Original Article

X-Ray Sensing by Mercury Iodide Poly-Methyl Methacrylate Composite

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Abstract

In Today's technological era, digital devices have made their place in every field. In the medical field, digital X-ray applications have contributed a lot. As a transducer, a good X-Ray sensor is always in demand. Many solid-state sensor materials are used as X-ray sensors, like lead iodide, Zinc Sulphide, Titanium dioxide and Mercury iodide, etc. Mercury Iodide is highly suitable for making X-ray sensors. It has favorable properties like a high mobility-life time ratio. This makes it a good material for fabricating an X-ray sensor. However, it is very difficult to handle a single crystal of mercuric iodide due to its limited mechanical strength. The previous study revealed that composites are best among the three forms i.e. single crystal, thin film, and composites. In the present study polymer composites of Mercury Iodide using Poly Methyl Methacrylate as polymer matrix and Mercury iodide as reinforcement material with different concentrations were fabricated. Polymer composite sheets were prepared for various concentrations of Mercury Iodide. These sheets were then subjected to X-Rays switching studies at room temperature. Studies revealed that Mercury Iodide - PMMA sheets show a stable detection of X-rays for all concentrations and the value of (Imax-Imin) increases in the concentration of Mercury iodide to 5.85 %, and after 8.65 % concentration value of (Imax-Imin) starts decreasing. An optimized range of concentration is worked out (5.85 % to 8.65%) at which the maximum value of photocurrent is obtained. These sheets show quite low thermally generated charges at room temperature and high photocurrent. X-ray switching studies conducted on these sheets show low rise and fall time making the material good for imaging applications. Rise time was recorded as 0.65 sec and fall time was 0.48 for a pulse of 2s and more.

INTRODUCTION

In today's technological era, digital devices have made its place in every field. In medical field also digital X-rays applications has contributed a lot to diagnose a problem. As X-rays are not secure to human body and radiation exposure can cause cell mutations that lead to cancer. Therefore, there is need to limit X-ray dose to patient. It can be done only if our x-rays detectors are very sensitive to high energy radiation. So, a good X-ray sensor is always in demand. Many solid state materials are used as X-ray sensors like Lead iodide, Cadmium iodide, Zinc sulphide, Titanium dioxide and Mercury iodide etc. Mercury Iodide is very much suitable for making X-ray sensor and has favourable properties to fabricate a sensor [1,2]. It is very difficult to handle single crystals mechanically and previous study revealed that, composites are best among the three forms i.e. single crystal, thin film, composites [3]. It was decided to prepare composites of Mercury Iodide with the polymers like PMMA (Polymethyl methacrylate) with different concentrations of mercuric Iodide. Some of the latest sensor materials are listed in table-1 and its environmental hazards are also listed according to MSDS data from sciencelab.com.

Table1. Material Safety Data Sheet (MSDS) rating of several solid state detector materials along with Mercuric Iodide in respect of health hazards caused by them (sciencelab.com).

S. No	Sensor	Health Hazards (NFPA)(0-4)
1	Mercuric iodide	3
2	Cadmium Iodide	3

3	Gallium Arsenide	3
4	Bismuth (III) oxide	1
5	Lead Iodide	3
6	Cadmium Iodide	3
7	Cadmium telluride	2
8	Titanium dioxide	1

The data from table-1 reported that Mercuric Iodide is a poisonous material that is harmful to the environment if it is used directly as a sensor. Polymer composites are heterogeneous substances consisting of two or more materials that have their individual characteristics. The combination of materials brings about new physical, chemical, and mechanical desirable properties [5]. Therefore, its toxic nature can be reduced to no harmful level by making its polymer composites and it can be used as a sensor without any adverse health effects. A good solid-state detector material is based on the following major properties [4]:

1. Material should have a high band gap. This helps in minimizing thermally generated noise.

2. Constituent atoms should have a high atomic number. This is for maximum absorption of X-ray energy.

3. Mobility-life time product of the material should be high. This helps in better charge collection.

4. Operating electric field should be low. It helps in keeping electronics involved simple.

5. Operating temperature likely to be room temperature for ease in operation.

6. Response time should be small for faster data processing.

7. Fabrication of the detector should be simple with the flexibility of design, shape, and size.

8. Highly stable or have low degradation.

Mercuric iodide is the most important material, due to its following promising properties:[8]

1. It is a wide band gap semiconductor (having a band gap of 2.13 e.V. which leads to a reduction in the density of thermally generated free carriers as compared to the density of X-ray generated carriers. This helps in keeping a low dark current in comparison to a photocurrent.

2. Due to the high atomic number (Hg-80u and I-53u) of its constituents, it has a high photon absorption coefficient.

3. Mobility-life-time product of the material is quite high i.e. 10 – 5cm2/V, which ensures better charge collection.

4. The operational electric field is fairly low | 104 V/cm which removes the requirement for a high voltage power supply.

5. Material processing temperature is low i.e. 1000C therefore, its processing is easy.

Each solid-state detector has properties that make them suitable for the detector, but the most important thing is flexibility in designing the shape of the detector. X-rays are high-energy radiations. When they interact with these materials they are partially reflected and partially refracted through the material. Photocurrent induced on the exposure of X-rays depends on the Quantum efficiency of material, the area exposed, reflectance coefficient, intensity wavelength product, and thickness of the sample [8]. The limited portion is absorbed by the material. If the detector is properly shaped, it is possible to maximize the absorption of X-rays. Single crystalline material gives limited flexibility in designing the shape of the detector. Keeping this in mind, it was planned to develop composite detectors. To have a better spectrum of physical and chemical properties it was planned to blend Mercuric Iodide with polymer (poly-methyl methacrylate). Poly-methyl methacrylate (PMMA) is a well known polymer. It is selected due to its following properties [6].

1. Low linear mold shrinkage (0.003-0.0065 cm/cm)

2. Good mechanical properties (Hardness (63-97 Rockwell, M) and Tensile strength (47-79 MPa).

3. Most resistant to direct sunshine exposure.

4. Low water absorption (0.3-2%) makes it very suitable for electrical device making.

5. Very low electrical conductivity (10-14-10-15Ω-1.cm-1)

6. High dielectric strength (17.7-60kV/mm).

Therefore, composites are the best option to enhance its mechanical properties. Polymers provide additional strength and flexibility in the shape of such detectors. In present study, a polymer composite of Mercuric Iodide has been fabricated with Poly Methyl Methacrylate as polymer matrix and reinforcement material as Mercuric Iodide. Further, they are subjected to X-ray switching studies.

Material and Sensor Development:

Mercuric Iodide has been purified by recrystallization using a multiple sublimation technique [1]. Fig. 1 shows crystals having dendrite growth which is attributed to the fast growth rate. On reducing the growth rate, it is found that crystals are very small in size therefore it is very difficult to separate them from the residual charge.

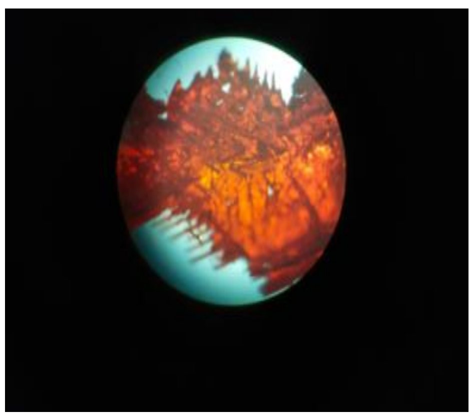

Fig1. Dendrite growth of purified Mercuric Iodide Crystals.

Polymer composites of Mercuric Iodide were prepared by setting of solution due to gravity at room temperature. Polymer Composites were prepared by dissolving a polymer and Mercuric iodide in a suitable solvent. PMMA polymer was used as matrix material with different concentrations of Mercuric Iodide. Seven samples of composites of mercuric iodide with different concentrations were prepared as mentioned in table 2.

Table 2: Quantity of Mercury iodide and PMMA in various samples.

SAMPLE NO.	QUANTITY OF HgI_2 (mg)	HgI_2 + PMMA (gm)	CONCENTRATION PERCENTAGE OF HgI_2
1	0	1.38	0 %
2	35.37	1.43	2.47 %
3	70.75	1.21	5.85 %

4	106.13	1.24	8.56 %
5	141.51	1.26	11.23 %
6	176.89	1.21	14.62 %
7	212.26	1.34	15.84 %

Samples of polymer composites with different concentrations of Mercuric iodide were shown in fig 2. While making the composite, the amount of mercuric iodide has been taken from 0% to 15.84 %, as can be seen in the figure 2.

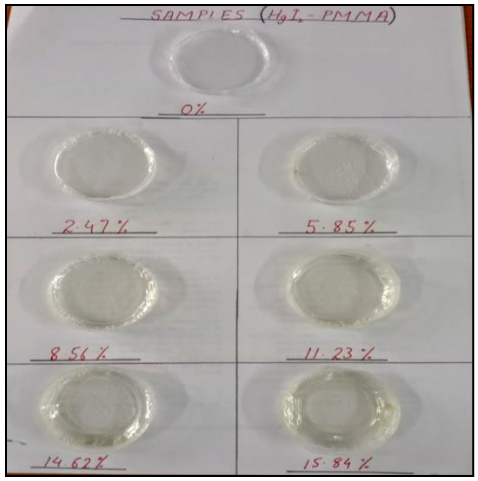

Fig 2: Polymer composites of Mercuric iodide with different composition.

These samples were cut in the form of rectangular sheets and electrodes were made by using silver paste. During sensor development, silver pasting was done at the top and bottom of the rectangular sheets such that a longitudinal configuration is obtained. Samples were then subjected to microscope studies to find out any possibility of cracks etc. Only those samples were used for studies that were found free from any crack. These prepared sheets are of the order of 1mm thickness.

X-Ray Switching Studies:

X-ray switching studies have been performed on composites of Mercuric iodide with PMMA to know about the response time of the detectors. The experimental setup used for X-ray switching studies consist of:

- X-ray source
- Time controller device
- Sensor holder
- Software/hardware to record photocurrent.

The voltage across the samples was fixed of the order of 25 V. All the samples were repeatedly exposed to an X-ray. Each exposure was of 2 sec. The photocurrent was recorded by Keithley 6485 Pico meter. The schematic longitudinal configuration of device is shown in fig 3.

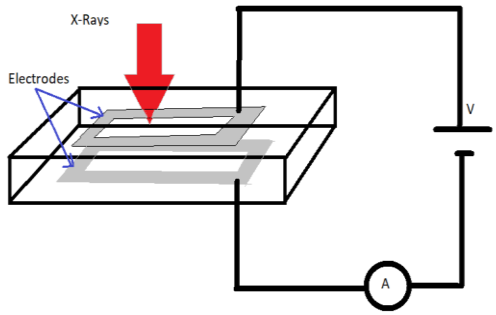

Fig 3: Circuit Diagram to measure photocurrent.

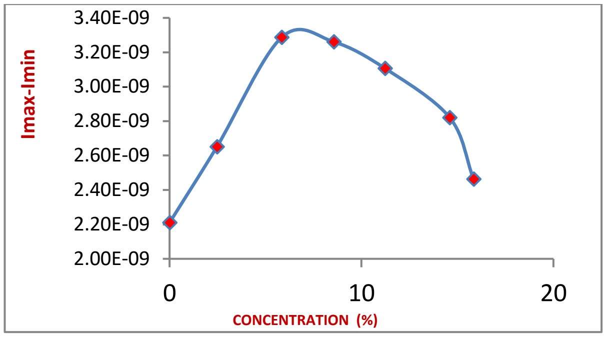

Fig 4: The variation of (Imax-I min) at different concentrations

The voltage applied to the sheets is of the order of 25 V and the variations in (Imax-Imin) were observed for all concentrations as shown in table 3. The graph in fig 4 shows the variation of (Imax-I min) at different concentrations:

Table 3. Observations of (Imax- Imin) with different concentrations.

Concentration	I_{max}- I_{min}
0	2.21E-09
2.47	2.65E-09
5.85	3.29E-09
8.56	3.26E-09
11.23	3.11E-09
14.6	2.82E-09
15.84	2.46E-09

It is observed that,for a particular value of electrode voltage of the order of 25V the value of (Imax-Imin) increases with increase in the concentration of Mercuric iodide to 5.85 % value and the value of (Imax-Imin) starts decreasing after 8.56 % concentration. In the case of polymer composites, PMMA is acting as a charge bridging material between Mercuric Iodide grains [9]. Switching curves were obtained for all seven concentrations, and (Imax–Imin) was observed as shown in table 3. The optimized range of concentration of mercuric iodide at which maximum photocurrent was observed is 5.85 % to 8.56 %. Response times for both the concentration were shown in Fig 5 and Fig 6.

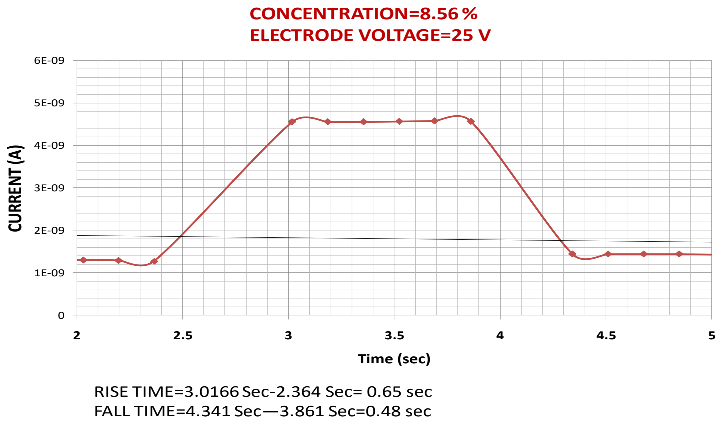

Fig.5: Response time for the concentration 8.56 %, Voltage=25 V

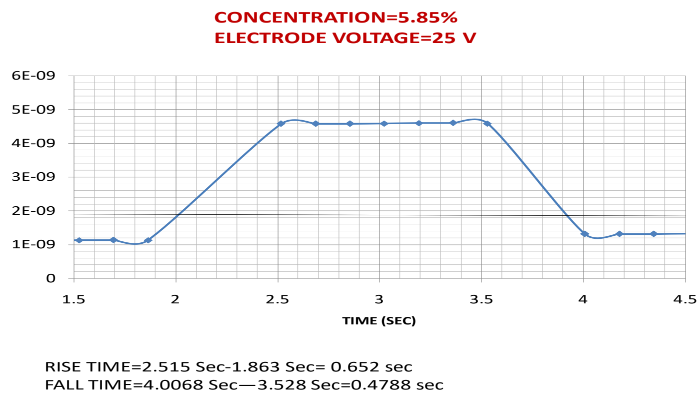

Fig.6: Response time for the concentration 5.85 % , Voltage=25 V

Conclusion:

Mercury Iodide – PMMA composite sheets show stable detection of X-rays for all concentrations but the value of (Imax-Imin) increases as we increase the concentration of Mercury iodide up to 5.85 % and after 8.65 % concentration value of Imax-Imin starts decreasing. This can be understood as initially increase in concentration of mercuric iodide, increases charge carriers. At low concentration of mercuric iodide, structural deformations were limited. As concentration of mercuric iodide is increased, structural deformation become significant. Thus, on increasing concentration beyond this limit, structural deformation starts playing dominating role. Hence, increasing the concentration of Mercuric Iodide in composite, polymer chains of PMMA starts breaking. This results in the trapping of charge carriers [7]. Thus, overall photoelectrons collection reduces.

So, an optimised range of concentration (5.85 % to 8.65%) has been obtained at which the sensor shows the maximum value of photocurrent. These sheets show quite low thermally generated charges at room temperature and high photocurrent. X-ray switching studies conducted on these sheets show low rise and fall time making the material good for imaging applications. Rise time was recorded as 0.65 sec(Fig 5,6) for both concentrations and fall time was 0.48 sec(Fig 5,6) for both concentrations for a pulse of 2 second and more.

Acknowledgement

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