iodide and mercuric iodide detectors, are suitable for operation at room temperature (without any cooling) due their large bandgaps.

Favorable properties of ideal room temperature radiation detectors are:

- High atomic number for efficient electron-holes creation and collection.
- Large bandgaps, low noise (low leakage currents) and very high resistivity ($> 10^{12}$ Ω cm).
- Minimum impurities and internal structural defects.
- Have almost ideal electrodes / contacts to reduce defects and charge carriers.
- Should be associated with very low noise electronics.

Lead iodide nuclear radiation detectors are excellent candidates for many applications which require portability and low price. They are suitable for high outdoor temperature applications. They would make the detection of hazardous materials in the vicinity of humans more feasible while medical imaging tools for quick diagnostics would become more affordable.

Table 1 shows a comparison between the physical properties of lead iodide and mercuric iodide radiation detectors.

The purpose of this work is to study the effects of added rare earth elements on the quality and the behavior of lead iodide radiation detectors. Small percentages $(0.05-0.8)$ by weight of some selected rare earth elements had been added to pure lead and pure iodine during the purification processes.

Several earlier papers have reported the results of adding rare earth elements to different materials during purification or during crystal growth [1, 2]. Narrow line width emissions were obtained at selected wave lengths from the ultraviolet to infrared when GaN was doped with rare earth elements [3]. It was shown by Favennec et al [4] that the thermal quenching of Er-doped semiconductors decreases with increasing band gap, so wide-band semiconductors such as lead iodide could be attractive hosts for rare earth elements. Also, it was found that for GaAs epitaxial layers, a considerable decrease in the free charge carrier concentration may be achieved by optimal quantitative combination of Al and Yb dopant during growth [5]. Lee et al [6] have indicated that holmium (Ho) doping greatly reduces residual impurities and improves the quality of In GaAs P epilayers. Finally it was found that holmium (Ho), dysprosium (Dy) and erbium (Er) are n-type dopants in lead telluride [7].

Parameter	HgI_2	PbI_2	Comments
Atomic Number (Z)	80,53	82,53	Absorption improves with increasing Z
Bandgap (eV) at room temp.	2.1	2.3	Wide bandgap reduces dark current /Noise
e-h pair formation energy (eV)	5	5.5	Gain increases with lower values
Mobility life-time product ($\mu\tau$) cm^2/V	10^{-4}	10^{-5}	High $\mu\tau$ better charge collection
Electron mobility cm^2/VS	$100-120$	8	Higher μ_n better charge collection
Hole mobility cm^2/VS	$4-15$	2	Higher μ_p better charge collection
Melting Point °C	259	408	Lower temperature, less sophisticated crystal growth setup
$\ell(\Omega$ cm)	$10^{11}-10^{13}$	$10^{10}-10^{13}$	Higher resistivity, lower noise

Table 1 Comparison of physical properties of HgI_2 and PbI_2.

Most of the reported lead iodide crystal growth methods were based on a pure commercial starting material. This material was then purified using a horizontal float zone purification heater [8, 9]. Crystals were grown using the Bridgman method. Matuchova et al [10] pursued a different approach for the purification/crystal growth of lead iodide. A summary of their approach follows.

A direct synthesis of lead and iodine was conducted to produce lead iodide material needed later for further purification and then for crystal growth. High quality lead wires were cut into small lengths and were cleaned. An admixture of holmium was then added to the cut, cleaned lead wires. At the same time small balls of iodine of superior quality were inserted into a small ampoule. A special quartz ampile divided into two zones was used to house the iodine ampoule in the first zone and the lead wires with the added holmium in the second zone. This special quartz tube was evacuated to about 10^{-6} mbar. The Iodine zone was heated to around 200 °C and the lead zone was heated to around 700 °C (to melt the lead). These conditions helped in producing lead iodide. Later, several purification runs were performed to remove any possible residual impurities followed by crystal growth using the Bridgman – Stokbarger method.

2 Measurements

The crystals prepared by Matuchova et al [10] were characterized using low temperature photo-luminescence (PL) and capacitance – voltage (CV) measurements. Some of the best results obtained from the above measurements were for lead iodide with holmium (Ho) rare earth element. Figure 1 shows PL measurements using small concentrations of

erbium (Er) and holmium (Ho) admixtures.

The PL spectra with the holmium admixture are noticeably sharper than with the erbium admixture.

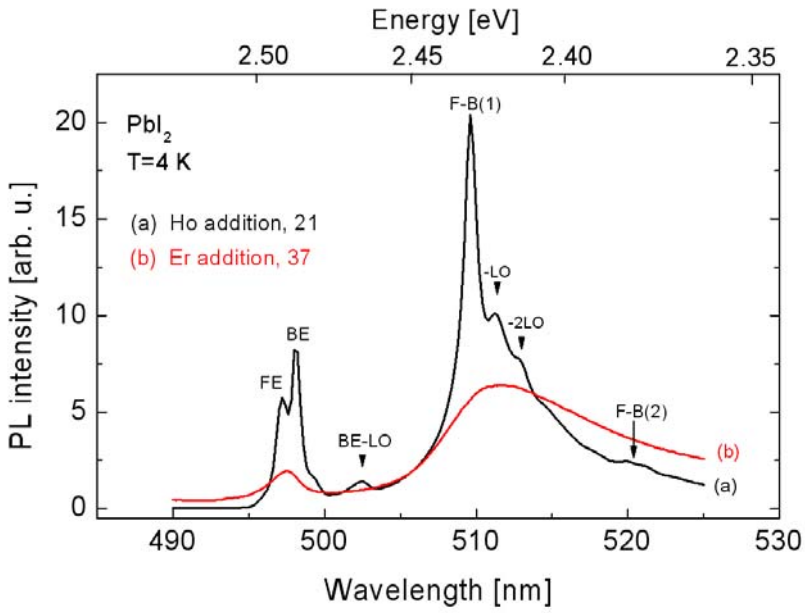


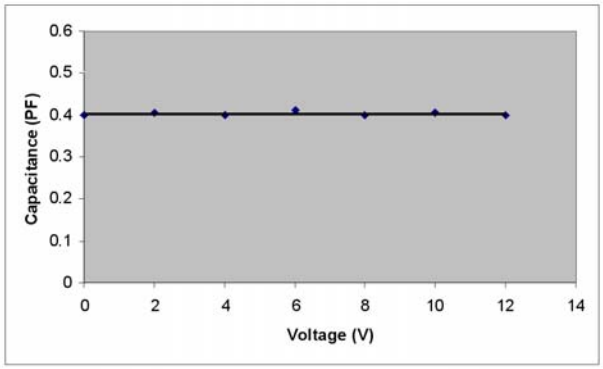
Fig. 1 Low temperature PL spectra of PbI_2 samples prepared by direct synthesis with the addition of Ho and Er.

The curves show exciton transitions FE, BE (free exciton, bound exciton) at about 2.49 eV and free to shallow donor levels (F-B) related to the transitions at 2.43 eV and 2.38 eV. The band BE-LO (longitudinal optical) at 2.46 eV is due to phonon replicas of the exciton band [10]. Figure 2 shows results of capacitance-voltage measurements for detectors (around 2mm × 2mm × 0.1mm) in the dark. Figure 2(a) shows a typical detector synthesized with holmium. Figure 2(b) shows a typical detector without any rare earth element synthesis. In the first case, capacitance variations are almost minimal, while in the second case, variations are very obvious [12].

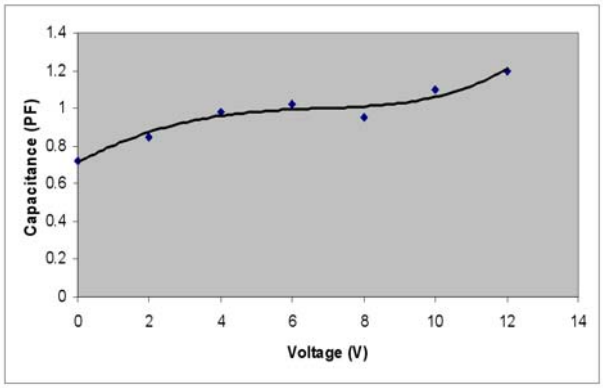
3 Discussion

Two types of samples were used for PL and CV measurements. The first group of samples is with an admixture of rare earth elements and prepared using direct synthesis. The second group has no rare earth element admixture and is also prepared using direct synthesis. Matuchova et al [10] confirmed that XRF measurements on the samples showed that these samples were of very good quality with impurities less than 10 ppm. The reasons for adding selected rare earth elements in small controlled quantities were:

- To remove undesirable impurities. This was successful due to the high chemical affinity of rare earth elements, i.e., the tendency of a rare earth element atom to



a)



b)

Fig. 2 Results of capacitance – voltage measurements for PbI_2 detectors prepared by direct synthesis with and without the addition of Ho, shown by curves (a) and (b), respectively.

combine by chemical reaction with an atom of unlike composition (i.e. to combine with impurities).

- To reduce the number of purification runs.
- To study the effects of adding rare earth elements on the quality of detectors.

The following rare earth elements were used during the preparation of samples:

- Cerium (Ce)
- Gadolinium (Ga)
- Holmium (Ho)
- Erbium (Er)
- Ytterbium (Yb)

It was found that the addition of holmium produced the best results. Its PL spectra are very sharp and its CV measurements are almost constant. This is a good indication of a material with a pure and uniform internal structure [11].

Additionally, it was also noticed that low percentages (< 0.05) of added rare earth elements gave better results.

4 Conclusion

Initial investigations have confirmed that lead iodide crystals with holmium rare earth element and direct synthesis is a good choice for several applications. We feel that rare earth elements could play an important role in preparing optimized lead iodide radiation detectors. More work is planned to better understand the effects of rare earth elements on the quality of lead iodide radiation detectors. Some of the major points that will be studied in future work are:

- The effects of rare earth elements on the purification process.
- Doping effects of rare earth elements on the behavior of lead iodide radiation detectors (i.e., the contribution of adding rare earth elements to energy resolution and efficiency of the detector).
- Lead iodide crystal growth optimization in order to reduce structural defects which are responsible for the low charge collection of lead iodide detectors.

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